Title

Impact of Increased Renewable Gases in Natural Gas on Combustion Performance of Self-aspirating Flames

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Impact of Increased Renewable Gases in Natural Gas on Combustion Performance of Self-aspirating Flames

DISSERTATION

submitted in partial satisfaction of the requirements for the degree of

DOCTOR OF PHILOSOPHY

in Mechanical and Aerospace Engineering

by

Yan Zhao

Dissertation Committee:
Professor Scott Samuelsen, Chair
Professor Vincent McDonell
Professor Derek Dunn-Rankin

2020

DEDICATION

To my family and friends.

Without the fun we had together, this could have been finished much earlier.

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NOMENCLATURE

Acronyms

HHV	Higher heating value
LHV	Lower heating value
WI	Wobbe Index
SG	Specific gravity
IG	Industrial grade
UHP	Ultra-high purity
AFUE	Annual fuel utilization efficiency
NG	Natural gas
UHCs	Unburned hydrocarbons
CFD	Computational fluid dynamics
CRN	Chemical reaction network
QCL	Quantum cascade laser

Symbols

$\eta_{cooking}$	Cooking efficiency
$c_{p,water}$	Heat capacity of water
m_{water}	Mass of heated water
h_{vapor}	Latent heat of water vapor
Δm_{vapor}	Mass of vaporized water
V_{fuel}	Volume of fuel
p_1	Natural gas pressure in the household pipeline
p_2	Natural gas pressure in the atmosphere before combustion
u	Flow velocity
$\dot{V}_f$	Fuel volumetric flowrate
A	Flow cross section area
I	AGA flame index
K	Flame lifting constant
f	Primary air factor
a	volume of air theoretically required for complete combustion
Y	Yellow tip coefficient
J	Weaver Index
D	Specific density
S	Flame speed of fuel in air
Q	Percentage of oxygen in the gas
N	The number, in 100 molecules of gas, of the total carbon atoms minus one
R	The ratio of hydrogen to carbon atoms contained in the fuel
a-subscript	Designating adjustment (original) gas
s-subscript	Substitute gas

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VITA

EDUCATION

2020 **University of California, Irvine, CA, U.S.A.**
Ph.D. in Mechanical and Aerospace Engineering
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2016 **Chinese Academy of Sciences, Beijing, China**
Masters in Power Engineering
Thesis: “Investigation of Fuel/Air Mixing and Combustion Characteristics of a Lean Premixed Burner for Gas Turbine Applications”

2013 **Hebei University of Technology, Tianjin, China**
B.S. in Energy and Power Engineering

RESEARCH EXPERIENCE & PARTICIPATED PROJECTS

2019/9- **Apparatus Design & Optimization of Blending Renewable Hydrogen into Existing Natural Gas Infrastructures**

- Proposed the hydrogen blending strategies for an existing natural gas pipeline system in California
- Conducted 3-D simulation by ANSYS-ICEM/FLUENT to optimize the hydrogen blending system
- Working on safety codes for hydrogen mixing and gas quality standards on hydrogen/NG mixtures

2017/7-2019/11 **Experimental & Numerical Assessment of Residential Appliances Performance Operating on Hydrogen/Biogas/Natural Gas Mixtures**

- Designed and set up the gas mixing system to generate fuel mixtures with varying species
- Conducted combustion tests on residential appliances operating on NG/biogas/hydrogen mixtures
- Developed CFD-CRN models predicting emission performance of multiple appliances
- Investigated the influence of hydrogen embrittlement (SEM-EDS) on flame behaviors

2016/11-2017/7 **Low Swirl Burner Flame Stabilization Strategy and Emission Control**

- Conducted tests evaluating the influence of pilot fuel on the low swirl flame stability and emissions
- Generated a 2-D model to predict the flame position variation with the pilot fuel percentage

2015/2-2016/5 **Lean Premixed Gas Turbine Burner Optimization and Testing**

- Utilized ANSYS to optimize a lean-premixed gas turbine burner
- Conducted laser diagnostic tests (LDV, PIV, PLIF) to characterize the flow field & flame structure

2014/12-2015/8 **Combustion Performance of a Premixed Burner Operated on Natural Gas/Methanol Mixtures**

- Set up the experiment system for liquid methanol gasification and mixing with natural gas
- Performed laser diagnostic and emission tests on a burner operating on NG/methanol mixtures

TEACHING EXPERIENCE

2020, 2019	**Teaching Assistant** Prof. Vincent McDonell University of California, Irvine	Fundamentals of Experimental Design

AWARDS & FELLOWSHIPS

2019	Association of Energy Engineers Scholarship, AEE Southern California
2015	Outstanding Student Honor of Chinese Academy of Sciences
2014	Chinese Academy of Sciences Scholarship
2012	National Endeavor Fellowship of China
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Sep. 2020	Presented at ICEPAG 2020 (Online presentation)
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Jun. 2019	Presented poster at 10th Anniversary Combustion Summer School (Princeton, NJ)
May. 2019	Presented poster at Global Climate Leadership Council Meeting (Irvine, CA)
Apr. 2019	Attended Sustain SoCal 10th Annual Conference & Expo (Irvine, CA)
Mar. 2019	Presented at 11th U.S. National Combustion Meeting (Pasadena, CA)
Nov. 2018	Paper presented at 14th ASME-IMECE (Pittsburgh, PA)
Sep. 2018	Presented at Air UCI Research Workshop (Lake Arrowhead, CA)
Aug. 2018	Presented at UCI COSMOS Summer Camp (Irvine, CA)
Jul. 2018	Presented at Workshop with Fortis BC and SoCal Gas (Irvine, CA)
Sep. 2017	Attended 9th Energy Efficiency in Domestic Appliances and Lighting (Irvine, CA)

JOURNAL ARTICLES

Zhao, Y., McDonell, V. and Samuelsen, S., 2020. Assessment of the Combustion Performance of a Room Furnace Operating on Pipeline Natural Gas Mixed with Simulated Biogas or Hydrogen. International Journal of Hydrogen Energy. 45(19), pp.11368-11379.

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Shao, W., Wang, Z., Liu, X., Zhang, Z., Xiao, Y. and **Zhao, Y.**, 2019. Numerical and Experimental Parametric Study of Emission Characteristics in an Axial Fuel Staging System. Energy & Fuels, 33(12), pp.12723-12735.

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CONFERENCE PAPERS

Zhao, Y., Shao, W., Liu, Y., Tang, X., Xiao, Y. and McDonell, V., 2020. Numerical and Experimental Study of Geometry Effects on Fuel/Air Mixing and Combustion Characteristics of a DLN burner. ASME 2020 Power Conference.

Zhao, Y., Morales, D. and McDonell, V., 2019. Influence of Blending Hydrogen and Biogas into Natural Gas on the Combustion Performance of a Tankless Water Heater. ASME 2019 International Mechanical Engineering Congress and Exposition.

Zhao, Y., McDonell, V. and Samuelsen, S., 2019. Experimental assessment of the combustion performance of an oven burner operated on pipeline natural gas mixed with hydrogen. 11th U.S. National Combustion Meeting.

Zhao, Y., Choudhury, S. and McDonell, V., 2018, November. Influence of Renewable Gas Addition to Natural Gas on the Combustion Performance of Cooktop Burners. ASME 2018 International Mechanical Engineering Congress and Exposition.

ABSTRACT OF THE DISSERTATION

Impact of Increased Renewable Gases in Natural Gas on Combustion Performance of Self-aspirating Flames

by

Yan Zhao

Doctor of Philosophy in Mechanical and Aerospace Engineering

University of California, Irvine, 2020

Professor Scott Samuelsen, Chair

The depletion of fossil fuels and the urgency of reducing greenhouse gas emissions demonstrate a pressing need for alternative energy sources. Currently, natural gas consumption is a major contributor to global warming, not only due to the carbon dioxide emissions in the combustion products but also due to natural gas leakage in the mining, transmission and storage facilities. Therefore, replacing the pipeline natural gas with renewable gas can decrease greenhouse gas emissions as well as reduce dependence on fossil fuels. This dissertation investigated the feasibility of blending renewable gases (i.e., biogas, hydrogen), into existing natural gas transmission and distribution pipelines from the perspective of fuel properties and combustion performance of residential appliances.

The fuel interchangeability technical barriers and challenges were first investigated, including fuel properties, flowrate control, flame performance, etc.. Moreover, the source of reddish hydrogen flame color was also investigated experimentally. The results indicated that hydrogen cracking and embrittlement could be a concern influencing flame appearance. A multi-fuel blending system was fabricated under this dissertation to generate fuel mixtures of various percentages. Burner configurations of existing residential burners were investigated, and representative self-aspirating burners were selected to conduct fuel interchangeability testing on different fuel classes. Monitored combustion performance included ignition behaviors, blowoff/flashback limits, emissions (CO, UHCs, NO, NO_2, N_2O, NH_3, etc.), efficiency, burner temperature, combustion noise, etc.. It was found that CO and UHCs emissions and the risk of flame blow-off increase significantly when the CO_2 percentage in the fuel exceeds 5 vol%. Self-aspirating burners were found to have distinct H_2 tolerance levels with a limiting factor of the flashback, which is more inclined to occur at ignition conditions. This limited the H_2 injection limits below 20 vol%. At a low level of H_2 addition to natural gas, most of the combustion performance of the tested self-aspirating devices didn't differ much and some of them were improved, which shows a positive sign for natural gas replacement with renewable hydrogen for self-aspirating flames.

CHAPTER 1: Introduction

1.1 Motivation

The adoption of renewable gases (biogas, renewable hydrogen, etc.) in our existing natural gas infrastructure is of interest in reducing greenhouse gas emissions thus combating climate change. This on-going energy transition from traditional fossil fuel to renewable energy sources brings a lot of challenges for system adaption. In this fuel exchange process, interchangeability studies regarding fuel properties, combustion devices performance and related issues need to be addressed.

Residential sector has always been in the pioneer position relative to energy transition for the past few centuries from fuelwood to coal, then to liquid and gaseous fuels [1]. However, compared to the astonishing combustion technology advancement in power generation, transportation system and industry applications, the residential sector didn't get as much attention or improvement. Take the power generation sector for example, a series of technologies were developed and adopted to reduce NO_X emissions, including Rich burn-Quick mix-Lean burn (RQL) technology [2], Dry Low NO_X (DLN) lean premixed combustion [3], Mild combustion [4], etc.. Post-combustion pollutants elimination technologies, like Selective Catalytic Reduction (SCR), are also being adopted to further decrease gas turbine NO_X emissions down to single digit ppm level [5]. In comparison, the actual emission species and levels from the residential sector are still not widely investigated and understood, and international consensus on regulating emission species of domestic appliances haven't been reached. Currently, regional emission regulations on residential appliances exist in some places, however, the emission measurement and evaluation methods are of all kinds without unified criterion.

Due to the emphasis on the indoor air quality improvement and the concept of smart building, interest in 100% electrification of residences is increasing. Therefore, the current energy consumption status both in the residential and power generation sectors needs to be studied on to investigate the feasibility of residential sector electrification and potential challenges. Furthermore, as the pipeline natural gas being replaced by renewable fuels, the renewable fuel combustion versus electrification should be evaluated.

Most of the current self-aspirating residential burners on market are designed to operate on natural gas. Replacing natural gas with renewable gases like biogas or renewable hydrogen in these burners needs comprehensive theoretical analysis and extensive experiments. In the manufactured gas era, residential burners once operated on different fuel mixtures, including hydrogen-rich fuels. Therefore, for the benefit of adopting renewable gases in existing natural gas combustion devices, lessons can be learned from the fuel involvement from manufactured gas to natural gas/propane back to the early 20th century. While learning from the previous experience, it should also be noted that residential self-aspirating flames require attention from much more perspectives. These include combustion safety, efficiency, multiple emission species, flame appearance, burner life span, ease of operability, etc. and even a decent outlook of the burner design. Most of these combustion performances of residential self-aspirating flames were not investigated in the manufactured gas era. Therefore, the technology gap in the energy transition from fossil fuel to renewable gases in self-aspirating flames needs to be addressed.

Currently, self-aspirating flames are widely adopted in residential appliances, commercial applications and light industry combustion devices [6]. To incorporate more renewable fuels in self-aspirating flames, burner configurations are needed to be conducted to characterize the working principles of different combustion devices. The tolerance level for renewable contents of existing burners adopting self-aspirating flames haven’t been tested. Moreover, the current emission regulations on domestic burners mainly focus on CO emission for safety concerns.

Emissions like NO, NO_2, N_2O, UHCs, NH_3, etc.. should also be tested for future regulation needs. Apart from the flame stability and emissions, other combustion performances like primary air entrainment, ignition behavior, burner temperature, combustion noise, etc. of self-aspirating flames haven't attracted much attention. Therefore, more comprehensive performance evaluation methodology on self-aspirating flames needs to be advanced.

1.2 Goals

The goals of this dissertation are to

(1) address the knowledge gap in the upcoming fuel transition of residential self-aspirating flames.

- The emission species and levels of residential appliances adopting self-aspirating flames.
- The tolerant level of self-aspirating flames on renewable gases.
- The influence of renewable gases on combustion performances: ignition behavior, flashback/blow-off, efficiency, emissions, combustion noise, burner temperature, etc..
- The aspects that need to be modified in current gas quality and emissions regulations.

(2) develop models predicting the burner performance and guide burner design and future policies on residential self-aspirating flames.

1.3 Objectives

The following tasks are required to achieve the goals of this dissertation:

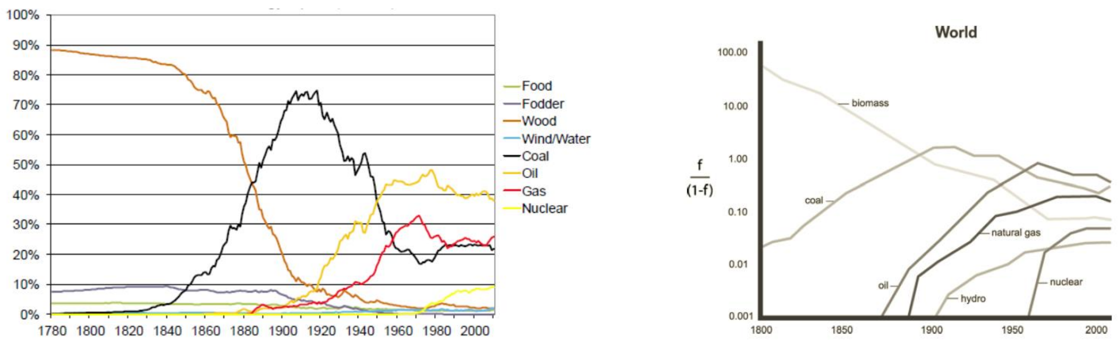
- Investigate residential burner configurations and working principles.
- Fabricate the fuel mixing apparatus and establish fuel mixtures of interests.
- Test the fuel flexibility of selective residential burners.
- Evaluate numerical models to predict the emissions of self-aspirating flames.
- Provide reference for fuel flexible burner design and future regulations.

CHAPTER 2: Background

This chapter summarizes the historical residential fuel transitions and research progress which indicates the research direction. Topics in this chapter range from past and ongoing energy transitions, to fuel interchangeability studies and related regulations.

2.1 Residential Fuel Transition

The fuel consumption shares among different energy sources vary along the human society development. Figure 1 (a) and Figure 1 (b) show the energy consumption variation for the past two centuries of the U.S. and the world, respectively. As shown, before the 19th century, wood was the dominant fuel. With the start of the Industrial Revolution, wood could no longer meet the massive energy demand due to its low energy density and the supply scarcity caused by fast forest destruction. Meanwhile, coal consumption significantly increased with its worldwide discovery and the advancement of coal mining technologies. Coal consumption surpassed that of wood in the late 19th century and played an essential role in the Industrial Revolution. The oil and gas industry started to come up around the 1900s and their market share surpassed the coal consumption in the 1950s. Till today, oil, natural gas and coal are still the dominate energy sources.



(a) Shares of the U.S. energy consumption, 1780–2010 [7] *(b) Global primary energy transition, 1800–2010 [8]*

Figure 1: Transition of energy consumption in the U.S. and the world

The energy transition of U.K., France, Netherland, Russia, Japan, China, Nigeria, etc. were also investigated [8][9][10]. Although the timeline or specific numbers differ, a similar trend of energy transition was found worldwide: from biomass (crop residuals, fuel wood, etc.) to coal, then oil and gas, afterwards from fossil fuel to nuclear or other renewable powers.

Although power generation, transportation and industrial applications do reflect the majority of fuel consumption, the residential sector is also an important and often the most price-sensitive sector relative to fuel utilization. Yet, this sector has significantly less benefit of technology and research and development efforts, likely due to the low profit margins and relative low cost associated with appliances. However, it was noted that domestic energy consumption and needs shaped the use of energy sources, especially at the initial stage of their rising [11]. For example, in Great Britain, the large consumption of coal was initiated by the increasing need for cheaper fuels in domestic heating as the fuelwood price was beyond the affordable level. This situation occurred when the British population grew rapidly and its once seemingly inexhaustible forests shrank at an alarming rate [12][13]. In the U.S., studies also show that the major market for anthracite coal during the first decade of the trade was American homes [14]. This is the same situation for oil industry. The initiation of crude oil exploration and extraction was not motivated by the transportation or industrial utilization, but by residential lighting [15][16][17].

Therefore, investigating residential sectot of energy consumption is of great importance, especially in considering future fuel consumption scenarios., The following sections conclude the development of residential fuel consumption.

2.1.1 *Before the 1900s: transition from biomass to coal*

The transition from wood to coal in the residential sector was accompanied by population growth and the rapid development of industry. This combined effect created a large demand increase for fuelwood. As a result of possible limited supplies of wood, people sought alternative fuels to replace what potentially looked like increasingly more expensive fuelwood. This residential fuel transition on a massive scale first occurred in England.

The population in England increased dramatically by more than 80% from 3.02 million in 1541 to 5.47 million in 1656 [18]. Despite this, the population explosion didn't exert significant influence on the fuelwood supply for residential houses. The residential fuelwood could still be secured in a lot of regions by planting of trees in backyards, on roadsides, on otherwise infertile slope land or in fuelwood groves to supply nearby farms or villages [8]. In England, the fuelwood prices were actually fairly stable between 1550 and 1650 [19]. Wilson [20] also noted that, from 1450 to 1650, while timber prices in England increased, however, they did not increase as much as the cost of other agricultural products. In fact, timber became cheaper year by year relative to inflation, even with an increasing demand from the residential sector. Hence, the population growth didn't exert that much high demand for wood supply and was not the major reason for the fuelwood price increase.

Before the 18th century, industry applications also used fuelwood as their energy source. Surprisingly, even though coal possesses more than 33% more energy density than fuelwood and also at a lower price, the industry didn't switch their fuel from fuelwood to coal that fast. The smoky and sulphurous nature of coal made its adoption difficult for industries that required smokeless or odorless fuels [21]. Further, the slow transition from fuelwood to coal in industry was not because of a lack of clean coal burning technology, but more because the industry was still profiting a lot from burning fuelwood. The energy density benefit of replacing fuelwood with coal couldn't balance out the capital investment of adjusting the combustion devices and coal transportation. Similar to the current situation, momentum in the industry and conservative tendencies usually slow new technology adoption. Fuel interchangeability studies from fuelwood to coal in industrial applications were already conducted in the Elizabethan Age. In 1590, John Thornbrough, the Dean of York, was granted a seven-year project to remove the “piercing and acrimonious spirits” for beer making and processing alum. By 1610, there were already patent applications using coal as fuel in baking of malt, bread, bricks, tiles and smelting of bell metal copper, brass, iron, lead and glass [22][23]. Although the possibility of utilizing coal was realized and numerous studies were conducted to adapt combustion devices from wood to coal, coal remained an insignificant industry fuel until the 17th century.

The first recorded transition from fuelwood to coal in England started in the residential sector of London. From 1500 to 1600, the population of London increased four-fold which led to a great increase in demand for food and industrial products. The forests in the land surrounding London were cleared to provide fuel sources and more land for agricultural purposes [13]. Although the need for residential heating increased with the population growth in London, the amount of wood consumption in the residential sector was still much less than that consumed in industry during that time. According to the study results on historical energy consumption, 20 kg of charcoal could be used to produce only 1 kg of iron (600 MJ/kg) in England during the Middle Ages [8]. The glass industry was also wood-intensive. To produce 1 kg of glass, 2.4 tons of wood needed to be consumed for both heating and to obtain potassium (90 MJ/kg). Salt production is also one of the major wood-consuming sectors, which demands as much as 500-600 MJ wood consumption per kilogram of salt produced [24].

With the development of industrial technologies, although the energy consumption per unit industrial product kept decreasing, it still couldn’t change the fact that fuelwood has a lower energy density and the supply was limited due to the slow recovery of forests. This large amount of fuelwood requirement in the industrial sector caused severe forest destruction and eventually fuelwood prices increased. Therefore, residents sought cheaper alternative fuels. In the latter half of the 17th century, the coal price in England was less than half of fuelwood per unit of gross energy [25]. This fuel price gap between fuelwood and coal was more significant in densely populated London than the England average. In Table 1, as the fuel prices and the general living cost level in London are all normalized to 100 from the year 1451 to 1500, coal saw a steady increasing trend with the living cost over the next 190 years. Fuelwood prices also started to increase significantly by the end of the 1500s. By 1642, the cost of fuelwood was more than double the price of coal.

Table 1: Indices of estimated price movements in London 1451-1642 [9]

Commodity	1451-1500	1531-40	1551-60	1583-92	1603-12	1613-22	1623-32	1633-42
General	100	105	132	198	251	257	282	291
Firewood	100	94	163	277	366	457	677	780
Coal	100	89	147	186	295	371	442	321

As the residents in London increasingly adopted coal as the substitute for fuelwood, imports of coal to London increased significantly in the last decade of the 16th century. Table 2 shows the coal imports records from the late 16th century to the late 17th century. As can be seen, the coal consumption in London increased significantly from 10,785 ton up to 361,189 ton in a century.

It should be noted that this coal consumption increase was mainly caused by the residential sector needs, since the technology for using coal in industry was still not widely adopted in the 17th century. The evidence could be found in the iron industry. In the 17th century, iron smelting was a voracious consumer of fuelwood in London. The dependence of smelting on the increasingly scarce and high-priced fuelwood supplies in the 17th century led to a situation in which the conversion of iron ore into metal in England was temporarily checked. However, this situation didn’t make the iron industry turn to coal combustion. Instead, a policy was released by the government to encourage imports of intermediate product pig iron instead of raw iron ore. Therefore, the rapid growth of coal production from the mid-16th century to the late 17th century represents largely the substitution of coal for wood as a household fuel [9].

Table 2: Imports of coal into London 1580-1680 [26]

Year	Period	Tons	Notes
1580	12 March-18 Sept.	10,785	
1585-6	Michaelmas-Michaelmas	23,867	
1591-2	Michaelmas-Michaelmas	34,757	
1605-6	Christmas-Christmas	73,984	
1614-15		91,599	One week missing
1637-8		142,579	Two weeks missing; a year of bad trade
1667-8	Midsummer-Midsummer	264,212	
1680-1	Michaelmas-Michaelmas	361,189	

The earliest coal shipped to London was from Newcastle as ballast, which was known as "sea-coal" [27]. The burning of sea-coal was reported to cause smoke with pungent odor [28]. Therefore, presumably sea-coal was of low quality and might have relatively high sulfur content in it. Sea-coal was first adopted by the poor residents in London due to its cheap price. To adapt the wood burning fireplace to the sea-coal, chimneys were built or modified to vent the smoke out of the house. Therefore, the number of chimneys of residential houses in London increased greatly around the 1550s. The acceptance of the sea-coal by the upper classes and nobility followed the steps of lower-class residents with a time leg. The use of sea-coal as a domestic heating source in royal house was a slow process, since Queen Elizabeth I was "greatly grieved and annoyed with the taste and smoke of sea-coal". After her death in 1603, James VI of Scotland became James I of England, during whose governing period, the popularity of using coal in residential houses was gained. Scotland was short of wood but had indigenous less sulphurous coals which led to coal being used in houses of Scottish nobles much earlier than in England. Therefore, the new king continued his old habit of using coal for domestic heating in England. Such behavior aided the adoption of coal as a domestic fuel even for wealthy London households [22].

It was the high demand for coal consumption in residential households that spurred the early development of coal mining and transportation. Coal provided the power source and encouraged the Industrial Revolution. Although a bit later, the U.S. went through a very similar pathway in the fuel transition from fuelwood to coal. In the U.S., coal surpassed fuelwood's energy supply in the 19th century, which is around 200 years later than the energy transition of Britain [29]. The early records in the U.S. of coal trading was in Pennsylvania and the major market for the first decade of the anthracite coal trade in Pennsylvania was the American homes [30]. Therefore, when the early coal boosters tried to quantify the potential market in New York, they counted the numbers of homes, not the numbers of factories or steam engines [11].

Similar to the situation of Britain, the early coal utilization in the U.S. was hindered by the poor transportation situation around the coalfields in the east coast. Even if Pennsylvania's anthracite coalfields were only a hundred miles away from the eastern seaboard, transporting coal to Philadelphia and New York was still extremely expensive in the early 19th century. In the year 1810, it cost more to ship coal less than a hundred miles to Philadelphia than delivering comparable shipments three thousand miles from England [31]. However, the war of 1812 tripled the price of the imported coal from Britain. During this time, emphasis on energy security resulted in local anthracite coal in Philadelphia gaining a higher market share in the east coast [32]. The real booster of the U.S. domestic coal consumption in the east coast was the construction of the Schuylkill Canal in the 1820s. In the 1810s, one ton of anthracite coal cost around 20 U.S. dollars. However, this price kept dropping to 8.4 dollars in 1820 and then 6.5 dollars in 1830. In the 1840s, the coal price in the east coast of the U.S. went down to around 4 dollars per ton [33]. Due to the availability of cheap coal, fuel interchangeability studies on fuelwood to coal in residential appliances were conducted by scientists and appliance manufactures. Between the year 1815 and 1839, 329 patents for coal combustion stoves were issued in the U.S. [34]. In the early adoption stage, a coal-burning stove cost around 30 dollars while the average labor only earned 2 dollars a day [35]. However, by the early 1830s, a domestic coal stove cost less than 10 dollars [11].

The Industrial Revolution further speeded up the energy transition from fuelwood to coal, especially when steam engines became widely adopted in the late 18th and early 19th century. However, as noted above, this energy transition was first initiated by the residential sector. After coal was widely adopted in residential cooking and heating, the accessibility to cheap coal paved its way to more applications in industry thus fostered the Industrial Revolution.

2.1.2 *From the 19th century to the 1950s: coal to petroleum and manufactured gases*

Besides the transition to coal utilization, the residential market also played an essential role on the development of the petroleum industry and trade, especially in its initial development stage in the latter half of the 19th century. The combination of urbanization and industrialization in both America and Europe led to a great need for lighting. Therefore, petroleum was first refined to obtain kerosene, which was a lighting source to replace diminishing and increasingly expensive whale oil [11][36][37]. The early development era of petroleum industry is also called “The Age of Illumination” [38].

In the early age of petroleum industry, instead of being a major petroleum product as it is today, gasoline was only a surplus byproduct which was usually burned at refineries, converted to gaseous fuel for gas lights or just dumped into the atmosphere due to its high volatility. Being a waste product, cheap gasoline stimulated the development of engines in the late 19th and early 20th century. For example, when Mercedes entered the U.S. market in the early 20th century, the price per car was at $12,450 ($353,261 in 2019 values) but the gasoline price was only 7 cents/gallon (around $2/gallon in 2019 values) [37]. With the accessibility to cheap and abundant gasoline, auto industry invested more money into automobile technologies and the prices dropped significantly. For example, when Ford first introduced its Model T to the market in 1908, the price was at $950, which dropped down to $269 by 1923 [39]. However, before the electric lighting and automobiles gained popularity in the first decade of the 20th century, kerosene was the major focus of the petroleum industry for providing light. Since the beginning of the petroleum industry, techniques were focusing on maximizing the production of kerosene until the second decade of the 20th century. As shown in Fig.2, although most of the crude oil is refined to gasoline and jet fuel for planes today, in the 1870s, more than 80% of a barrel of crude petroleum was transformed into illuminating oil [7][38].

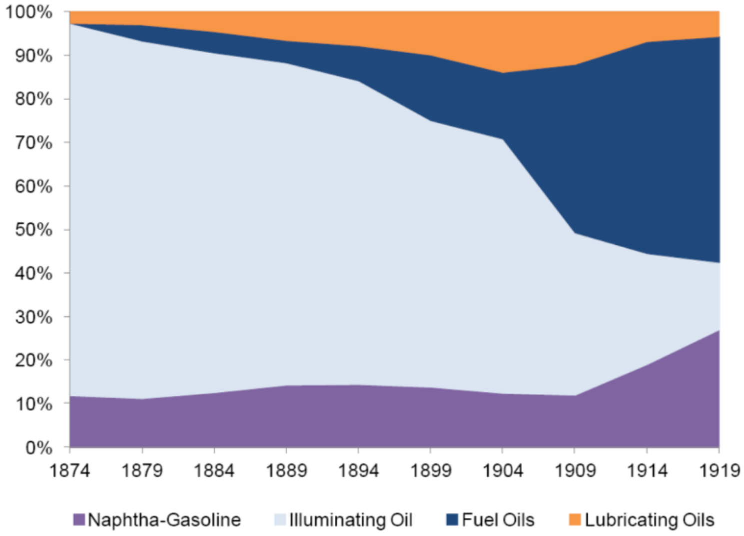


Figure 2: Shares of U.S. oil refinery output from 1874 to 1919 [7]

As petroleum (kerosene) found its use in domestic lighting applications in the late 19th century and early 20th century, the energy transition in domestic cooking and heating was from coal to different kinds of manufactured gases. Although it is hard to track who was the first to produce manufactured gas, Becker and Clayton are believed to be the early pioneers who invented methods to produce combustible gases from coal. In 1681, Johann Becker from Germany discovered that combustible gases could be generated by heating coal in the absence of air. Three years later, John Clayton from England collected combustible gas using a similar coal treatment method and called this gas “Spirit of Coals”. The commercialization of manufactured gas in residential and industrial applications was initiated by the foundation of London and Westminster Gas Light and Coke

Company in 1812. In the same year, this company built the world's first commercialized gas networks in Great Peter Street, Westminster, London by laying wooden pipes and illuminated Westminster Bridge with gas lamps on New Year's Eve in 1813 [40]. Due to its clean combustion performance and ease of transporting through pipelines compared to coal, manufactured gas quickly gained popularity in the early 19th century. The prosperity of the gas industry was accompanied with emerging inventions of gas combustion appliances. In 1826, the world's first gas stove was designed in England by James Sharp [41]. In the 1850s, Robert Bunsen invented the aspirated burner (Bunsen burner) which significantly influenced the gas applications in residential and industrial applications till today [42]. In the 1870s, Dr. Carl Auer von Welsbach invented the incandescent gas light mantle, which solved inefficient combustion of open-flame burners in streetlamps [43]. Moreover, water heaters, room heaters and many other appliances such as soldering irons and hair-curling tongs appeared on the scene in the mid to late 1800s [40]. To meet the increasing customer base, more gas networks were built in Britain along with manufactured gas productions sites, which are shown in Figure 3. Due to the widespread application of manufactured gas, manufactured gas was also called “town gas” in U.K. [44].

Figure 3: Known and suspected locations of manufactured gas sites in the British Isle [45]

The United States started the manufactured gas era at the same time as Britain. In 1816, the first manufactured gas company in America-the Gas Light Company of Baltimore was founded.

On 7th February 1817, the first manufactured gas streetlamp was lit in Baltimore. This was followed by the foundation of Boston Gas Light Company in 1822, and the New York Gas Light Company in 1825 [40].

It should be noted that the early manufactured gas companies generated their gases from coal, except for the Gas Light Company of Baltimore which produced its gases from the distillation of pine tar. However, it switched to coal gas in 1822, due to the maturity of the coal gas production technologies and more satisfactory gas qualities [40]. Manufactured gas is a general term for multiple man-made gases, including coal gas, producer gas, water gas, carbureted water gas, oil (petroleum) gas, etc.. The name of a particular manufactured gas depends both the source and the gas production procedure. Overlaps in the naming system for manufactured gases is apparent. For example, producer gas can also be called coal gas if it is generated by coal. Water gas was produced in a gasification process in a carbon fuel bed with steam, and coal was decomposed into hydrogen and carbon monoxide [46]. Carbureted water gas is the water gas with added carbon contents to increase the heating value of the gas, which was achieved by adding oil to the hot gas in the presence of steam then was subsequently thermally cracked to gaseous constituents [47]. Oil (petroleum) gas is produced in a gasification process where oil is thermally cracked in the presence of steam to produce a fuel gas.

Due to the variety of sources and production procedures, manufactured gases vary in composition and heating values. Table 3 shows the species percentages in selected gaseous fuels. The major combustible fuels include hydrogen, carbon monoxide, methane and ethane. The diluents are mainly nitrogen and carbon dioxide. It should be noted a range exists for given specific gas species in different manufactured gases or natural gas. Table 3 shows a few representative values from Reference [48] and [49].

Table 3: Properties of selected manufactured gases and natural gas [49]

Contents	Producer gas	Water gas	Carbureted water gas [48]	Coke oven gas	Blast furnace gas	Oil gas	Natural gas
H_2	15%	49.7%	40.5%	57.0%	3.7%	50.0%	0.0%
CO	24.7%	39.8%	34%	5.9%	26.3%	10.2%	0.0%
CH_4	2.3%	1.3%	8.9%	29.7%	0.0%	27.6%	94.5%
C_2H_6	0.0%	0.1%		1.1%	0.0%	-	0.5%
CO_2	4.8%	3.4%	-	1.5%	12.9%	2.6%	0.2%
N_2	52.2%	5.5%	-	0.7%	57.1%	5.1%	4.0%
O_2	0.2%	0.2	-	0.0%	0.0%	0.2%	0.3%
Lower Heating Value (MJ/m^3)	5.8	11.4	20.1	21.5	3.9	19.7	35.7

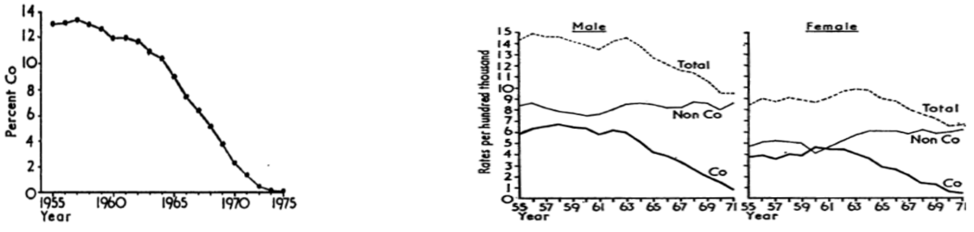
Compared to wood and coal burning, gaseous fuel combustion offers numerous advantages, including absence of ash, cleanliness, ease of end user control and flexible combustion performance, etc.. Moreover, the pipeline construction decreased the price of gaseous fuel transport dramatically compared to solid fuels. The calorific value of manufactured gases can also be altered by mixing various gases, based on the end user's needs.

However, the variety of gaseous species and the lack of fuel species regulation or control also brought numerous problems. The major threats brought by manufactured gases were explosion accidents due to the wide flammable range of carbon monoxide and hydrogen, and also carbon monoxide poisoning. For example, the Fire Research Station of the U.K. reported 1,007 town gas explosion accidents between 1957 to 1968, which is a time period approaching the end stage of town gas era [50]. The safety awareness in the 1950s and 1960s period was relatively high, and the technologies of handling manufactured gas were also mature, but explosion tragedies still occurred occasionally. Therefore, it is believed that more accidents happened in the early age of the manufactured gas era in the 19th century without detailed records.

Carbon monoxide poisoning was another significant issue with manufactured gas use. The leakage of manufactured gas from the pipeline into residential houses can be detrimental. As shown in Table 3, the carbon monoxide percentage in manufactured gases range from around 6%

to 40% and it can result in human death at ppm level in minutes [51]. Studies show that carbon monoxide poisoning can be more harmful for the elderly compared to younger people. For example, Chalke et al. [52] pointed out that people over 65 years old are more prone to carbon monoxide poisoning due to the loss of sense of smell.

Moreover, the ease access to carbon monoxide through the residential pipeline also tended to increase the suicide rate [53]. Figure 4 shows the relationship between CO percentage in the residential gas supply and the suicide rates in England and Wales. In the 1940s, the CO percentage in town gas of U.K was between 10% and 20%. In the 1950s, a method of manufactured gas from oil products and naphtha began to be adopted, which decreased the CO percentage in this noncoal-based gas down to around 1%. Meanwhile, natural gas was discovered worldwide and it quickly gained popularity in the U.K. due to its high heating value, CO-free, clean combustion performance and continuing decreasing price due to its abundant supply. By 1971, around 69% of the gas in the U.K. domestic gas supply system was natural gas. As can be seen in Figure 4(a), CO was almost depleted from the U.K. residential gas supply system by 1975. Figure 4(b) shows the correlated suicide rates of England and Wales, which clearly notes the rate of CO-poisoning suicide decreased significantly as there was less access to CO from the pipeline in homes.



(a) CO percentage in domestic gas of U.K.　　*(b) England and Wales: suicide rates by mode of death*

Figure 4: Relationship between residential gas species and suicide rates of U.K. [54]

2.1.3 *From the 1950s to the 1990s: manufactured gas to natural gas and LNG*

The nation-wide organized transition from manufactured gas to natural gas occurred in the U.K., which is a good example of government assisted energy transition. In 1948, the U.K.

Parliament passed the Gas Act and nationalized the gas industry through amalgamation. At that time, 1,050 gas works existed, supplying a total of 2119 million therms to 11.3 million consumers [40]. The Gas Act also established the British Gas Council with 12 Area Gas Boards which were responsible for arranging their own supplies and finances. The British Gas Council consisted the 12 chairmen of the Area Boards, a Deputy Chairman and a Chairman, whose role was to advise the government of the policies in gas industry and coordinate different Area Boards.

A series of policies promoted by the British Gas Council changed the British gas industry significantly. For example, after the Gas Council was founded, 622 old gasworks were closed, some of the larger works were extended and linked together, and 21,200 miles of new gas mains were laid. Therefore, by 1962, the town gas production units deceased from 1050 in 1949 down to 341, of which 74 units produced around 73% of the nation’s town gas [55]. With the development of the industrial sector, the gas consumption in industry rose from 639 million therms (year 1953) to 819 million therms (year 1960).

Contrary to the wide adoption of town gas in industry, the town gas consumption in residential houses decreased from 1366 (year 1953) to 1268 million therms (year 1960). This was mainly due to the price increase of town gas which led the residential sector to look for alternative fuels. From 1950 to 1960, the town gas price increased by 65%, however, the electricity price only increased 25% over the same time period. The major reason for the town gas price increase was increasing cost and difficulty in sustaining suitable supplies of coking coal, for which the steel industry was also a major customer [56]. Facing these difficulties, the British Gas Council proposed three solutions:

1. Gasification of alternative lower grade coal.
2. Gas production from petroleum instead of coal.

3. Introduction of natural gas to enrich manufactured gas.

Considering the cost and technology availabilities, the third solution became the final answer to solve the dilemma of British gas industry. In 1953, the British Council started to work with the BP Exploration Company to find natural gas reserve all over the British Isle. The discovery of natural gas fields around the world also stimulated the British Gas Council to explore the overseas market. In the late 1950s, British Gas Council started the plan to explore the North Sea and also started to import LNG from the American Gulf Coast, Algeria, etc.[55].

In this manufactured gas to natural gas transition, combustion devices adaptation to natural gas posed a significant challenge. Natural gas has a volumetric heating value around twice that of town gas. Other fuel property differences like density, flame speed, etc. also brought difficulties for existing combustion devices to switch from town gas to natural gas without modification. At first, it was proposed to blend other gaseous fuels into natural gas to make a fuel mixture that possess similar burning properties as town gas while still keeping the heating value of this new fuel higher than that of town gas. This type of gas was once called “GS gas”. However, experiments showed that the old town gas appliance burners still needed modification, even with the specially made “GS gas”. Therefore, with the abundance supply and the decreasing price of natural gas, the British Council determined to take a one-step action, which was to replace the old residential appliance burners with new ones that were compatible with natural gas, instead of adopting the more expensive two-step way to alternate both the fuel and appliances.

Starting from 1966, the appliance-conversion act took almost 10 years with 40 million appliances from 14 million end users. By 1972, appliances from 6 million families were converted, and in the same year the British Gas Council changed its name to British Gas Corporation with more concentrated power authorized by the government after the 1972 Gas Act [56][57][58]. By the end of 1978, around 13.4 million town gas end users converted to natural gas [40]. Table 4 shows the progress of the British gas end users converting from town gas to natural gas.

Table 4: Progress of customer conversion to natural gas in the U.K from the 1960s to the 1970s [40]

Year (April to March)	Total customers	Annual number converted to NG (000s)	Cumulative number converted to NG (000s)
1967/68	13,210	51	51
1968/69	13,265	418	469
1969/70	13,347	1,093	1,562
1970/71	13,372	2,029	3,591
1971/72	13,390	2,407	5,998
1972/73	13,506	2,100	8,098
1973/74	13,559	2,108	10,206
1974/75	13,682	1,674	11,880
1975/76	13,925	1,131	13,011
1976/77	14,200	329	13,340
1977/78	14,516	98	13,438

This town gas to natural gas transition in residential homes was a huge task in British energy development history. More than 13.5 million sites had to be visited with 35 million appliances (200 million burners) and more than 8000 residential appliance models to convert. Because conversion always brings inconvenience to the residential gas end users, the British Gas Council (British Gas Corporation after 1972) determined to provide advantageous terms for new appliance purchase or even free of charge, when the old appliances couldn’t be converted successfully. The British Gas Council also decided to supply the replacement parts to residential houses, so the conversion can be done on site. This was estimated as a more efficient and cheaper way to complete the conversion, compared to shipping the appliances to specific places and conducting conversion there. Figure 5 shows an on-site town gas to natural gas transition test on streetlamps in England. To achieve this fuel conversion, not only training/education programs were provided to technicians, the marketing strategies/advertisement of natural gas to the public were also equally, if not more, emphasized. For example, the first demonstration fuel conversion project was done on Canvey Island, which gained public confidence of the fuel conversion. To assist the transition, the Conversion Executive was founded to coordinate among the British Gas Council, Area Boards and the Society of British Industries, which represented the benefit of appliance manufactures and contractors. At the same time, public relation strategies were adopted including providing conversion handbooks/manuals, developing better understanding of local conditions and approaching gas end users of different social classes/income levels using varying methods [59][60].

The successful conversion in Central London of Buckingham Palace, Parliament, the Bank of England and Westminster Abbey acquired symbolic status in fuel conversion of the British upper class. However, different problems were encountered in other places of Britain. For example, cultural and linguistic issues became a hindering stone in North East/West, East Midlands, etc.. Specific lobby teams were developed to reach out to Asian immigrant communities, where especially women expressed greater concern on the work of fuel conversion [55]. In May 1968, a town gas explosion occurred at Ronan Point of London, which resulted in the death of 4 people and injury of 17. The tragic accident further shook the public confidence in town gas during its campaign with natural gas. This also provided a window of opportunity for the natural gas supporters to reduce the public concern on converting from town gas to natural gas. For example, the government and the British Gas Council commissioned a report by Prof. Frank Morton of Manchester University addressing the concerns with natural gas and delivering the advantages of the fuel transition like reducing explosion accidents and gas poisoning [55][61].

Figure 5: Town gas to natural gas transition test on streetlamps in a Midlands town in England [40]

The final cost of the fuel transition from town gas to natural gas in the U.K. was £577 million (1978 currency), which was less than the estimated £400 million in 1966 considering inflation. However, other costs, like writing off obsolete plants, brought the total bill to £1027 million. In the April 1977/March 1978 Annual Report, British Gas Acclaimed: “The cost of the entire conversion programme was met without external subsidy of any kind.” [40].

The United States experienced a different procedure of adopting natural gas in residential homes compared to the British way. Before the gas transition, Britain already built a relatively mature town gas industry and gas transporting system since the 1800s, and then experienced an organized state-led town gas to natural gas transition by an almost monopolistic strategy developed by the British Gas Council and the British Government. However, being the world’s largest bastion of free market and enterpreneurialship, the U.S. natural gas industry was developed in parallel with its manufactured gas industry due to the abundance cheap supply of natural gas from international trades. From the late 1800s to the early 1900s, U.S. gas companies provided both manufactured gas and natural gas separately to customers depending on their specific needs. In the U.S., the sale of natural gas exceeded that of manufactured gas in 1935. In the late 1940s, the manufactured gas began to be phased out and ultimately replaced by natural gas in the 1950s [40]. However, in that time, U.K. hadn’t started the town gas to natural gas conversion project yet.

The first recorded usage of natural gas in the U.S. was from Fredonia, New York in 1821. Local residents used logs and later lead pipes to transport the gas to nearby houses for illumination [62]. This first known natural gas practical application in New York was 4 years before the foundation of New York Gas Light Company. However, in the United States, from the discovery of natural gas in 1821 to its sale exceeding manufactured gas in 1935, enormous volumes of natural gas were flared due to the lack of local usage or in hoping that oil might be lying under the gas. It was estimated that around 76 trillion cubic feet natural gas were wasted from the early days of oil industry to the late 1940s [40]. In the late 1800s, the natural gas industry started to thrive due to the development of pipeline transporting technology and discovery of natural gas on the U.S. land. In 1883, the Penn Fuel Company started to distribute natural gas by pipeline to customers in Pittsburgh’s East Liberty and Lawrenceville neighborhoods from Haymarket Well [63]. The first long distance pipeline (217-mile/14- to 18-inch) transporting natural gas was completed in 1925 by the Mongolia Gas Company of Texas from northern Louisiana to Beaumont, Texas [64]. In 1947, the original oil-transporting pipelines (the “Big Inch” and the “Little Big Inch”) connecting Texas and the U.S. east coast converted the fuel to natural gas after they were sold to Texas Eastern Transmission Corporation after the Second World War [65]. Natural gas found wide application through the U.S., and natural gas pipelines were lying in every one of the Lower 48 states by the year 1966. The natural gas consumption by American residential end users also increased dramatically, as shown in Table 5.

Table 5: U.S. domestic natural gas consumption from 1945 to 1977 [40]

Year	Total customers (millions)	Total consumption (trillion Btu)	Average consumption (million Btu/customer)
1945	18.6	775	41.6
1950	22.1	1384	62.6
1955	26.3	2239	85.2
1960	30.4	3188	104.8
1965	34.3	3999	116.5
1970	38.1	4924	129.2
1975	40.9	4991	121.9
1977	41.7	4946	118.7

The natural gas consumed in the U.S. was not only from domestic sites but also imported from Canada, Mexico or Algeria in liquid form since the 1950s. The natural gases on the energy market possess distinctive properties depending on their sources. Table 6 summarizes the components and their relative abundance in investigated natural gases on the U.S. market. As shown, the major functional content of natural gas is methane, together with some higher hydrocarbons. The diluents are usually nitrogen, carbon dioxide, or a trace of oxygen. It should be noted that the contents of the natural gas in Table 6 are only the ones investigated, natural gas from non-investigated resources might extend the percentage of these species into a larger range.

Table 6: Variation in composition of natural gas on the U.S. market [66]

	Volume (%)			
Component	Mean	Standard deviation	Minimum	Maximum
CH_4	93.0	5.5	73	99
C_2H_6	3.0	2.6	0	13
C_3H_8	1.0	1.4	0	8
C_4H_{10}	0.5	1.0	0	7
C_5H_{12}	0.1	0.3	0	3
C_6H_{14}	0.1	0.1	0	1
N_2	1.5	2.9	0	17
CO_2	0.5	0.5	0	2

With natural gas species varying from different sources, the natural gas either mined or purchased by the U.S. gas utility companies also varies significantly. Lacking the unified national natural gas quality regulation, gas companies in the U.S. developed their own standards on the natural gas species, heating value, sulfur contents, and so on. Table 7 shows the natural gas quality regulations of some gas utility companies in the United States. Some of these regulations are modified every few years. For example, in 2019, SoCalGas decreased their natural gas minimum heating value from 990 Btu/scf to 970 Btu/scf [67].

Table 7: Natural gas quality standards adopted by gas companies of the United States [67][68]

Pipeline Company	Heating value (Btu/scf)		Water content	Various inerts			Hydrogen sulfide (H_2S)
	Min	Max	Lbs/MMscf	CO_2	O_2	Total inerts	(Grain/100scf)
SoCalGas	970	1150	7	3%	0.2%	4%	0.25
Dominion Transmission	967	1100	7	3%	0.2%	5%	0.25
Equitrans LP	970	-	7	3%	0.2%	4%	0.3
Florida Gas Transmission Co.	1000	1110	7	1%	0.25%	3%	0.25
Colorado Intrastate Gas Co.	968	1235	7	3%	0.001%	-	0.25
Questar Pipeline Co.	950	1150	5	2%	0.1%	3%	0.25
Gas Transmission Northwest Co.	995	-	4	2%	0.4%	-	0.25

Fuel interchangeability studies in the United States started from the manufactured gas era, and is still under progress for natural gas from different sources. Although lacking government-led uniform gas transition strategy like the U.K., early records of fuel interchangeability studies being conducted on manufactured gases and natural gas in the late 1800s and early 1900s from some U.S. institutes are noted.

In the late 19th century, the large cost increase of gas-making materials resulted in frequent manufactured gas price increases in the United States. In some localities, the gas price increase compelled a decrease in standards of quality. By that time, natural gas production was not as abundant as today. Therefore, a variety of gases were available on market including natural gas, manufactured gas or artificially lower-standard manufactured gas. As a result, the investigation of residential burner mechanisms operating on multiple gases were conducted. The American Gas Association started to work on the project of improving the gas utilization efficiency, which was one of its earliest tasks after its foundation in 1918. Meanwhile, the engineering section of the U.S. Bureau of Standards started to investigate on the design and operation of atmospheric burners, which were, and still are, commonly adopted in residential appliances. Through these studies in the early 1900s, better understanding of residential appliance burner working principles were obtained [69]. Later on in 1932, other than the burner efficiency, reducing emissions from combustion like carbon monoxide also became a consideration in residential burner design [70]. Therefore, more detailed residential burner optimization started to be conducted considering efficiency, flame characteristics, carbon monoxide emission, etc. [71]. The test setup for residential burners is shown in Figure 6.

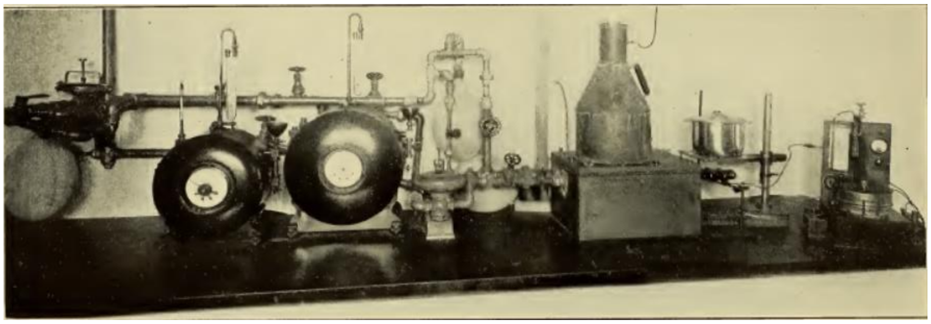


Figure 6: Apparatus setup for studying burner performance in Bureau of Standards of the U.S. in the 1930s [71]

American Gas Association starting in 1927. This 6-year long project was called Mixed Gas Research, which conducted around 175,000 individual tests involving more than 250 gases. In 1936, AGA released their general formula “AGA Index C” to represent the gas interchangeability where C= the index of change in performance of appliances”. AGA Index C=H/Sqrt(D), where H is the gas heating value and D is the gas specific gravity [72]. This index is nowadays referred as Wobbe Index, which is widely adopted around the world in gaseous fuel interchangeability studies. Not soon after, researchers realized that a single Wobbe Index was not enough to predict all the combustion behaviors, therefore, more indices regarding flame blow-off, flashback, yellow tip and so on were developed to examine the performance of residential burners. From the perspective of technical development, these preliminary fuel interchangeability studies in the U.S. since the 1900s assisted the transition from manufactured gas to natural gas and also benefited the historical energy conversion of the U.K. in the 1950s.

2.1.4 *Since the 1980s: renewable gases*

With the depletion of natural gas reserves and the urgency of reducing carbon emissions to combat climate change, replacing pipeline natural gas with renewable gases has been of interest not only to fossil fuel dependent countries but also to the whole human society. Although many complexities remain to be sorted out, reducing carbon emissions by replacing fossil fuels with renewable energy is a strategy that has international consensus as evidenced by the endorsement of United Nations Framework Convention on Climate Change in 1992 [73]. For the residential sector, one of the most effective ways of adopting renewable energy is to replace the pipeline natural gas with renewable gases, as shown in Figure 7. The benefits of adopting renewable fuels in natural gas pipeline infrastructure include:

1. Reducing carbon emissions, thus combating climate change.
2. Extending the gaseous fuel from singular fossil fuel sources to more renewable sources like biogas, or hydrogen.
3. Obtaining energy security by alleviating the influence of natural gas price fluctuation.

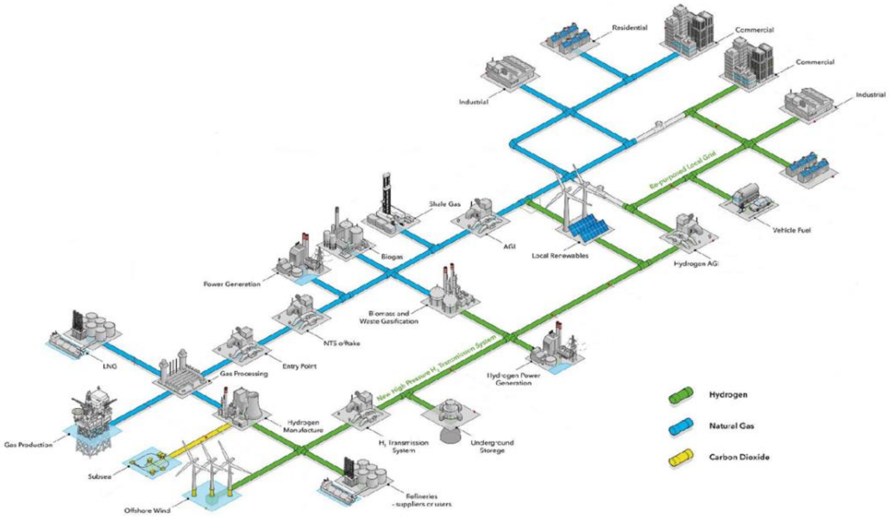


Figure 7: Blending renewable gases into existing natural gas infrastructure [74]

2.1.4.1 Biogas

Biogas can be produced from various sources, including wood and agricultural products [75][76][77], landfill waste [78][79][80], sewage [81][82][83], etc.. Although the biogas production sources may vary, the functional part in different sources is mainly biomass. The biogas sources influence the contents of biogas significantly and Table 8 shows typical biogas contents.

Table 8: Typical biogas contents [84]

Species	Units	Biogas		
		Sewage gas	Agricultural gas	Landfill gas
CH_4	%	66-75	45-75	45-55
CO_2	%	20-35	25-55	20-30
CO	%	<0.2	<0.2	<0.2
N_2	%	3.4	0.01-5	10-25
O_2	%	0.5	0.01-2	1-5
H_2	%	trace	0.5	0.0
H_2S	mg/Nm^3	<8000	10-30	<8000
NH_3	mg/Nm^3	Trace	0.01-2.5	trace
Siloxanes	mg/Nm^3	<0.1-5.0	trace	<0.1-5.0

The variation of biogas species is not only related to biomass sources, but also on gasification technology and procedures. The two major species in biogas are methane and carbon dioxide. Other common fuel species like carbon monoxide, hydrogen are present at much lower levels. Other species in raw biogas include nitrogen, oxygen, ammonia, hydrogen sulfide, etc.. However, some of these trace gases might be detrimental to pipeline or combustion devices through direct contact or combustion. Therefore, before blending biogas into the pipeline or supplied to combustion devices, the gas needs to be purified or processed to be “upgraded” to meet certain standards.

The major functional specie in biogas is methane, which is usually called biomethane due to its production source. The first known large scale biomethane blending into natural gas grid project was in Moenchengladbach, Germany. The sewage gas plant had a raw gas capacity of 400 m^3/day and injected around 20 million m^3 of purified biogas (biomethane) into the local gas grid from

1982 to 1996 [85]. This practice was later followed by the other countries in Europe [86], and also U.S. [87], Canada [88], Japan [89], etc. [90][91].

2.1.4.2 Renewable hydrogen

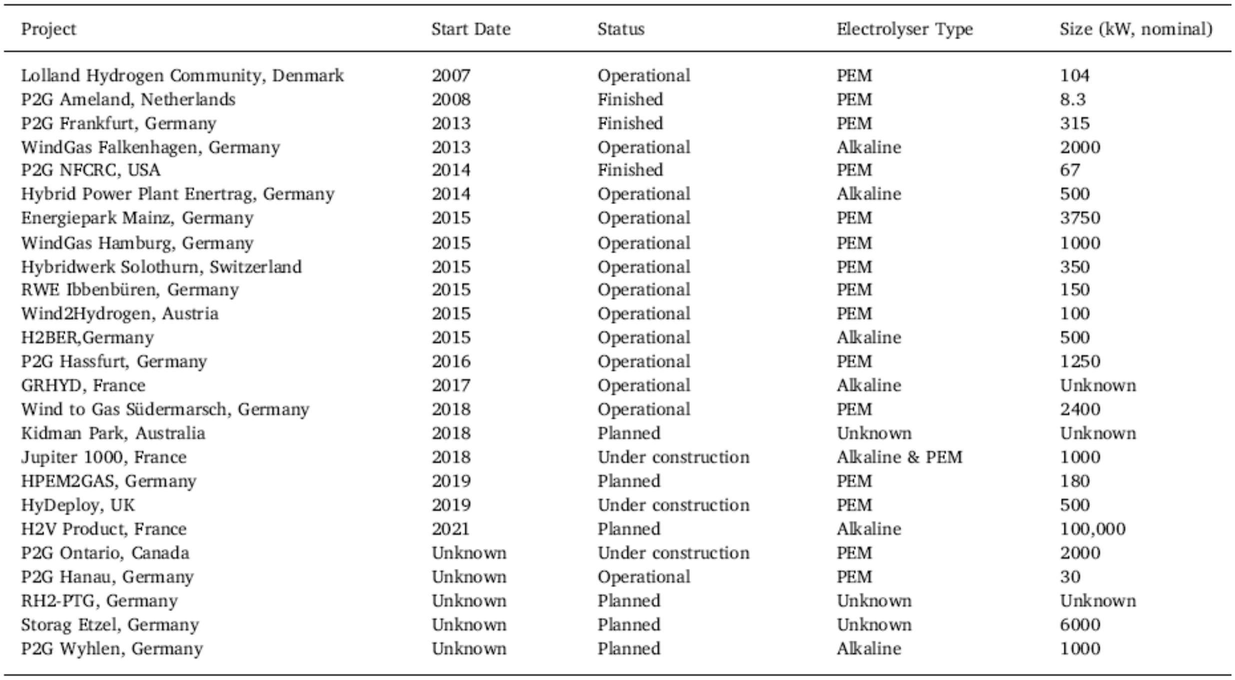
Hydrogen has long been touted as a long term solution for carbon reduction. Hydrogen from renewable sources has gained attention these years [92][93]. Hydrogen is the most abundant element in the universe and also a carbon-free combustible fuel. Besides combustion, hydrogen can also be readily used in other applications, like fuel cells, industrial refineries or ammonia production for agricultural fertilizers, etc.. Moreover, being the lightest element, hydrogen has a much higher heating value on the mass base compared to fossil fuels, which also makes it a competitive energy carrier in addition to being a fuel. Therefore, storing renewable energy (solar, wind, etc.) into hydrogen and transport hydrogen through pipelines or directly blend hydrogen into the existing natural gas infrastructure can be an effective way to achieve renewable energy adoption and carbon reduction goals.

Renewable hydrogen as a fuel supply and an energy carrier to power the world is far from being a new idea. The notion of using hydrogen from electrolization dates back to 100 years ago. In 1923, the famous British scientist J.B.S. Haldane published the book < Science and the future> [94], which noted “The country (Britain) will be covered with rows of metallic windmills working electric motors which…will be used for the electrolytic decomposition of water into oxygen and hydrogen. These gasses will be liquefied, and stored in vast vacuum jacketed reservoirs, probably sunk in the ground”. Considering hydrogen as a highly condensed energy carrier, Haldane also mentioned that hydrogen “will enable wind-energy to be stored, so that it can be expended for industry, transportation, heating, and lighting, as desired”. Compared to coal and petroleum, which were the fuels widely used in Britain of the 1920s, Haldane also mentioned the environment benefit of using hydrogen as “no smoke or ash will be produced”. Despite the promising utilization of hydrogen, Haldane did express his concerns regarding the capital investment by saying “the initial costs will be very considerable”. As the development of natural gas storage and transporting technology, the current cost of utilizing hydrogen in existing natural gas systems should be much lower than the year 1923 when Haldane made this prediction. Till today, hydrogen already found its wide application in industry, transportation, and is considered for residential heating.

Beyond Haldane’s suggestion of utilizing wind power, technologies are now available to generate renewable hydrogen from other sources, including renewable feedstocks [95], solar energy [96], nuclear power [97] or directly from water electrolysis by renewable grid power (P2G, Power to Gas) [98]. Moreover, hydrogen has already been integrated into pipeline infrastructure as discussed above. Recall that Britain once had a high percentage of hydrogen (around 50 vol%) in their town gas for over half a century. Therefore, blending hydrogen into our existing pipeline shouldn’t be as difficult as the very early adoption of hydrogen in the manufactured gas era. However, since current pipeline infrastructures around the world are mainly designed for natural gas, blending hydrogen into natural gas system warrants some technical considerations regarding the fuel property differences between natural gas and hydrogen.

Hydrogen has a much lower density compared to natural gas, and its high flame speed and wide flammability range limit its percentage in the current natural gas infrastructure. Therefore, the hydrogen utilization in natural gas grid is still in demonstration stage and hydrogen blending percentage is still very low (generally under 10 vol%). Table 9 lists the demonstration projects of hydrogen injection into existing natural gas pipeline infrastructures.

Table 9: Power-to-gas projects that include hydrogen injection to the gas grid [99]



Project	Start Date	Status	Electrolyser Type	Size (kW, nominal)
Lolland Hydrogen Community, Denmark	2007	Operational	PEM	104
P2G Ameland, Netherlands	2008	Finished	PEM	8.3
P2G Frankfurt, Germany	2013	Finished	PEM	315
WindGas Falkenhagen, Germany	2013	Operational	Alkaline	2000
P2G NFCRC, USA	2014	Finished	PEM	67
Hybrid Power Plant Enertrag, Germany	2014	Operational	Alkaline	500
Energiepark Mainz, Germany	2015	Operational	PEM	3750
WindGas Hamburg, Germany	2015	Operational	PEM	1000
Hybridwerk Solothurn, Switzerland	2015	Operational	PEM	350
RWE Ibbenbüren, Germany	2015	Operational	PEM	150
Wind2Hydrogen, Austria	2015	Operational	PEM	100
H2BER,Germany	2015	Operational	Alkaline	500
P2G Hassfurt, Germany	2016	Operational	PEM	1250
GRHYD, France	2017	Operational	Alkaline	Unknown
Wind to Gas Südermarsch, Germany	2018	Operational	PEM	2400
Kidman Park, Australia	2018	Planned	Unknown	Unknown
Jupiter 1000, France	2018	Under construction	Alkaline & PEM	1000
HPEM2GAS, Germany	2019	Planned	PEM	180
HyDeploy, UK	2019	Under construction	PEM	500
H2V Product, France	2021	Planned	Alkaline	100,000
P2G Ontario, Canada	Unknown	Under construction	PEM	2000
P2G Hanau, Germany	Unknown	Operational	PEM	30
RH2-PTG, Germany	Unknown	Planned	Unknown	Unknown
Storag Etzel, Germany	Unknown	Planned	Unknown	6000
P2G Wyhlen, Germany	Unknown	Planned	Alkaline	1000

Before trying to increase the percentage of hydrogen in residential pipelines, multiple sectors need to be considered including pipeline material, sealing, compression system adjustment, etc.. From the combustion perspective, the ability of end uses like residential appliances to safely operate on hydrogen/natural gas mixtures is important to assess. Further, what adjustment should be made to existing appliances to increase their tolerance level of hydrogen. Ironically, the process may look like a reversal of the retrofit program undertaken in the U.K. to adapt appliances to switch from town gas to natural gas.

Some preliminary studies have been conducted and are still ongoing in several countries. In the 1970s, Pangborn et al. [100] projected technical considerations on a typical residential atmospheric burner (designed for natural gas) operating on hydrogen. In Europe, the NATURALHY project was initiated in 2004, which aims to prepare for hydrogen economy. After the start of this project, De Vries et al. conducted both theoretical and experimental research to investigate the feasibility of using hydrogen in our current natural gas appliances. However, these studies mainly stayed on either theoretical level or only tested on fundamental burners [101][102][103][104].

In 2018, Jones et al. conducted tests on a commercial cooktop burner designed for natural gas and successfully injected 34.7% hydrogen into the cooktop burner without inducing flashback [105]. Around the same time, California Energy Commission (U.S.) sponsored University of California, Irvine to conduct a more comprehensive study on existing natural gas appliances operating on natural gas/biogas and natural gas/hydrogen mixtures. Using both experimental and numerical methods, this study investigated the combustion performance of multiple commercial residential appliances, including cooktop burner [106][107][108], oven burner [109], water heaters [110][111], room furnace [112], etc.. The investigated combustion performance includes flashback/blow-off limits, ignition behavior, burner temperature, emissions (CO, UHC, NO, NO_2, N_2O, NH_3), combustion noise, efficiency, etc.. In 2019, University of Zaragoza worked with BSH Home Appliances Group in Spain and worked on reaction mechanism simplification to generate numerical models predicting combustion behaviors of appliance operating on hydrogen-rich fuel [113]. Also in 2019, an appliance test project was initiated by E.ON, which is one of the largest companies in Europe working on P2G programs. E.ON planned to raise the hydrogen percentage in natural gas pipeline grid to as much as 20% in Schopsdorf (Germany). To help achieve this goal, 400 commercial residential appliances were selected to conduct tolerance tests [114]. These activities are summarized in Table 10.

Table 10: Tests on residential appliances operating on hydrogen enriched natural gas

Organization	Starting time	Appliances/Burners	Description	References
Gas Technology Institute, U.S.	1977	Atmospheric burner	Provided technical considerations of appliances operating on hydrogen-rich natural gas.	[100]
DNV GL Oil & Gas/University of	2004	Fundamental aspirating burner	Conducted fundamental analysis on natural gas/hydrogen mixture	[101][102][103][104]

Groningen, Netherlands			properties and conducted experiments on a Bunsen burner.	
Swansea University, U.K.; King Saud University, Saudi Arabia	2018	Cooktop burner	Conducted experiments on a cooktop burner operating on over 30 vol% hydrogen.	[105]
University of California, Irvine, U.S.	2018	Cooktop burner, oven burner, room furnace, water heaters, etc.	Conducted experiments on multiple appliances, and tested multiple performance (ignition, emissions, burner temperature, etc.) of various appliances operating on hydrogen/natural gas mixtures.	[106][107][108][109][110][111][112]
University of Zaragoza, BSH Home Appliances Group, Spain	2019	Cooktop burner	Developed simplified reaction mechanism to simulate the combustion performance of a cooktop burner.	[113]

2.1.5 *Conclusion*

When it comes to fuel consumption, technology advancement or emission control, the power generation, industry and transportation sectors are usually first in line. The residential sector, which is most related to our daily lives, is usually overlooked. However, as discussed above, nearly every energy transition in human history was either initiated by, or highly influenced by the residential sector. From fuel wood to coal, then to oil and different gaseous fuels, industry consumed large amounts which led to higher prices less affordable to residents. The old fuel depletion due to the industrial sector drives the residential sector to seek less expensive alternative fuels. For large fuel consumption sectors like industry, when the benefit of adopting the new fuel can balance out the cost of getting rid of their old fuel consumption devices, the fuel transition will occur in these high energy consuming sectors. Therefore, in the energy transition, industry usually has a time delay of adopting the new fuel due to the capital investment of their devices to adapt to the new energy source. However, the residential sector is more flexible and price sensitive due to the relatively low capital investment in appliances and more flexibility in consumer adoption/replacement tendencies. Examples besides appliances include electric vehicles, rooftop solar, IOT devices in the home, etc.. This places the residential sector in the pioneering position when energy transition occurs. Therefore, investigating in the energy transition in the residential sector is not only beneficial to the residential sector study itself, but can also help foresee future fuel transitions in other energy consuming sectors.

Fuel transitions have also been assisted or promoted by other factors, like government policies and stricter emission regulations. For example, after the London Great Smog in 1952, the necessity of replacing coal with cleaner fuels was finally widely accepted by the U.K. society. The Clean Air Act 1956 passed by the Parliament of the U.K. was principally in response to the smog, which accelerated the transition of the U.K. from coal consumption to gas adoption. This successful fuel transition is a great example of how rational policy can play a leading role in the energy transition and emission control.

Yet it is clear that air pollution problems in London were evident well before the 1950s. People were suffering from respiratory deceases due to coal combustion and church roofs were being damaged by the high sulfur coal ashes in the smog hundreds of years ago before the London Smog in 1952. Numerous policies were put in place before the Clean Air Act 1956, but they all failed in some manner. The successful adoption of the Clean Air Act of 1956 is usually attributed to the human reflection and awakening after the death tragedy in the 1952 London Smog, which finally ended the coal era in London. Under this government-led energy transition, another critical factor that helped facilitate the transition is the large amount of natural gas discovered in North Sea in the 1950s. While this discovery did not have the public impact of the London Smog episodes, it enabled Europe to use cheaper and cleaner natural gas than coal. As a result, one must ask: Did the discovery of cheap natural gas end the smog tragedy or did the Clean Air Act 1956? If the Clean Air Act were proposed in the Victorian era, would it be as successful as the one in 1956 or it was just another failed policy like its predecessors? It is apparent that transitions require many factors to properly align in order for action to occur.

The discussion above touches on general trends in fuel transition. Of course, not all countries in the world were participating in this transition or experienced the same processes. Even in the U.K. and the U.S., not every city was participating in the energy transition. For example, while the U.S. transitioned into the natural gas era more than 50 years ago, around 10% of U.S. residential houses still use fuelwood or coal for residential heating. Other countries may not follow this procedure or may skip several steps. For example, some developing countries in Africa or Asia continue to use biomass combustion for residential heating, including dry bushes, tree leaves, residuals of crops, etc., which corresponds to fuel consumption stage even before the fuelwood era in the developed world. However, these late developed countries may be able to skip some energy transition steps, and enter directly the renewable gas era by adopting biomass gasification technologies if assisted by the developed world. Certainly, the world must adopt renewable resources in the near future to help combat climate change.

2.2 Residential Electrification vs Combustion

2.2.1 *Lighting market*

A current debate is ongoing relative to transitioning from gas fuel combustion appliance to those run on electricity [115][116]. Such debates between residential devices operating on electricity vs gas fuel and have been taking place since the introduction of technologies that enabled widespread availability of the electric infrastructure in the 1870s. The first battlefield between gas and electricity industries was in the lighting market.

Before the 1870s, the lighting market was mainly dominated by kerosene and gas lighting. But, as shown in Table 11, electrical systems started to be introduced at this time, offering a possible alternative for lighting. In 1877, the first electric arc lamp was tested on the street of Paris, which triggered interest in adopting electric lighting in Europe. In 1878, London started to test on electric streetlamps, however, this project was ended a few months later due to the high cost of relatively scarce electricity. While the electric lighting systems were being tested in the 1870s, the gas industry invested in high efficiency gas lamps to compete with electric lamps. In 1877, Dr. Carl Auer von Welsbach opened the first factory in London to produce incandescent gas light mantles, but the gas mantles didn't gain much popularity until the 1890s. From the 1870s to the 1880s, electric lighting technologies advanced rapidly, including the invention of light bulb by Thomas Edison. The U.K. government passed the Electric Lighting Act in 1882 to promote electric lighting adoption. Ironically, the market share of electric lighting remained limited. In fact, the gas mantle lighting system started to take market share back from electric lighting due to electricity price fluctuations.

In the time around World War I, electric lighting started to gain favor due to shortages in coal gas and related price fluctuations. As the technologies for electricity generation and transport matured after World War II, electricity prices dropped significantly, and electric lighting dominated all market sectors as it still does today.

Table 11: Lighting system development path since the 1870s

Year	Event	References
Early 1870s	Introduction of the electric dynamo posed a competitive threat to kerosene and gas lighting.	[117]
1877	Carbon arc lamp was first used in Paris in May 1877, which triggered the interest of London.	[118]
	Dr. Carl Auer von Welsbach of Vienna invented the incandescent gas light mantle. Welsbach established the first factory to make gas mantles in London in 1877.	[119]
1878-1879	On December 14th 1878, the Holborn Viaduct electric lighting was switched on in London. Afterwards, four circuits each with four lamps were powered by a 20 horse power Robey steam engine drove a Gramme alternator and exciter. However, the experimental lighting was shut down on May 9th 1879. The city engineer reported that the cost was 3.75 times that of gas.	[118]
1879	Edison invented the long-life light bulb and applied for a patent.	[120][121]

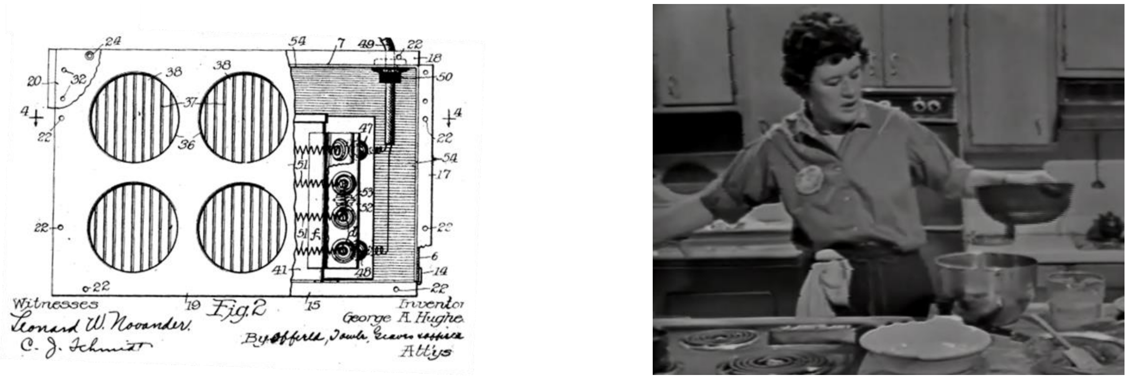
1882	Electric Lighting Act (U.K.) led to the establishment of electricity undertakings on a statutory basis, but the high cost of electricity hindered the wide application of electric lighting.	[122]
1890s	Gas mantle started to be widely used for streetlights.	[123]
1900s-1910s	By the early 1900s, the cost of electric lighting was getting closer to that of gas. However, only the wealthy could afford to wire their houses until the 1910s.	[11]
After 1910s	The World War I resulted in coal shortage and the coal gas quality fluctuation, which gave a chance for electric lighting popularization. After the World War II, the power generation technologies and electric grid were rapidly developed fast which brought down the electric lighting price, and the electric lighting found wide application not only for street lighting, but also in residential houses.	[124]

2.2.2 *Cooking, air/water heating and other applications*

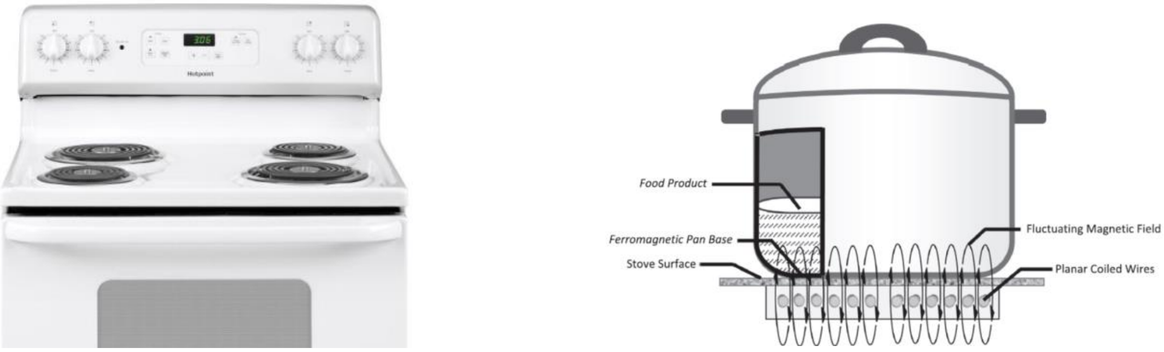
The majority of energy consumption in residential homes is associated with lighting and heating. Heating includes cooking, air (space) heating, water heating and other small applications like irons, dryers, kettles and so on.

It is believed that the first successful commercial cooking range was from the United States. In 1915, George A. Hughes from Iowa applied for a patent on his invention-an electric range with simple heating element wires set in clay bricks [125]. In 1918, Hughes merged his company with General Electric. In the 1920s, electric appliances started to appear in residential homes in both the U.S. and Europe, due to the thrust of appliance manufactures like General Electric, Westinghouse, and Siemens [126][127][128]. Figure 8 shows the evolution of the appearance of electric cooking ranges. Not much change is evident even over 100 years. When it was first developed (as shown in Figure 8 (a)), the electric wires were in parallel, then they were optimized into coil shapes to promote better contact and support to the cooking utensils. Figure 8 (b) shows an image from the TV show “The French Chef” with Julia Child cooking on electric stove tops in the early 1960s. Even today, the basic electric stove tops remain similar, as shown in Figure 8 (c).

In recent years, another type of electric cooking technology, induction, has gained popularity. Induction cooking technology is based on the principle of static magnetic fields. Alternating current flowing through an induction coil, which generates a magnetic field with changing polarities. This alternating field “induces” eddy currents that are trapped within the steel material that forms the base of the pan and transfers heat to the food it contains. The induction cooking surface provides a “clean” appearance and higher efficiency compared to traditional electric stoves. However, it is generally more expensive and also requires specific cooking pots and pans which are made of magnetizable materials.



(a) Cooking range invented by Hughes, 1915 [125] *(b) Julia Child cooking with electric range, 1963 [129]*

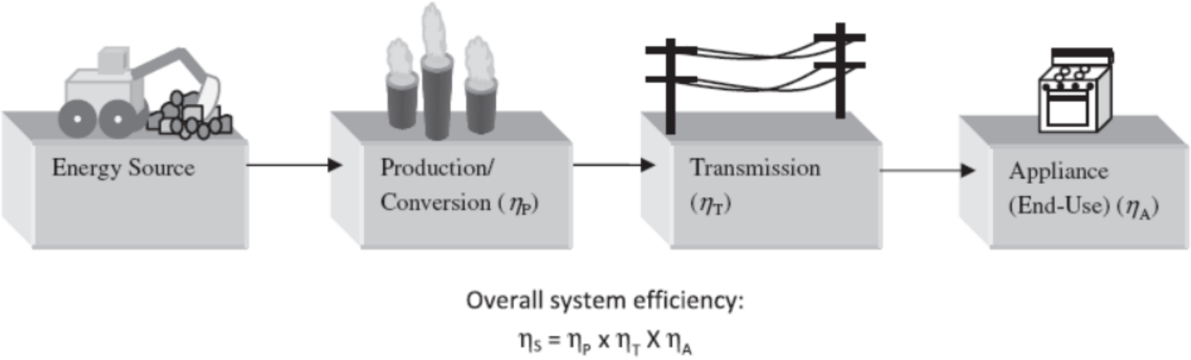


(c) Electric cooking range on market, 2020 *(d) Induction cooking schematic [130]*

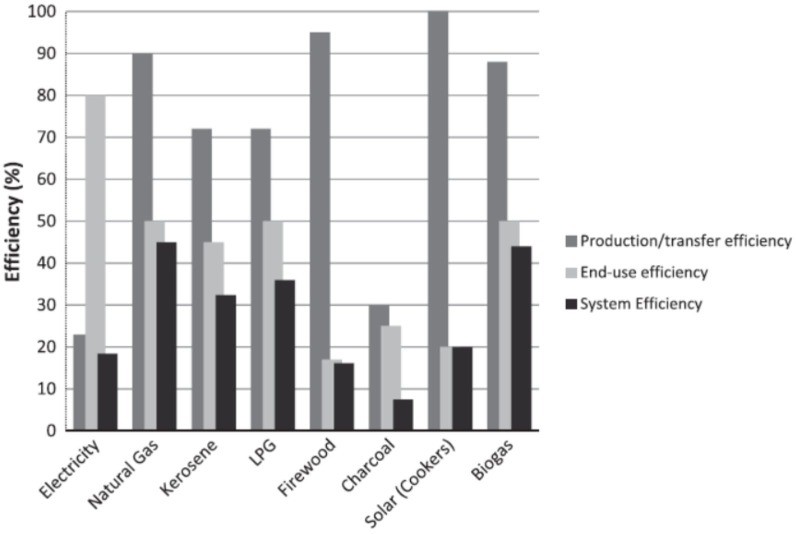
Figure 8: Historic appearance variation of electric cooking range

Compared to gas cooking appliances, electric cooking appliances usually have higher operating efficiencies. In the U.S., the average efficiency of gas stoves is around 40% which is significantly lower than the 74% efficiency of their electric competitors [131]. In Europe, the electrical cooking appliances have similar efficiencies as that of the U.S., but the gas cooking appliances have a higher range of efficiency from 52% up to 69% [130].

However, despite their early appearance in the market and evolution over 100 years, electric appliances do not dominate the current residential market. The high price of electric appliances and electricity itself is part of the reason. Also, when considering the “source to pot” efficiency, it is evident that many losses occur. As shown in Figure 9, the total system efficiency of utilizing electric appliances not only includes the appliance efficiency itself, but also includes the efficiency of electricity production and transmission process. Currently, most of the electricity in the world is generated from the fossil-fuel based power plants, whose highest efficiency is around 60% [132]. Further, according to the U.S. EPA, in 2019 the average efficiency of fossil-fueled power plants in the United States is only 33% [133]. Figure 9 (b) compares the efficiencies of cooking appliances using multiple energy sources. As shown, even if the end-use efficiency of electrical cooking appliances is assumed to be relatively high at 80%, the total efficiency is only around 18% which is much lower than the natural gas appliance efficiency of 45%.



(a) System efficiency considerations of utilizing electric appliances



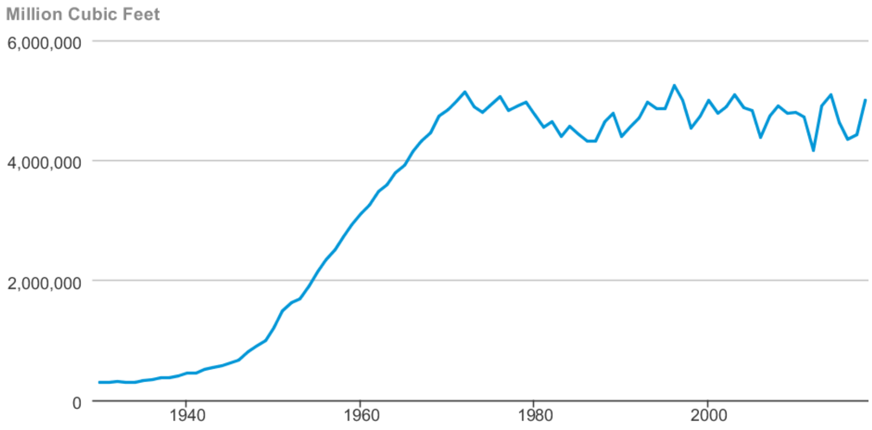
(b) Efficiency comparison among cooking appliances

Figure 9: Efficiency of residential cooking appliances using different energy sources [130]

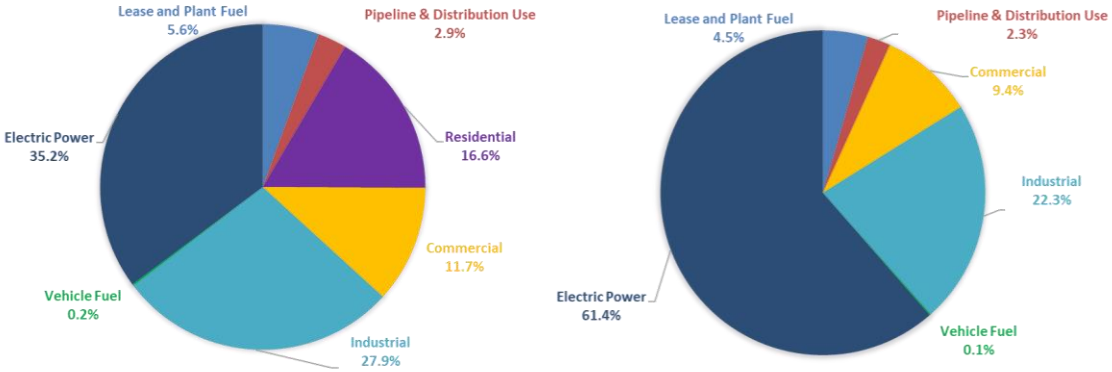
Figure 9 (b) shows efficiency comparison of different types of residential cooking appliances. Cooking applications exhibit smaller variations in system efficiency due to the large energy loss in the cooking process. However, when it comes to air heating and water heating appliances, the gas appliance total system efficiency will be even higher than their electric competitors. The operating efficiency of residential gas-fired air furnaces is typically greater than 80% [112][134]. Tankless water heater efficiencies are typically greater than 90% [110]. With the large gap between electrical appliances and gas appliances in the water heating and space heating applications, it does not seem likely that energy savings will result from electrifying residential homes.

One possible advantage of electrifying residential houses is shifting the emissions of greenhouse gases and other pollutants like CO/NO_X to the electricity generating sector, which is mainly power plants. It is relatively easy to control the emissions from a few power plants rather than from millions of households. However, as electrification increases, more demand for electricity will occur which will likely drive prices up. Further, with higher penetration of clean, but intermittent renewable energy sources like solar and wind, electrifying residential homes may add extra numbers of dispatchable fossil-fuel based power and/or expensive energy storage devices. This might increase the total greenhouse gas emissions, which is quite the opposite of what we intended to do. Combined with the decrease in electric appliance efficiency compared to gas fueled appliance, an even greater impact on the electrical system results again foreshadowing higher electricity prices.

Figure 10 (a) shows the U.S. natural gas consumption in the residential sector from 1930 to 2018. After 1970s, the natural gas consumption reached a plateau and has remained essentially level. As shown in Figure 10 (b), in 2018, about 16.6% of the natural gas was consumed in the residential sector which is the third largest percentage after power generation and industrial sector. In the near term, if renewable power supplies are not available, electrification of the residential sector will require this 16.6% to be provided by power generation. As shown in Figure 10 (c), this electrification will increase the total natural gas consumption by 25% and the natural gas consumption percentage in the electricity generation sector will increase from 35.2% to 61.4%, if not assisted with renewable electricity supply. This assumption is relatively conservative, since the efficiency difference of the cooking appliances in Figure 9 (b) is used to predict the system efficiency decrease in the residential sector. If considering the larger system efficiency gap of water heaters and air furnaces between using gas and electricity, even more natural gas would be consumed in the electric power generation sector.



(a) U.S. natural gas residential consumption from 1930 to 2018 [135]



(b) U.S. NG consumption in 2018 [135]

(c) U.S. NG consumption prediction

Figure 10: Natural gas consumption situation in the United States and future prediction

It is apparent that electrifying residential homes must be assisted with renewable energy sources. This is exacerbated by the fact that current renewable electricity availability is far less than that needed to replace the fossil fuel power supply. Governments around the world are trying to establish policies to increase the percentage of renewable electricity generation. The State of California in the United States is one of the most ambitious regions in the world promoting renewable energy and reducing carbon emissions. In September 2018, California passed Senate Bill 100 setting a world-leading precedent by committing to 100% renewable electricity by 2045 [136]. Together with the renewable electricity goal, California also aims to electrify home appliances [137][137][138][139]. Yet, even with these actions, a significant time lag in available renewable electricity resources is likely which warrants an approach with diverse sources in the near term.

The challenge for this transition is significant. Figure 11 shows the natural gas consumption by different appliances in residential homes of California (U.S.), which reveals that water heating, air (space) heating and cooking use by far the most natural gas [140]. The column charts show the energy sources for these three sectors in representative large cities of California. More than 60% of the households in California use natural gas to cook and more than 80% of the households use natural gas to heat water and space [141]. Based on the current status, electrifying California residential homes will face great challenges. The increased demand for renewable electricity will likely increase utility costs for consumers which may hinder the progress of electrifying residential homes. It is estimated by the California Building Industry Association (CBIA) that it could cost 1) $7,200 per household to upgrade wiring and electrical panels to purchase new appliances, 2) $877 higher annual utility costs per household, and 3) an overall cost increase of $4.3 to $6.1 billion per year in California [142]. It should be noted that when the U.K. was undergoing the town gas to natural gas transition, the cost of replacing appliances across the country was covered by the government owned British Gas Council. For countries with a free market today, the cost of converting gas appliances to more expensive electrical appliances will be less likely covered by electricity utility companies. No matter how these costs are paid, the money will be eventually from California taxpayers. For example, since solar panel adoption in California started, concerns have been expressed from disadvantaged families about rent or utility increases due to the construction of renewable energy infrastructures. Therefore, in this appliance transition process, low-income communities or disadvantaged households should be given extra attention and care.

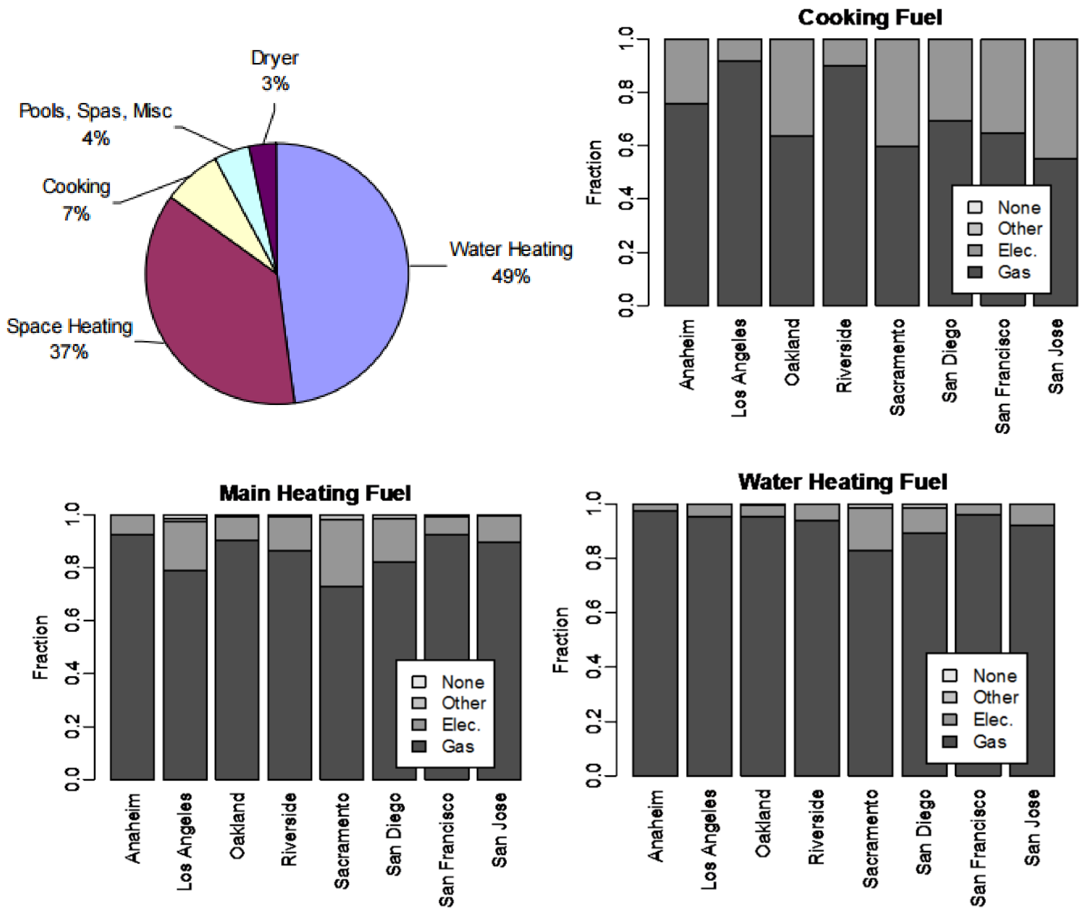


Figure 11: Energy consumption status by residential appliances in California, United States [140][141]

The discussions above focused on energy consumption and cost concerns. But another important challenge is people's acceptance of using electric over gas appliances. If not considering possible higher electricity bills, people may have less concern of adopting electric appliances for heating air and water because they can't feel the difference between gas and electrical appliances just from the heated air or water. However, when it comes to cooking, a clear preference of gas over electricity is evident. In 2014, a survey was conducted among 100 professional chefs across the United States, and 96 reported they prefer using gas burners. The reported advantages of using gas stoves include fast response speed and controllability, high adaptability of cookware, simple to clean and inexpensive maintenance [143]. Advantages of gas over electric cooking appliances have been highlighted since the introduction of electric cookstoves 100 years ago. Given the limited advances in either gas or electrical cooking appliances over the past decades, sentiment regarding the preferred fuel has not changed. Even though Julia Child used electric stoves on the TV show in the 1960s, it still didn’t popularize them enough to displace gas ranges. It is also interesting to note that while Julia Child used electric appliances on TV, she actually had a gas stove in her own house [144]. The reason she used the electric stove on TV was more of a sponsor issue. When WGBH planned to shoot the scene at the demonstration kitchen of the Boston Gas Company, they learned that the kitchen was dismantled in 1962. As a result, the Cambridge Electric Light Company offered to provide their electrical demonstration kitchen for the “The French Chef” [145]. Moreover, General Electric was the co-sponsor for this show and provided the electrical stove. Besides her personal use of a gas range, in published letters to her book editor and friend Avis Devoto, it was also clear that Julia Child strongly preferred cooking with gas [146].

2.2.3 *Conclusion*

It took less than 40 years for the electricity taking over the residential lighting, since the first arc lamp was lighted in 1877 on the street of Paris. However, more than 100 years have passed since electric appliances for cooking and heating were first introduced and they remain a small portion of the residential appliances market. This might be caused by the intrinsic disadvantage of electric appliances for cooking and heating: people don’t necessarily need electricity to heat food, air and water, it is just the heat that people really need. For the lighting systems, electricity has its intrinsic advantage of converting to light with high efficiency. However, combustion converts most of the energy in the fuel into heat, which is what needed for cooking and domestic heating. If the heat needs to convert first into electricity in power plants and the output electricity is transported into residential homes and is converted to heat again, the exergy loss in this process is significant.

Currently, the major consideration of electrifying residential homes is to reduce emissions from the residential sector and allocate the emissions into the power generation sector. As discussed above, due to the efficiency loss in this appliance conversion process, there might be an increase in greenhouse gas emissions if this conversion is not assisted with high penetration of renewable electricity from solar, wind or hydro power. This renewable electricity adoption might be practical for cities that have access to renewable energy sources. California, for example, has implemented policies to reduce carbon emissions by requiring solar panels be added to any new residence constructed from 2020 onward [147]. However, this policy might not be practical for cities like London or Seattle where solar energy is much less abundant as California. Even if London can utilize the electricity from offshore wind farms, energy loss in the electricity transport through grid will occur.

Therefore, instead of electrifying all residential homes, another strategy has already been in commission: replacing pipeline natural gas with renewable gases including biogas and hydrogen. This method of adopting renewable energy needs much less investment for replacing appliances and wiring adaptation for electrical appliances and takes advantage of existing infrastructure widely prevalent in most developed regions.

Table 12: Various aspects comparison of electrification vs using renewable fuels

	Electrifying residential homes	Adopting renewable gases in pipelines
System efficiency	System efficiency decreases due to the energy loss in the electricity generation process.	The efficiency should not change significantly
Carbon reduction	Shifting the carbon emissions to power plants may increase total carbon emissions if not assisted with renewable energy sources.	Carbon emissions will drop in proportion to the amount of biomethane or renewable hydrogen adopted
Combustion pollutants (CO/NO_X, etc.)	Shifts pressure to power plants to reduce emissions.	Pollutants don’t change much [106]-[112].
Appliance upgrade	Need to replace all the gas appliances with electrical appliances, together with upgrading the house wiring system.	Don’t need appliance upgrade for biomethane adoption. Need studies on hydrogen fuel interchangeability.

Cost	Electrical appliances are usually of a higher price and electricity price is more expensive than gas.	Gas price might increase due to limitation of renewable gas source availability.
Technology availability	Technology available.	Technology available.
Energy Security	Might be vulnerable when blackout occurs due to a single type of energy source.	Energy system is more diverse.
People's acceptance level	People prefer gas cooking appliances	People are more used to gas appliances for cooking and air/water heating.

A full energy transition from gas to electricity in the residential sector remains a current area of debate. Regardless if it is ultimately achieved, residential gas pipelines should be and already are adopting renewable fuels.

2.3 Gas Appliances Combustion Performance Evaluation Methods

There are multiple combustion performances that are considered while evaluating residential appliance performances, including efficiency, emissions, etc..

2.3.1 *Efficiency*

Efficiency, which is directly related to utility bill, is one of the primary considerations for a residential householder to choose an appliance. Therefore, improving efficiency has always been a motive for appliance manufactures to upgrade their products. To guide the residential sector to higher energy efficiency level, regulations are also being improved to promote this process world widely [148]. Among all the energy efficiency promoting programs, the most influential one might be the ENERGY STAR program executed by the U.S. Environmental Protection Agency and the U.S. Department of Energy in 1992 [149]. Over the past few years, this program has been adopted world widely, including European Union, Japan, Australia, etc..

For gas appliances, the efficiency of an appliance is closely related but not equal to the burner combustion efficiency. The combustion efficiency of a burner is the ratio of total heat release over the total chemical energy in the fuel. Therefore, the major combustion efficiency loss is from the unburned hydrocarbons or carbon monoxide emissions. In modern residential appliances, these emissions are mostly at ppm level which makes the combustion efficiency close to 100%. Therefore, the appliance efficiency loss is most likely from aspects regarding the appliance inner structure design, including burner location, heat exchanger arrangement, etc..

It should also be noted that a high appliance efficiency can also be coupled with a low combustion efficiency, which might increase the emission levels. For example, in a cooktop burner support rack optimization process, it is found that lower rack can shorten the distance between the flame and the cooking utensil which helps reduce radiation loss to the environment thus increase the efficiency. However, this also results in flame impingement onto the cooking utensil surface and can bring up the carbon monoxide emission level and also degrade the cooking utensil life span [150]. Therefore, improving appliance efficiency is a task involves numerous aspects, sometimes a compromised efficiency level should be adopted to optimize other appliance performance.

Among various residential appliances, water heating and air heating efficiency is relatively easy to measure using energy output/input method. Most of the existing room furnaces can achieve an energy efficiency of 80% easily and water heater efficiency can be higher than 90% with a latent heat recovering (water condensing) system [110][112].

Compared to the mature efficiency testing method for air/water heating appliances, there is yet no universal efficiency testing standard for cooking appliances. Differing from air/water heating, heating the food to a certain temperature is not the only purpose of cooking, so it is relatively hard to define a perfect cooking process. However, efforts are still being made around the world to quantify the cooking efficiency thus help improve the cooking appliance performance. As shown in Table 13, the most common ways to evaluate the cooking efficiency are food cooking, water boiling and object heating. Cooking a specific food is an intuitive way to evaluate the cooking performance. For example, Oberascher [151] used boiling eggs and cooking potatoes to evaluate the cooking performance of difference cooking appliances. To evaluate the cooking results, the egg yolk status was scaled to five levels: very liquid, liquid, firm outer edge, can be separated from egg white, firm and dry. The egg yolk status was also coupled with the yolk color judgement to distinguish the cooking efficiency of different appliances. Cheng [152] conducted experiments on meat and used meat moisture loss rate as one of the cooking efficiency measurement methods. Food cooking method is effective to evaluate the cooking performance at one location and one time, but it is relatively hard to repeat the results for other testing locations or even the same location in different times due to the tested food quality variation. Moreover, it is also hard to generate a universal quantified standard to evaluate the cooked food characteristics related to cooking efficiency.

Therefore, water boiling test is more widely adopted to evaluate the cooking appliance efficiency. To avoid the influence of cooking utensils on the cooking efficiency, American National Standards Institute (ANSI) regulated the dimension of the water boiling pot and testing method [153]. Although boiling water deviates from the actual cooking process, it is relatively easy to quantify the cooking efficiency using Equation (1). The efficiency is defined as the heat absorbed by water (including the latent heated from escaped water vapor) over the total heat release from the fuel. Most cooking appliances can’t recover heat from the water vapor in the exhaust, therefore, lower heating value is usually adopted to calculate

$$\eta_{cooking} = \frac{c_{p,water} * m_{water} * \Delta T + h_{vapor} * \Delta m_{vapor}}{V_{fuel} * LHV} \qquad \text{Equation (1)}$$

Heating intermediate object, instead of food or water, is also a method to evaluate the cooking efficiency by measuring the object temperature variance over time. The material should be able to resist high temperature and have high thermal conductivity, therefore, metals like steel or aluminum are usually adopted [158][159]. This method requires the temperature of the intermediate object being relatively uniform over time, so it is more commonly adopted in oven testing.

Table 13: Cooking appliance efficiency evaluation methods

Name	Method	Reference
Food cooking	Cooking a specific food (egg, etc.) and evaluating the food appearance to interpret efficiency	[151][152]
Water boiling	Boiling water and calculate the ratio of heat absorbed by water over the total heat release rate	[106][107][153][154][155][156][157]
Object heating	Heating some materials (usually metal rod/disk) and measure the temperature change of the material to quantify the efficiency	[158][159]

It should be aware that the cooking speed and cooking efficiency are different from each other when evaluating cooking performance of different fuel classes. If the heat release rate is constant for two testing conditions, faster cooking time can be regarded as higher efficiency. However, when the fuel classes have different heating value or Wobbe Index, less cooking time can also be resulted from higher heating load of the burner. In this case, faster cooking, which might result in food overburn, may become a non-ideal situation. For example, Zhao et al. [106] evaluated the cooking efficiency of a cooktop burner operating on natural gas and simulated biogas. The results show that the cooking time increased as natural gas was replaced by biogas due to the heating load decrease. However, the cooking efficiency stayed at a constant level. The cooking performance of a cooktop burner operating on natural gas/hydrogen mixtures was also tested, it was found that both the cooking time and efficiency didn’t change much up to 50 vol% hydrogen added into natural gas [107].

2.3.2 *Emissions*

Besides increasing efficiency, reducing emissions from appliances is also one of the major incentives for appliance manufactures due to the stricter emission regulation worldwide. In the past few decades, combustion technologies were advanced to reduce emissions mainly in power generation, transportation and industry. However, the emissions from the residential sector didn't draw as much attention as other energy utilization sectors. Residential appliances are closely related to our daily life, so the emissions from them are not only doing harm to the environment but might also threaten our health. Therefore, the existing emission regulations on appliances and the current emission levels of existing appliances should be understood to project a bigger picture for the future.

2.3.2.1 Residential appliance emission regulations

The emission regulations on power generation, transportation and industrial applications are frequently updated and widely adopted in the world nowadays. These regulations also play a significant role on motivating combustion technology advancement, which experience should be learned by the residential sector.

One of the most active agencies promoting appliances standards are American National Standards Institute (ANSI) and Canadian Standards Association (CSA). They update the ANSI-CSA standards on appliance performance every few years. Nowadays, ANSI-CSA standards are not only adopted in North America, but also being learned and adopted by more countries in the world. However, the ANSI-CAS standards only cover the carbon monoxide emission considering the safety reason, while lacking the regulation on other emissions like nitrogen oxides. Therefore, ANSI-CSA standards have to be coupled with other standards to form a more comprehensive emission regulation series, especially the ones regulating on nitrogen oxides emissions. One of the most pro-active regions in regulating NO_X emissions and promoting new combustion pollutants control technologies is California (U.S.). To deal with the Los Angeles smog hazard caused by NO_X-O_3 photochemical reactions, several environment protection and emission regulation promoting agencies were founded, after the U.S. Congress passed the Clean Air Act in 1963. These agencies include U.S. Environmental Protection Agency (EPA, 1970) promoting federal regulations, California Air Resources Board (CARB, 1967) regulating on vehicle emissions, South Coast Air Quality Management District (SCAQMD, 1976) issuing emission standards on standard sources of air pollution. Among these agencies, SCAQMD issues emission regulations on multiple sectors including the industrial and residential applications.

As shown, Table 14 lists the existing emission regulations on residential appliances in North America, which is also considered as one the of the strictest regulation series in the world. It should be noted that most of these regulations are only adopted in very few places around the world. For example, some of the NO_X emission regulations from SCAQMD are not even extensively adopted in California itself. However, these regulations should project a future view on our future appliance emission level around the world.

Table 14: Representative emission regulations for residential appliances in North America

	Regulation title	Agency	Adopted time	Major contents
Water heating appliances	ANSI Z21.10.1 [160] ANSI Z21.10.3 [161]	ANSI-CSA	2017	CO < 800 ppm (heating load ≤75,000 Btu/hr) CO < 400 ppm (heating load >75,000 Btu/hr)
	Rule 69.5.1 [162]	SDCAPCD	2017	NO_X < 10 ng/J (calculated as NO_2) or NO_X < 15 ppm (@ 3% O_2, dry)
	Rule 1146.2 [163]	SCAQMD	2018	NO_X < 14 ng/J (calculated as NO_2) or NO_X < 20 ppm (@ 3% O_2, dry)
Space heating appliances	HSC-1988 [164]	California Law	1997	No person shall sell, or offer for sale, any new or used unvented heater that is designed to be used inside any dwelling house or unit, with the exception of an electric heater, or decorative gas logs for use in a vented fireplace.
	ANSI Z21.86 [165]	ANSI-CSA	2016	CO: less than 200 ppm in the air free sample
	Rule 4905 [166]	SJVAPCD	2018	NO_X: 14 ng of oxides of nitrogen (calculated as NO_2) per joule of useful heat delivered to the heated space
	Rule 1111 [167]	SCAQMD	2018	NO_X: 14 ng of oxides of nitrogen (calculated as NO_2) per joule of useful heat delivered to the heated space

Cooking appliances	Rule 1153 [168]	SCAQMD	1995	VOC of commercial bakery ovens (≥ 2 million Btu/hr) should be less than 50 pounds/day
	Rule 1138 [169]	SCAQMD	1997	Chain-driven charbroiler must be equipped with catalytic oxidizer in reducing PM and VOC
	Rule 1131[170]	SCAQMD	2003	The VOC content of each solvent used ≤ 120 grams per liter of material
	Rule 1153.1 [171]	SCAQMD	2014	Commercial ovens: CO:800 ppm (@3% O_2) NO_X:40 ppm (@3% O_2)-500 °F-60 ppm (@3% O_2)
	ANSI Z21.1 [153]	ANSI-CSA	2016	CO: cooking appliances less than 800 ppm
	Rule 4692 [172]	SJVUAPCD	2018	In lieu of SCAQMD-Rule 1138. The catalytic oxidizer shall have a control efficiency ≥ 83% for PM-10 emissions and a control efficiency ≥ 86% for VOC emissions

For residential water heating appliances, ANSI-CSA regulates that CO should be lower than 800 ppm in an undiluted exhaust sample for appliances with the heating load not higher than 75,000 Btu/hr (~22 kW) [160]. For the water heaters exceeding that threshold, the CO emission should be lower than 400 ppm [161]. The most widely adopted NO_X standard in the U.S. is the SCAQMD-14 ng/J emission regulation [163]. The water heaters adopted this regulation are usually labeled as low NO_X water heaters on market. The ultra-low NO_X 10 ng/J standard is from San Diego County Air Pollution Control District (SDCAPCD) [162], which is being adopted by more and more residential water heater manufactures.

Space heating is also a major emission contributor from residential houses. In 1997, California banned the sale of ventless space heaters in residential homes. For current vented heaters being sold or installed in California, the CO emission should be lower than 200 ppm [165]. Besides, the NO_X emission standard becomes stricter from 40 ng/J to 14 ng/J, and penalty should be paid by the manufactures if not meeting this new standard by the year 2021[112][166][167].

Currently, the major emission regulations on cooking appliances are on mainly on VOC or particle emissions from the food, which requires ventilation in the kitchen. The threatening emissions to human health from kitchens are mainly particle emissions instead of emissions from kitchen flames [173][174]. However, residential houses with bad ventilation might have CO and NO_X accumulation in the living space which will bring health issues. Even if the direct influence of residential flame emissions on human health is negligible compared to particle emission from food itself, reducing kitchen flame emissions should also be a task to reduce the total emissions into the atmosphere. Due to the uncertainness of the kitchen burner operating conditions, ANSI-CSA imposes a relatively “generous” restriction on CO emission (800 ppm) compared to water and space heaters. Although there are no direct NO_X emission regulations on residential cooking appliances, SCAQMD does restrict the NO_X emission of commercial ovens being lower than 40 ppm at an operating temperature lower than 500 °F. With the oven temperature exceeding 500 °F, NO_X emission limit is extended to 60 ppm.

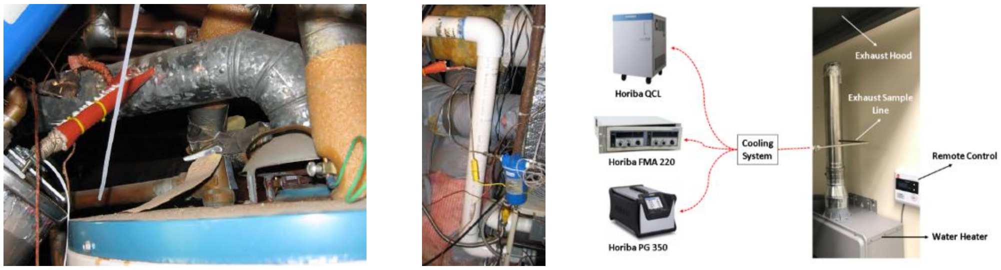
It should be noted that the emission regulations in Table 14 are mainly for gaseous fuels. There are still a lot of developing countries even some areas in developed countries using wood, coal or biomass as residential heating/cooking energy sources. For these areas, the particle emissions might be a larger or more direct threat to human health.

2.3.2.2 Emission evaluation methods conclusion

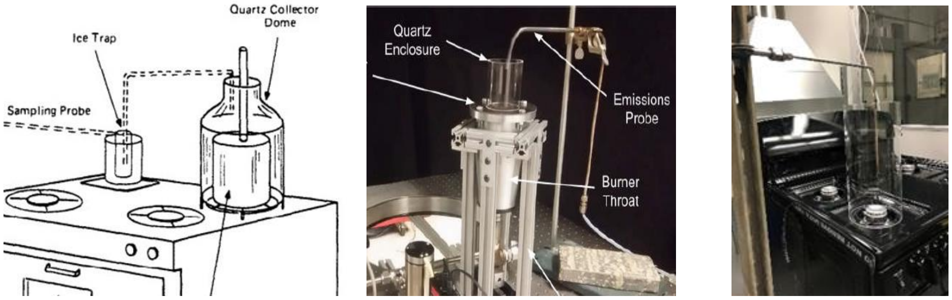
As can be seen in Table 14, there are multiple ways to interpret emission levels of an appliance: volume percentage in an undiluted exhaust sample (ppm), volume percentage at a corrected oxygen concentration (ppm @ 3% O_2), emission level per unit energy output (ng/J), etc.. This shows the emission level interpretation of residential appliances is still lacking universal standards. In fact, not only the expressions of residential appliances are hard to achieve a consensus, there are also various sorts of emission testing methods.

As shown in Figure 12 (a), the secured exhaust sampling is an ideal way to sample the emissions from an appliance exhaust venting location. This is an effective to avoid air dilution in the exhaust. However, this method is only practical for confined combustion appliances like room furnaces or water heaters. Before all the exhaust is vented into the atmosphere, it will first be directed into a pipe, which the sampling probe can be located at.

However, a large number of residential burners have open-air flames, which means combustion takes place in an open space and dilution from the surrounding air is inevitable. Therefore, quartz enclosure and hood sampling method are usually adopted as shown in Figure 12 (b) and (c) respectively. The quartz enclosure sampling method is more suitable for small size burners. When adopting this method, it is necessary to make sure that the sampled mixture is homogeneous. If not, multiple locations within the quartz enclosure should be tested over a certain time period and the average should be calculated to present the emission level of a burner. The exhaust hood sampling method is the one more commonly adopted for kitchen burners. The exhaust hood collects all the emissions before venting it into the atmosphere, before the sample is taken from the hood, the exhaust mixture is already homogeneous. What calls for special attention is the distance between the hood and the burner. The hood should be close enough to the burner, so all the exhaust can be collected, or the tested emission level will be lower than the actual level. However, if the hood is too close to the flame, it might cut off the source of the oxidants (air) to the flame and results in carbon monoxide increase in the exhaust. ANSI Z21.1 recommends this gap should be around 5 inches (12.7 cm).



(a) Secured sampling [175][110]



(b) Quartz enclosure sampling [176][177][106]

(c) Exhaust hood sampling [178][175][107]

Figure 12: Emission testing methods of residential appliances

As shown above, the emission tests on residential burners are sometimes naturally diluted. Even for confined combustion like water heater and room furnace burners, the secondary air from atmosphere also dilutes the emission to a lower absolute level. Therefore, the emissions are usually corrected under a certain reference level so they can be compared with each other.

2.3.3 *Flame characteristics*

American Gas Association (AGA) has its recommendations on residential flame characteristics, which are widely adopted in the world. As shown in Table 15, most flames of residential appliances should have a relatively small absolute code value. The flame cones are usually in blue color and have laminar flame characteristics. However, there are also exceptions. Since stable laminar flames are more likely having a low flowrate, they are more for lower heating load appliances. Higher heating load appliances, like room furnace, laundry dryer, etc., tend to have turbulent flames. Moreover, not all flames are supposed to have blue color. Gas fireplaces need to generate soot to promote radiation, therefore, the flames are usually yellowish or orangish. Although yellow tip index was developed to guide fuel selection avoiding yellow tips, outdoor grillers are allowed to have yellow tips which help increase the heat transfer rate to the food.

Table 15: AGA flame code classifications

Code	Flame Description
+5	Flames lifting from ports with no flame on 25% or more of the ports.
+4	Flames tend to lift from ports, but become stable after short period of operation.
+3	Short inner cone, flames may be noisy.
+2	Inner cones distinct and pointed.
+1	Inner cones and tips distinct.
0	Inner cones rounded, soft tips.
-1	Inner cones visible, very soft tips.
-2	Faint inner cones.
-3	Inner cones broken at top, lazy wavering flames
-4	Slight yellow streaming in the outer mantles, or yellow fringes on tops of inner cones. Flames deposit no soot on impingement.
-5	Distinct yellow in outer mantles or large volumes of luminous yellow tips on inner cones. Flames deposit soot on impingement.

When replacing natural gas with renewable gases, the flame characteristics should be reevaluated. For example, hydrogen flame is invisible under sunlight, which might limit its use in existing gas fireplaces or outdoor grillers. The light flame color of hydrogen-rich fuel might also cause danger if an existing flame can’t be identified. However, researchers around the world also find random reddish glow in hydrogen flames. It is believed the color is from the contaminant in hydrogen, which might be related to hydrogen embrittlement effect on metals [108].

2.3.4 *Ignition performance*

Appliance ignition is also a crucial aspect to evaluate the performance of an appliance. Failure of ignition can result in natural gas leakage into the residential homes which endangers the safety of residents. Moreover, long ignition time also results in methane leakage into the atmosphere for natural gas appliances. The greenhouse gas effect of methane is about 20 times that of carbon dioxide, therefore, the methane emissions from a delayed ignition also contribute to the greenhouse effect [180][181]. Some regulations set an upper limit for appliance ignition time. For example, ANSI Z21.1 [153] requires residential cooking appliances should finish the ignition within 4 seconds. Higher heating load appliances even needs stricter ignition time limit. ANSI Z21.86 [165] regulates that the room furnace ignition should be completed within 0.8 second.

Ignition is an unsteady process, which might become a limiting factor for fuel interchangeability studies. For example, Zhao et al. [107] replaced more than 75% natural gas with hydrogen without having flashback in a cooktop burner under steady operating condition. However, the ignition flashback occurs at only 20% hydrogen addition. This is because at steady operating condition, the mixture within the burner is usually fuel-rich and out of the flammability range. At ignition condition, when the fuel started to fill the burner, the existing air within the burner makes the mixture fall into the flammability range. This will cause ignition flashback.

Therefore, ignition performance should draw special attention in the burner design or residential appliance regulations.

2.4 Remaining Questions

Under this upcoming (already taking places in some countries) energy transition from traditional carbon-based fossil fuels to low carbon renewable fuels, various challenges need to be dealt with.

The residential sector has witnessed several energy transitions in the past few decades from solid fuel (crop residuals, fuelwood, coal, etc.) to fuel gases (manufactured gas, natural gas, etc.). Replacing pipeline natural gas with renewable gases (biogas, hydrogen) in residential applications is not an entirely groundbreaking technical revolution, since we once had more than 50 vol% hydrogen in our residential pipeline in the manufactured gas era. Technical considerations regarding hydrogen compression, leakage, pipeline material, etc. can draw lessons from the manufactured gas era. To increase the hydrogen tolerance level of current natural gas appliances, insight can also be obtained from the designs of manufactured gas burners. However, it should be noted that, since the manufactured gas era, modern residential burner designs have more requirements. Besides safety concerns, other requirements like low emissions, high efficiency, ease of operability, longer lifetime period, and visual appeal must be considered in modern residential appliances design. As a result, while “reversing” the transition from manufactured gas to natural gas can give guidance, shifting from natural gas to natural gas/hydrogen mixtures and perhaps ultimately 100% renewable hydrogen will require more effort.

Open questions regarding the fuel interchangeability from natural gas to renewables gases in residential self-aspirating flames include:

- What are the differences and similarities among modern gas appliances and what are their improvements compared to the manufactured gas era?
- What are the CO, UHCs, NO, NO_2, N_2O, NH_3 emission levels of current residential appliances? Is there a need for emission regulation update on residential appliances?
- Can the natural gas/propane gas appliances tolerate renewable gas contents like biogas or hydrogen? If so, what are the operating limits?

- What is the influence of renewable gas on the burner performances: ignition behavior, flashback/blow-off limit, efficiency, emissions, combustion noise, burner temperature, etc.?
- Which numerical strategy & reaction mechanism can more accurately predict the emissions and stability of residential flames?
- The air entrainment is critical to the combustion performance of self-aspirating flames. Can this behavior be predicted?
- The hydrogen flame color has been reported colorlessly or pale blue. However, there are numerous studies reporting reddish glow in hydrogen flames. What is the source of this random reddish color of hydrogen flames?
- If natural gas is to be partially replaced by renewable hydrogen, what gas quality regulations needs to be altered? What would the emission levels be like between appliances electrification vs renewable gas combustion?

The above questions are discussed and addressed in this dissertation. The approaches adopted under different tasks are outlined in Chapter 3. The results and analysis are given in Chapter 4&5. The final answers to these questions will be summarized in Chapter 6.

CHAPTER 3: Approach

To assist the energy transition from natural gas to low-carbon renewable gases in residential self-aspirating flames, this dissertation uses both experimental and numerical methods to address the knowledge gap bedded in the open questions.

In light of the existing questions, this dissertation established detailed approaches for addressing each task is described as following:

Task 1. Investigate residential burner configurations and working principles.

This task investigates the geometry of the residential gas burners, thus clarifies the mechanism of self-aspirating flames adopted in these burners. Various burner types bedded in residential appliances are studied under this task, including cooktop, oven, water heater, room furnace, etc..

Multiple burners from each appliance kind are examined to compare similarities and differences. Representative burners are selected and tested to further verify the working principle analysis. Preliminary fuel flexibility analysis is conducted to forecast the challenges in the experiment. Task 1 is the foundation of the work in this dissertation, which helps generate the Standard Operating Procedure of the experiments testing different perspectives of burner performance and also helps avoid accidents when hydrogen flashback occurs.

Task 2. Fabricate the fuel mixing apparatus and establish fuel mixtures of interests.

Under this task, a fuel mixing station is fabricated to generate fuel mixtures of different flowrates and species percentages. Different burners possess various heating load ratings, which require the flowrate varying for different operating conditions. Besides generating mixtures of multiple kinds, the mixing station should also be able to help achieve homogeneous mixing effect while regulating the fuel gas pressure to the burner operating requirement.

Carbon dioxide and hydrogen are mixed with natural gas separately to simulate biogas and renewable hydrogen blending into natural gas infrastructures. Before the gas mixtures are supplied to different burners, the pressure is regulated to around 8 inches of water (~2000 Pa) to mimic the household natural gas pipeline pressure.

Task 3. Test the fuel flexibility of selective residential burners.

Task 4 requires setting up a platform to test various appliance burners operating on natural gas/renewable gas mixtures. The installation uses a series of American National Standards Institute standards as reference to ensure the appliance testing is safe and similar to the real life operating conditions.

Combustion performances are monitored to investigate the influence of renewable gas on the self-aspirating flames. The percentage of renewable gas goes beyond the current gas quality regulations and up to the highest possible level until the combustion fails. Therefore, the limiting performance factors are obtained for the benefit of future burner optimization to incorporate more renewable gas contents.

Task 4. Establish numerical models to predict the emissions of self-aspirating flames.

Numerical simulation of self-aspirating flames is conducted using ANSYS-Fluent. CFD-CRN modeling is applied to several burners that are tested experimentally. The boundary conditions of the simulation are from the experiment data. The procedures of the numerical simulation are:

1. Build the numerical model by deriving the information from the physical burner geometry.

2. Setting the boundary conditions by obtaining the fuel flow data and the exhaust contents analysis.

3. Compare the simulation results with the existing experiment data in terms of emissions and flame extinction limits.

Task 5. Provide reference for fuel flexible burner design and future regulations.

The combustion characteristics of self-aspirating burners operating on natural gas/renewable gas mixtures and related analysis can guide the burner optimization to incorporate more renewable gases in natural gas. Currently, the number of regulations on residential self-aspirating combustion devices is limited compared to those on large energy consumption combustion units. The comprehensive testing in this dissertation can serve as a reference for future regulations from performance perspectives, including emissions, efficiency, etc.. The fuel property analysis will also help pave the way for gas quality regulation updates for adopting more renewable gases in existing natural gas infrastructures.

CHAPTER 4: Fuel Interchangeability Technical Considerations

4.1 Residential Self-aspirating Burner Configurations

Residential burners are usually mounted in domestic appliances. Compared to higher heating load gas burners designed for turbines or industrial applications, residential burners usually adopt simple designs which are easily manufactured, maintained or replaced if damaged. Lacking complicated fuel/air flow control systems, most residential burners adopt self-adjusting fuel/air mixing technologies. The most common if not the only burner type in residential appliances is self-aspirating or called aspirated burner, whose flame type is usually partially premixed. This combustion technology was first introduced into application in the 1850s by the German chemist Robert Wilhelm Bunsen, who invented the later called Bunsen burner and influenced the combustion applications and the gas industry significantly [42].

Figure 13 (a) shows a model of Bunsen burner. It is an atmospheric burner with a jet of gas aspirating part, and the air can be entrained through adjustable shutters into a fuel/air mixing tube before reaching the burner port for combustion. Due to the pre-mixing of fuel and air before combustion, the soot formation and carbon monoxide emissions are diminished significantly and the flame length or position is more controllable by adjusting the air shutter. Therefore, the Bunsen burner gained popularity not soon after it was introduced to the public, and was modified into various applications. Figure 13 (b) shows a domestic aspirating burner for cooking and water heating in the late 19th century. As shown, the fuel is supplied to the burner through a valve at the bottom and then entrains some air through the small circle openings on the burner surface before reaching the flame zone. Till today, self-aspirating burners are still dominant in residential appliances. Figure 13 (c) shows a representative cooktop burner that is available on market today. As shown, fuel is injected into the burner head while entraining a small amount of surrounding air.

Part of the mixture flows out of the ignition ports and arrive at the ignitor through the ignition tube. After ignition, the flame propagates to the burner head and a full flame will be established. Most of appliances on market are using self-aspirating technology. Their flames appear blue color due to its relatively low equivalence ratio at operating conditions, which helps reduce soot, unburned hydrocarbons and carbon monoxide emissions. However, due to the difference of heating purposes, some appliances might glow in yellow/red color. For example, outdoor grillers usually have yellow tips to enhance the heat transfer in grilling. Gas fireplace flame appearance is close to diffusion flame, because it needs the soot radiation to heat the surrounding space.

Due to the simple burner design and outstanding combustion performance, self-aspirating combustion is not only the most common combustion technique in residential homes, but also laboratories and light industry.

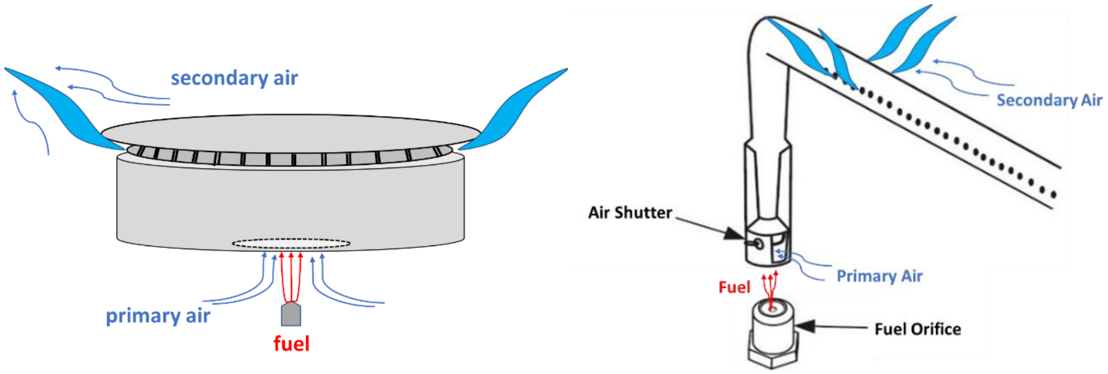
(a) Bunsen burner *(b) Domestic burner from the 19th century [40]* *(c) Current cooktop burner*

Figure 13: Domestic burner involvement

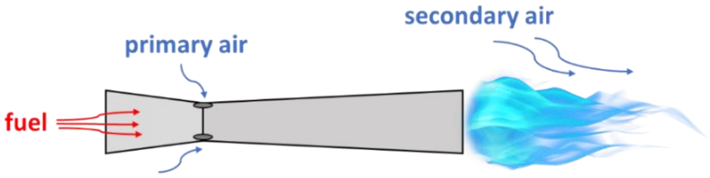
Figure 14 shows the most common residential gas burners on market, which are mainly from three sections: cooking, air heating and water heating. The cooking burners include cooktop burners, oven burners, broilers, grill burners. Central air furnace is one of the most common air heating devices in residential buildings. Gas fireplace is more common for outdoor heating and is getting depleted for indoor air heating due to its high emissions and inefficiency of heating a large space. Ventless space heating is another air heating method that is banned by more regions, due to the direct vent of exhaust (NO_X, CO, etc.) causing indoor air quality deterioration. Other air heating devices might have more indirect purposes than solely heating the air like laundry dryers. Water heating is another large energy consuming sector. Storage water heaters and tankless water hears are two common residential water heating applications. Although pool heaters have a lower total energy consumption due to the low ownership ratio per household, but it usually has a high energy consumption rate per unit.

Figure 14: Representative residential appliance burners on market

Although these residential burners adopt the same combustion technology, their appearances might still be distinct from each other. Figure 15 shows the most common self-aspirating burner types in residential burners. Figure 15 (a) shows the configuration of a flat self-aspirating burner, which usually has the fuel and primary air inlet at the burner bottom and flame at the top. The primary air is usually not adjustable for this simple design and hard to replace the designated fuel with alternative fuels. This burner type is widely adopted in cookstoves and storage water heaters. Figure 15 (b) is a tube burner, which is usually equipped with an air shutter at the fuel injection location. This burner finds its application in ovens, broilers, outdoor grillers, etc.. Figure 15 (c) is a Venturi jet burner, which has the primary air suction openings at the smallest diameter of the burner. The Venturi effect produces a low pressure at the primary air opening area thus helps aspirate more primary air into the burner. This burner type is usually used for higher heating load residential appliances like laundry dryer, room furnace, etc..



(a) Flat burner *(b) Tube burner*



(c) Venturi burner

Figure 15: Self-aspirating or aspirated burner types in residential appliances

4.2 Fuel Classes and Properties

As discussed in the earlier chapters, the major fuel in residential pipelines is natural gas, whose major content is methane. In the manufactured gas era, the major contents were carbon monoxide, hydrogen and some higher hydrocarbons. Due to the toxicity of carbon monoxide, it is considered to be a dangerous content in the residential pipeline and has already been eliminated in most of the residential pipeline systems around the world. However, due to the carbon-free property and ease of production from renewable energy sources, hydrogen might play an important role in future residential pipeline systems. Therefore, Table 16 lists the key physical and chemical properties of representative residential gaseous fuels. The major fuel properties are from Reference [182] and [183], with exceptions being noted.

Lower hydrocarbons have smaller sizes of molecule which result in lower density and ease of leakage. Currently, leakage of natural gas into atmosphere from residential buildings is also becoming a concern for climate change [181]. Also, if pipeline gas contains carbon monoxide, the leakage of this gas can be detrimental due to the density similarity between carbon monoxide and air, which makes it harder to dissipate out of residential houses. Hydrogen has the smallest molecule thus lowest density among all the combustible fuels, therefore, hydrogen is easier to leak out from the pipeline. However, some studies also show that the leakage rate of hydrogen is at the same level of natural gas in low-pressure infrastructures like pipeline in residential buildings [184]. Due to the high reactivity of hydrogen, the leakage danger can be a big challenge in its residential applications. Some researchers are also arguing that the leakage of hydrogen might be less dangerous compared to other fuels, because the low density of hydrogen increases its ventilation rate. Both numerical and experimental studies were conducted to evaluate the danger level of hydrogen leakage in residential houses and results show that the ventilation rate of leaked hydrogen is highly dependent on leakage rate and location [185][186]. There is no definite conclusion yet, regarding the danger level between natural gas and hydrogen leakage.

Another important fuel property is the flame speed of the fuel. Table 16 compares the laminar flame speed of different fuels at stoichiometric condition. As shown, listed hydrocarbons have very similar flame speed, which is between 0.4 and 0.5 m/s. Carbon monoxide has a lower flame speed. To be noted, the laminar flame speed of hydrogen is around 5 times that of methane. This might increase the danger level of flashback when replacing the pipeline natural gas with hydrogen.

Another property difference is the fuel flammability range in air. Methane, which is the major content in natural gas, has a relatively small range of 5%-15%. This property makes it a “safe” gas to be used in residential buildings. Ethane and propane have similar flammability properties. However, carbon monoxide and hydrogen have much wider flammable ranges, especially hydrogen. Hydrogen can be ignited when its volume percentage in air is between 4% and 75%. This property brings extra challenge in fuel/air flow rate control and burner design.

The high reactivity of hydrogen is also reflected on its low ignition energy and smaller quenching distance. However, these properties also have positive influences on the combustion performance. For example, the ease of hydrogen ignition can reduce ignition time, which decreases the fuel leakage in the ignition process. Moreover, the high reactivity can also increase the flame stability thus allow combustion to take place at fuel lean condition, which helps decrease thermal NO_X emissions. The adiabatic flame temperature of hydrogen is slightly higher than that of methane (around 4% higher at stoichiometric condition), which might bring up the thermal NO_X emissions at fuel rich combustion conditions but shouldn’t be a big concern for lean operating conditions.

An important fuel property for combustion is the heat release rate, which is closely related to the heating value and Wobbe Index. Heating value is either interpreted on a volume base or on a mass base. Higher heating value is usually used to describe the total heat release from the fuel, including the latent heat from the water in the exhaust, while lower heating value doesn’t consider the latent heat. It should be noted that natural gas is usually sold from utilities to customers by higher heating value, however, most of our residential appliances can’t not recover the latent heat. That means we are paying the gas bill for the higher heating value of the fuel, but we are only using the lower heating value of the gas. To be noted, some modern high efficiency appliances, which equipped with water condensing heat exchangers can recover the latent heat from the exhaust. These appliances are usually of higher heating loads, like room furnace or water heaters. The appliances with water condensing systems can usually be labeled as energy efficiency higher than 90%.

As shown in Table 16, the volumetric heating value of a gaseous fuel decreases as the fuel goes to lower hydrocarbons or carbon/hydrogen free. However, on a mass base, the heating values of hydrocarbon fuels are at a similar level. Although the volumetric lower heating value of hydrogen is at 9.8 MJ/m^3, which is around 30% of methane, the heating value of hydrogen on the mass base is more than twice that of natural gas. Wobbe Index is usually correlated to the heat release rate in combustion applications. It should be noted that although methane and hydrogen have distinct heating values, their Wobbe Index are very similar, which gives a positive sign for fuel interchangeability.

Table 16: Property comparison among common residential fuels (at 298.15K and 1 atm)

Fuel Properties	Unit	Propane (C_3H_8)	Ethane (C_2H_6)	Methane (CH_4)	Carbon Monoxide (CO)	Hydrogen (H_2)
Density	kg/m^3	1.808	1.219	0.648	1.131	0.0813
Viscosity	10^{-5} Pa·s	0.82	0.94	1.11	1.49	0.89
Laminar flame speed (ϕ=1)	m/s	0.46	0.45	0.43	0.17 [187]	2.1
Low flammability	ϕ	0.51	0.50	0.46	0.34	0.1*
	vol %	2	3	5	12.5	4
High flammability	ϕ	2.83	2.72	1.64	6.76	7.2*
	vol %	11	14	15	74	75
Ignition energy (ϕ=1)	10^{-5} J	30.5	42	33	-	2
Quenching distance (ϕ=1)	mm	2.0	2.3	2.5	-	0.64
Adiabatic flame temperature (ϕ=1)	K	2267	2259	2226	2400	2318

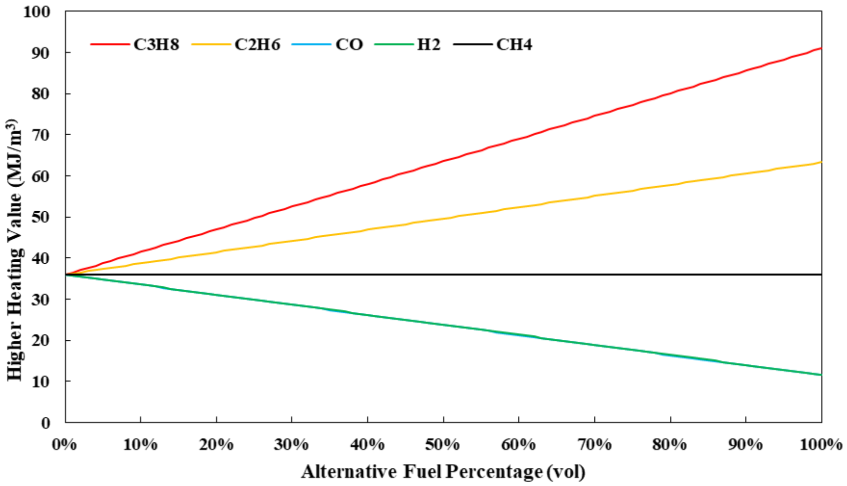
Lower heating value (LHV)	MJ/m^3	83.9	57.9	32.4	11.5	9.8
	MJ/kg	46.4	47.5	50.0	10.2	120.1
Higher heating value (HHV)	MJ/m^3	91.1	63.3	36.0	11.5	11.6
	MJ/kg	50.4	51.9	55.5	10.2	142.1
Wobbe Index	MJ/m^3	73.7	62.4	48.6	11.8	44.3

* It should be noted that the flammability range in Reference [182] has errors. The details are shown in Appendix A. This correction is confirmed with Prof. Stephen Turns through personal communication.

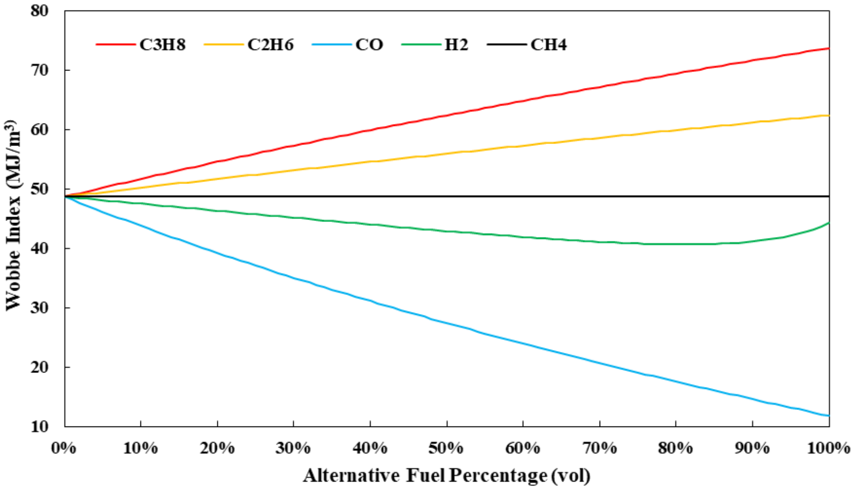
Although all of the fuel properties can influence the combustion performance, there are some essential parameters that need to be considered ahead of other ones, when it comes to fuel interchangeability. For example, heating value and Wobbe Index are two priority ones regarding the heat release properties of the fuels.

4.2.1 *Heating value and Wobbe Index*

The major function of a fuel is to convert its chemical energy into heat, which potential is presented as heating value. Due to the similarity between natural gas and methane properties, methane is used to conduct analysis. The higher heating value and Wobbe Index of methane and other fuel mixtures are plotted in Figure 16.



(a) Higher heating value of methane/alternative fuel mixtures



(b) Wobbe Index of methane/alternative fuel mixtures

Figure 16: HHV and WI of methane/alternative fuel mixtures

Figure 16 (a) shows the heating value curves of methane and other fuel mixtures. The volumetric heating value of methane is 36.0 MJ/m^3 (298K, 1 atm). With alternative fuels mixed with methane, the higher heating value of fuel mixtures show a linear change. When methane is mixed with fuels of higher heating value like propane and ethane, the volumetric heating value shows an increasing trend. However, when methane is mixed with hydrogen and carbon monoxide, the heating value of the mixture shows a decreasing trend. Due to the similarity of the higher heating value between hydrogen and carbon monoxide, the two lines are overlapping.

Differing from the fuel mixture heating value plot, the Wobbe Index plot doesn't show a linear plot due to the definition of Wobbe Index.

$$Wobbe\ Index = \frac{Higher\ Heating\ Value}{\sqrt{Fuel\ Specific\ Gravity}} \qquad \text{Equation (2)}$$

While the Wobbe Index of the fuel is widely adopted today, the concept of using heating value over square root of fuel density as a fuel interchangeability parameter originates from an American Gas Association (AGA) study in the 1920s, which is mentioned in previous discussion. In this study, a "C-index" of change in performance of appliances was established as a critical parameter, which is the predecessor of the widely used Wobbe Index today.

To illustrate the reason why Wobbe Index can be used to predict interchangeability of heat release rate, the Bernoulli equation can be used as a starting point, and a few assumptions made which are applicable to typical appliance operating conditions. By assuming the fuel flow within the pipeline feeding the device and the combustion device itself is at steady state, and the flow is incompressible (constant fluid density) and inviscid, the Bernoulli equation is reduced to Equation (3).

$$p_1 + \frac{1}{2}\rho_f u_1^2 = p_2 + \frac{1}{2}\rho_f u_2^2 \qquad \text{Equation (3)}$$

In the above equation, p_1 is the natural gas pressure in household pipeline, which is at a low level around 2000 Pa. For most appliances, combustion usually takes place at open-air condition, therefore, p_2 is the atmospheric pressure. Equation (3) can be rearranged to solve for the volumetric flow of the gas exiting the fuel port:

$$\dot{V}_f = A_2\sqrt{\frac{2\Delta p}{\rho_f(1-\left(\frac{A_2}{A_1}\right)^2)}} \qquad \text{Equation (4)}$$

As shown, the volume flow rate of the fuel is a function of the fuel density, fuel flow areas, and pressure drop from the household pipeline to the combustion device. However, due to the relatively small orifice area for the combustion device compared to the gas feed cross section, $(A_2/A_1)^2$ can usually be ignored, which leads to Equation (5).

$$\dot{V}_f = A_2\sqrt{\frac{2\Delta p}{\rho_f}} \qquad \text{Equation (5)}$$

Therefore, the heat output of a combustion device can be presented as

$$\dot{q} = \dot{V}_f HHV = \sqrt{\frac{2\Delta p}{\rho_{air}}}\frac{HHV}{\sqrt{sg_f}} \qquad \text{Equation (6)}$$

Based on the assumption of constant density and neglecting viscous effect, the heat output of the combustion device is only a function of the heating value over the square root of the fuel specific density.

$$Wobbe\ Index = \frac{HHV}{\sqrt{sg_f}} \sim \dot{q} \qquad \text{Equation (7)}$$

The reason of Wobbe Index working well for predicting the heat output of different combustion devices is the validity of the assumptions in practice. The Mach number of the gas flow in the household pipeline is much smaller than 0.3. Therefore, it can be assumed that the gas is incompressible in the pipeline. When the gas flows through the orifice of a combustion device, the gas pressure drop can accelerate the flow, which will result in friction loss. However, the gauge pressure in the pipeline is only at around 2000 Pa, which is less than 2% of the atmosphere pressure. Therefore, the pressure loss is not significant.

The Wobbe Index of methane and alternative fuel mixtures (assuming alternative fuel takes x% of the volume in the fuel mixture) are given by Equation (8).

$$WI_{Mix} = \frac{(1-x\%)HHV_{CH4} + x\%HHV_{Al}}{\sqrt{\frac{(1-x\%)\rho_{CH4} + x\%\rho_{Al}}{\rho_{Air}}}} \qquad \text{Equation (8)}$$

As shown in Figure 16 (b), although the heating values of hydrogen and methane are significantly different from each other, they have similar Wobbe Index values. This observation leads to the consideration that part of the pipeline natural gas can be replaced with hydrogen without influencing the heat output in current combustion devices. In other words, a 5 kW burner will retain a nearly 5 kW rating, when operating on natural gas/hydrogen mixtures.

4.2.2 *AGA indices*

Even though heating value and Wobbe Index are essential indicators for fuel interchangeabilities, they can’t predict all the other combustion performances like flame characteristics, flashback limits, emissions, etc.. For example, due to the nonlinear character of the Wobbe Index definition, 37% hydrogen/63% methane mixture has the same Wobbe Index as pure hydrogen. These two different fuel classes might have the same heat release rate in a combustion device, however, their combustion performance should differ from each other significantly. Therefore, there are also other flame indices can help predict the combustion performances besides heat release rate, like flame lifting, flash-back, yellow tip, etc..

The most widely applied flame indices are AGA flame indices regarding flame lifting (I_L), flame flashback (I_F) and yellow tip (I_Y) [72][188][189]. It should be noted here that in Reference [72], the flame lifting index (I_L) was wrong.

$$I_L = \frac{K_a}{\frac{f_a a_s}{f_s a_a}\left\{K_s - \log\left(\frac{f_a}{f_s}\right)\right\}} \qquad \text{Equation (9)}$$

$$I_F = \frac{K_s f_s}{K_a f_a}\sqrt{\frac{H_s}{39934}} \qquad \text{Equation (10)}$$

$$I_Y = \frac{f_s a_a Y_a}{f_a a_s Y_s} \qquad \text{Equation (11)}$$

- *K*: lifting limit constant; *a*: volume of air theoretically required for complete combustion; *f*: primary air factor; H: Higher heating value of the fuel (unit: kJ/Nm^3); *Y*: yellow tip coefficient.
- Subscripts *a* and *s*: designating adjustment and substitute gases, respectively.

Gas utility companies usually have their own AGA index regulations. For example, SoCalGas Company in the U.S. have the following requirement: $I_L \leq 1.06$, $I_F \leq 1.2$, $I_Y \geq 0.8$.

4.2.3 *Weaver indices*

Another series of interchangeability index are the Weaver index series [72]. This series included the considerations of all AGA indices and also three additional indices. The additional indices are the consideration of Wobbe Index, primary air entrainment and incomplete combustion prediction.

Heating load index (J_H) is the ratio of substitute gas Wobbe Index over the original gas Wobbe Index. As J_H getting close to 1, it is more possible to keep the heating load constant while replacing the original fuel with the substitute gas.

$$J_H = \frac{H_s\sqrt{D_a}}{H_a\sqrt{D_s}} = \frac{WI_s}{WI_a} \qquad \text{Equation (12)}$$

- D: specific density of the fuel

Primary air index (J_A) shows the change in the primary air that accompanies a change of gas. It is indicated by Weaver that this index not only provides an accurate measure of primary air to burn two gases but an almost accurate measure of secondary air as well. The ideal value of J_A is also 1.

$$J_A = \frac{a_s\sqrt{D_a}}{a_a\sqrt{D_s}} \qquad \text{Equation (13)}$$

- a: volume of air theoretically required for complete combustion

Lifting index (J_L) the relative tendency for flames of the two gases to lift from the burner ports. When J_L=1, the two gases are interchangeable regarding the flame position.

$$J_L = J_A \frac{S_s}{S_a} \frac{100-Q_s}{100-Q_a} \qquad \text{Equation (14)}$$

- S: flame speed of the fuel in air

- Q: percentage of oxygen in the gas

Flashback index (J_F) shows the relative tendency for flames of the two gases to flash back into the burners. J_F=0 indicates there is no difference between the gases in this respect.

$$J_F = \frac{S_s}{S_a} - 1.4J_A + 0.4 \qquad \text{Equation (15)}$$

Yellow tip index (J_Y) indicates the tendency of a fuel producing soot, therefore yellow tips, in flames. As shown in the definition, the higher content of carbon in a fuel, the higher the index can be. Therefore, the ideal vale for J_Y is 0.

$$J_Y = J_A + \frac{N_s - N_a}{110} - 1 \qquad \text{Equation (16)}$$

- N: the number, in 100 molecules of gas, of the total carbon atoms minus one, except for methane.

The last index in this series is incomplete combustion index, which is used to represent the tendency of a fuel to produce unburned hydrocarbons or carbon monoxide in the exhaust. When the two gases are equally likely to liberate carbon monoxide during combustion, J_I=0.

$$J_I = J_A - 0.366\frac{R_s}{R_a} - 0.634 \qquad \text{Equation (17)}$$

- R: the ratio of hydrogen to carbon atoms contained in the fuel

4.2.4 *Other indices*

AGA indices and Weaver indices are the two most widely applied flame index series regarding fuel interchangeability. Although Weaver index series (6 total) is more comprehensive in covering flame characteristics, the three AGA indices are more commonly adopted by gas utilities around the world. This is because gas utilities usually have their own regulations on fuel species, heating value and Wobbe Index first. These basic regulations from utility companies ensure that the combustion performance of the substitute fuel does not deviate from the original fuel by too much. After the substitute fuel is confirmed with the basic regulations, the application of AGA Index can help further test the flame lifting, flashback and yellow tip performance.

Besides AGA and Weaver indices, there are also other indices developed around the world to predict the interchangeability among gaseous fuels. They are usually considered or adopted in scientific research, because they are either too focused on one or two combustion performances or too complicated to be applied in industrial applications. The other indices are listed in Table 17.

Table 17: Flame indices besides AGA and Weaver indices

Name	Description	Reference
Knoy index	Similar to Wobbe Index, which predicts heat release rate interchangeability.	[190]
Dutton indices	This series of indices mainly consider the incomplete combustion behavior or near blow-off condition for higher hydrocarbons. It consists incomplete combustion index, lift index and soot index.	[191]
Delbourg indices	This series is consisted of two indices: yellow tip index and soot formation index.	[191][192][193]

4.3 Fuel Flowrate Control

As natural gas is replaced by renewable gases, different combustion systems might have distinctive responses to the fuel replacement. Some combustion flowrate control systems are equipped with fuel sensors that keep the gas volumetric flowrate constant or at a constant heat release rate (heating load). Some combustion devices even have air trimming systems, which help regulate the air fed to the flame while stabilizing the excess oxygen in the flue gas is at a constant level. This air trimming system ensures combustion take place at a similar equivalence ratio in spite of fuel species variation. However, a large amount of residential combustion devices don’t have such delicate fuel/air control systems, which leaves the Wobbe Index of the fuel significantly important for evaluating the heating load in terms of interchangeability.

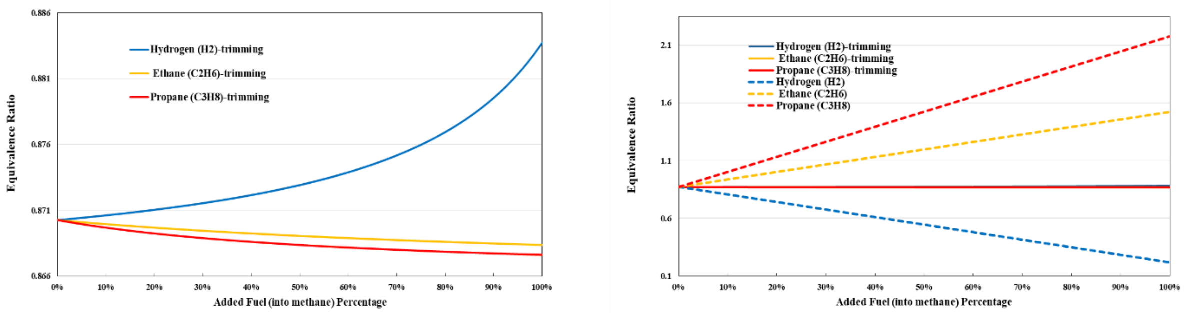
Here are the analysis for the fuel/air control system’s influence on the equivalence ratio and exhaust flowrate.

4.3.1 *Constant fuel volumetric flow*

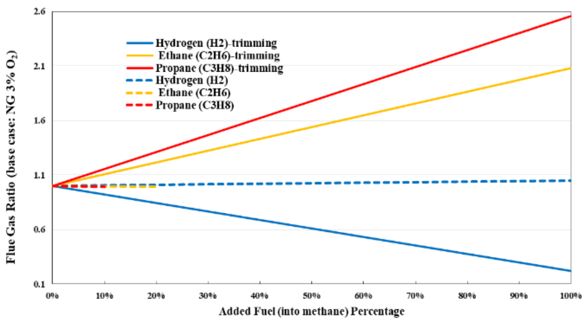
Fuel flowrate controllers of combustion systems are usually calibrated for a certain fuel gas, like natural gas or propane. As the designated fuel is replaced by other gases, most flowrate controllers without fuel sensors can’t response to the fuel composition changes, which means they still allow the same amount of gas flow through it, regardless of the gas property variation.

Figure 17 (a) shows the equivalence ratio variation as the fuel composition changes, with methane being the original fuel. For pure methane reacting in air, the equivalence ratio is 0.87 with

3 vol % excess oxygen in the exhaust on a dry basis. With methane replacing by other gases (hydrogen, ethane and propane), the equivalence ratio varies. With the oxygen trimming system, the equivalence ratio increases for hydrogen-enriched methane. On the other hand, higher hydrocarbon enriched methane mixtures see a decreasing trend. However, this equivalence ratio variation is relatively small. Take hydrogen for example, from 100% methane to 100% hydrogen, the equivalence ratio increased by less than 2% due to the oxygen trimming. As shown in Figure 17 (b), the equivalence ratio lines with oxygen trimming is almost a flat curve compared to the dash lines representing cases without oxygen trimming. Without oxygen trimming, the air supply doesn't change as fuel composition varies. Therefore, hydrogen enriched methane sees an increase of equivalence ratio due its low oxygen consumption per unit volume. On the other hand, the equivalence ratio of higher hydrogen carbon enriched methane increases.



(a) ER variation with oxygen trimming *(b) ER variation with/without oxygen trimming*



(c) Flue gas variation with/without oxygen trimming (dry basis)

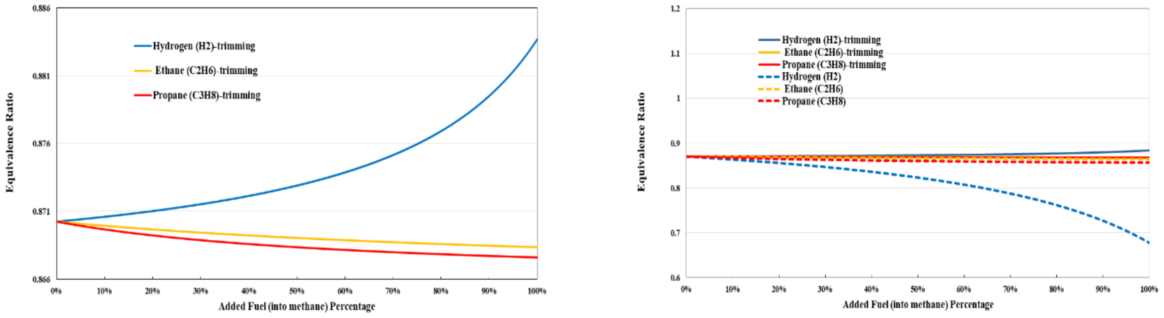
Figure 17: Equivalence ratio and flue gas variation of constant fuel volumetric flow control system

Flue gas flowrate is another important factor that needs consideration in the fuel interchangeability studies. Instead of extracting heat directly from the flame, most of the residential appliance burners extract heat mainly from the high temperature flue gas. In the cooking process, for example, the cooking pan is placed over the flames. Although flame radiation is an effective way of transferring the heat from the heat to the pan, the hot exhaust convection heat transfer is as important as radiation, especially for larger size cooking utensils. For water heating and air heating appliances, the flames are usually located away from the heat exchangers to avoid material degrade, thus to increase the life span of the appliances. Therefore, the flue gas plays a more significant role in these combustion applications. The hot exhaust flows through a heat exchanger and transfer the heat to either air or water. To extract as much heat as possible, some appliances adopt multistage heat exchangers or condensing heat exchangers. As shown in Figure 17 (c), the flue gas flowrate remains almost at a constant level for all fuel classes without oxygen trimming (dash lines). At this condition, the flowrates of both fuel and air remain constant. However, ethane and propane lines stop at 20% and 10% replacement respectively, due to the equivalence ratio exceeding 1. With oxygen trimming, higher hydrocarbon fuels see an increase in the flue gas. In contrast, hydrogen sees a decrease in the flue gas flowrate. This decrease is caused by two reasons: one is the low exhaust generation rate per hydrogen molecule; second is its carbon-free property generating only water in the exhaust. Therefore, if natural gas is replaced by hydrogen while maintaining constant volume flowrate, the total flowrate of the flue gas deceases, which results in a longer residence time in the heat exchanger. However, this is not necessarily a positive result, since the heat exchangers are usually designed for a certain flowrate rate range, low flowrate could result in deteriorated heat transfer. Moreover, as more water is generated into the exhaust for hydrogen enriched fuel, more latent heat will be wasted in the absence of the condensing heat exchanger.

4.3.2 *Constant heating load*

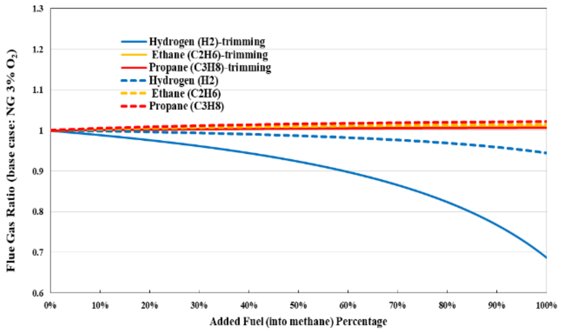
Constant heating load control system is a relatively reliable control systems in terms of fuel interchangeability in combustion devices. Most of the combustion devices, especially residential appliances mainly require the heat output from the fuel. Therefore, one of the major ratings on a combustion application' label is the designed heating load.

As shown in Figure 18 (a) and (b), constant heating load with oxygen trimming system helps keep the equivalence ratio at a constant level for different fuel mixtures. Even without the oxygen trimming system, the equivalence ratio of higher hydrocarbon enriched fuels doesn't vary much at constant heating load conditions. This is due to the similar chemical properties among hydrocarbons. However, hydrogen enriched methane shows a different trend. The volumetric heating value of methane is around 3 times that of hydrogen. Therefore, to keep the heating load of the combustion device at a constant level, fuel volumetric flowrate should triple its original level when methane is 100% replaced by hydrogen. However, even if the fuel flowrate is tripled, the total oxygen consumption of hydrogen is only 2/3 that of pure methane (same total heating load). Therefore, Figure 18 (b) shows a decreasing curve for hydrogen enriched methane without oxygen trimming.



(a) ER variation with oxygen trimming

(b) ER variation with/without oxygen trimming



(c) Flue gas variation with/without oxygen trimming (dry basis)

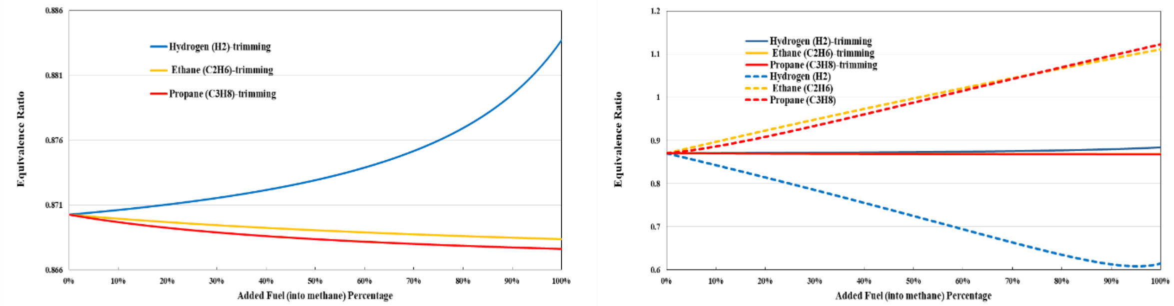
Figure 18: Equivalence ratio and flue gas variation of constant heating load control system

Figure 18 (c) shows the flue gas variation of different fuel mixtures. Compared with constant fuel volume flow condition shown in Figure 17 (c), flue gas variation of constant heating load is relatively small for all mixtures. The most obvious deviation is the hydrogen-methane mixture with fuel trimming system.

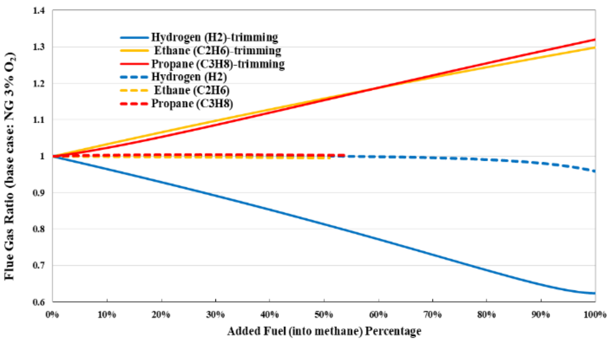
4.3.3 *Without fuel flowrate control (Wobbe Index)*

Numerous combustion applications don’t have delicate fuel control system, which means they are only designed to operate on certain fuel gases. These combustion applications usually operate at lower heating load and are less costive, like lower heat output appliances. The fuel flowrate control mechanism is as discusses in section 4.2.1 of this dissertation.

As can be seen in Figure 19, although the equivalence ratio and flue gas variation trend are similar to the constant volume flow case in Figure 17, the absolute values variation are obviously small. The reason can be found in Table 16. Even if the volumetric heating values of these fuel classes are significantly different from each other, their Wobbe Index values are actually seeing a smaller difference.



(a) ER variation with oxygen trimming *(b) ER variation with/without oxygen trimming*



(c) Flue gas variation with/without oxygen trimming (dry basis)

Figure 19: Equivalence ratio and flue gas variation without fuel flowrate control

4.4 Summary

Fuel interchangeability for combustion involves numerous technical aspects, from fuel property to its storage and transmission, at last end user combustion performance. Due to the inert property of carbon dioxide in biogas, it is usually purified to the fuel with higher methane percentage (biomethane). Biomethane has similar physical and chemical properties as natural gas, therefore, its interchangeability with natural gas is not as challenging as hydrogen.

Here are the technical challenges that need to be considered or dealt with for natural gas replacement with renewable fuels, especially renewable hydrogen:

- The wide flammability range and high flame speed of hydrogen increases the flashback risk in hydrogen enriched fuel combustion applications. Therefore, burners need to be modified (primary air entrainment optimization, flame ports adjustment, etc.) to incorporate more hydrogen in original designated fuels.

- The small molecule size of hydrogen makes it easier to leak compared to other fuel gases, especially for high pressure storage or transport systems without proper sealing. Moreover, the hydrogen attacking or embrittlement effects on materials needs extra attention while choosing materials to contain hydrogen.
- Fuel gas quality regulations on heating value, Wobbe Index or other flames indices need to be reevaluated for renewable gas adoption.
- Burner combustion performance needs to be tested while operating on natural gas/renewable gas mixtures. The monitored aspects should include efficiency, emissions, burner temperature, combustion noise, etc..

In conclusion, fuel interchangeability should be a comprehensive work from fuel quality monitoring to burner performance testing. Residential gas appliance performance evaluation didn't attract much attention before, compared with power plants, vehicle engines and industrial applications. Therefore, in this new energy transition from fossil fuel to renewable energy, new standards on the appliances could be developed to enhance residential house safety and contribute to a renewable future in the residential sector.

CHAPTER 5: Experiment Results of Residential Burner Testing

The objective for this chapter is to test the selective residential appliances performances operating on natural gas/renewable natural gas mixtures, thus helping guide fuel interchangeability studies on combustion devices adopting self-aspirating flames.

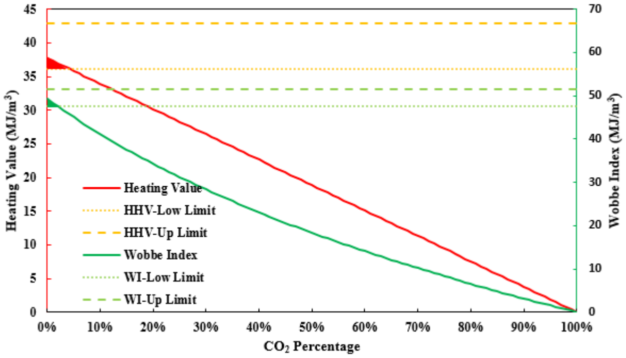
5.1 Overview

In the following experiments, natural gas is mixed with carbon dioxide (simulated biogas) and hydrogen separately, to test the combustion performance of different self-aspirating burners operating on different mixtures. The natural gas contents in UCI Combustion Laboratory is shown in Table 18. As can be seen, the methane content is 95.8%, together with some higher hydrocarbons like ethane, propane, etc.. The inert gas species are carbon dioxide and nitrogen. The natural gas is provided by SoCalGas Company, which uses SCG Rule 30 [67] to regulate their gas qualities. SCG Rule 30 regulates the gas quality from different perspectives: heating value, Wobbe Index, moisture content, sulfur content, inerts, etc..

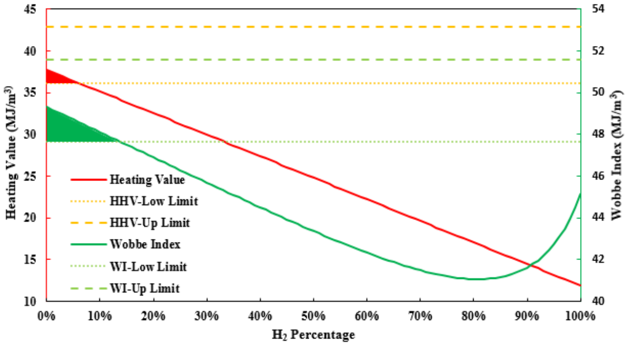
Table 18: Natural gas contents in UCICL

	Content	Molecular Formula	Molecular Fraction
1	methane	CH_4	95.8%
2	ethane	C_2H_6	1.4%
3	propane	C_3H_8	0.4%
4	iso-butane	C_4H_{10}	0.05%
5	n-butane	C_4H_{10}	0.05%
6	iso-pentane	C_5H_{12}	0.025%
7	n-pentane	C_5H_{12}	0.025%
8	C6	C_6H_{14}	0.017%
9	C7	C_7H_{16}	0.017%
10	C8	C_8H_{18}	0.016%
11	carbon dioxide	CO_2	1.9%
12	oxygen	O_2	0%
13	nitrogen	N_2	0.3%

Figure 20 (a) shows the heating value and Wobbe Index variation of natural gas/carbon dioxide mixtures. The dash lines represent the regulations of SCG Rule 30. According to the heating value and Wobbe Index regulation, only a small amount of carbon dioxide can be mixed into natural gas. In 2019, SCG decreased the heating value standard from 990 Btu/scf to 970 Btu/scf, which allows the maximum hydrogen percentage to increase from 3 vol% to 6 vol%. Due to the similarity of Wobbe Index between natural gas and hydrogen, 14 vol% hydrogen can be mixed into natural gas. However, the work of dissertation is not limited to these regulations. The test conditions purposefully go beyond the current regulations. The hydrogen is increased all the way from 0% to the point where an operability issue occurs.



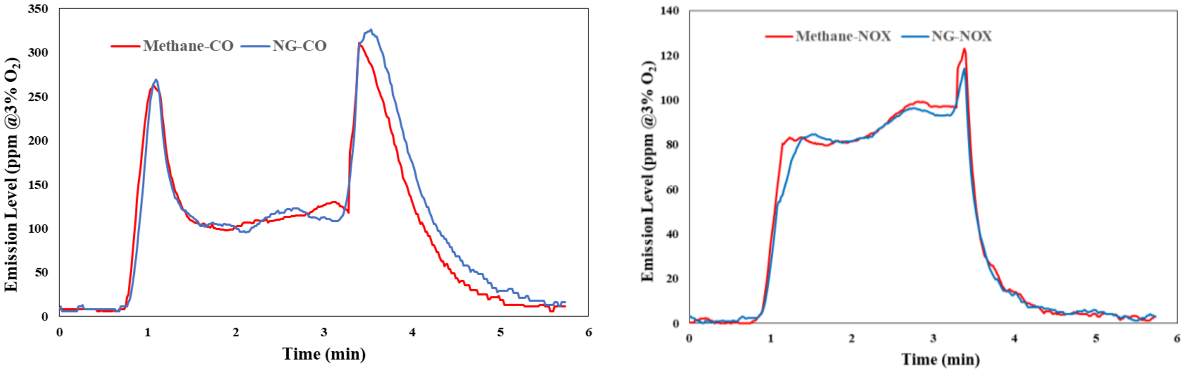
(a) Heating value/Wobbe Index of NG/CO$_2$ mixtures



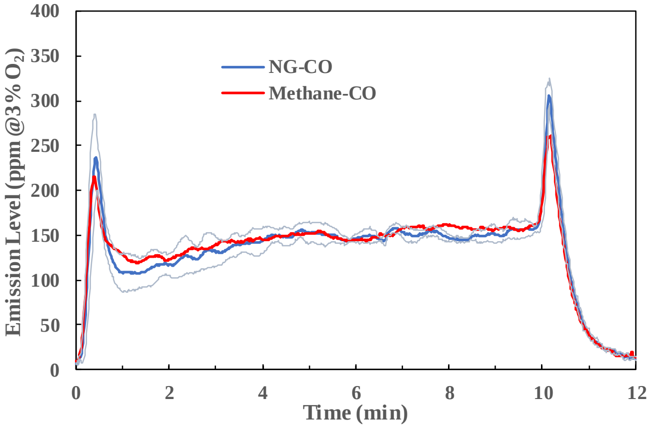
(b) Heating value/Wobbe Index of NG/H$_2$ mixtures

Figure 20: Fuel mixtures heating value and Wobbe Index variations under SCG Rule 30 regulations

Natural gas and methane are compared with each other in the combustion experiments on cooktop burners. Figure 21 (a) and (b) show the CO and NO$_X$ emissions of these two fuel classes for a 3 min combustion test. As can be see, the emission curves are almost overlapping. Figure 21 (c) shows the CO emissions comparison for 10 min tests. The emission data of methane falls within the variation range of the natural gas test. Therefore, natural gas is adopted instead of pure methane in this dissertation’s experiment.



(a) Methane-natural gas CO emissions comparison *(b) Methane-natural gas NO$_X$ emissions comparison*



(c) Methane-natural gas NO$_X$ emissions comparison

Figure 21: Emissions comparison between natural gas and methane on cooktop burners

Different fuel mixtures are generated in the fuel mixing station shown in Figure 22. The flow rates of different fuels are controlled by the pressure regulator-sonic orifice system. A certain pressure drop for a certain orifice presents a specific gas flow rate. Therefore, fuel inlet pressures are adjusted to give a designed fuel flow rate and then the percentages of different fuels in the mixture can be controlled. As shown, there are 4 fuel flow channels, and each channel has two sonic orifices. These sonic orifices are replaceable so to vary the fuel flow rate in a wider range. Different fuels will be injected into a manifold and mix with each other. After the manifold, the fuel mixture flows into an expansion tank, whose volume is less than half a liter. Before the fuel mixture is supplied to the appliances, there is a regulator between the expansion tank and the appliance, and the fuel mixture pressure is regulated down to around 10 inches of water to simulate the actual residential natural gas pipeline pressure.

A National Instrument CompactRIO system coupled with multiple Data Acquisition modules is adopted for flowrate controlling and other measurements. Then the data is acquired and processed by LabView. The fuel pressure signals of different flow channels are collected by pressure transducers and then export to NI-9203. The thermal couples are connected to NI-9213. NI-9215 is adopted to measure the voltage signals of the sound level monitor and emission analyzers.

(a) Fuel station front look

(b) Fuel station inside arrangement

Figure 22: Fuel mixing station setup

The testing tools and their technical details are presented in Table 19. Three emission analyzers are adopted to measure multiple species in the exhaust: Horiba PG-350 (NO/NO_X/CO/O_2/CO_2), QCL-1400 (NO/NO_2/N_2O/NH_3) and FMA-220 (UHCs). Extech-407762 is utilized to monitor the combustion noise. The burner temperature is measured by FLIR-T640. Two Nikon cameras are used to detect the ignition time and characterize the flames.

Table 19: Experimental tools adopted in the experiment

Name	Outlook	Function	Characteristics

Horiba PG-350		Emissions: $NO/NO_X/CO/O_2/CO_2$	Horiba Portable Gas Analyzer 350 (PG350): NO: 0-250 ppm, NO_X: 0-250 ppm; CO: 0-500 ppm; CO_2: 0-10 vol%; O_2: 0-25 vol%. Accuracy: ±2% of the maximum range setting.
Horiba QCL-1400		Emissions: $NO/NO_2/N_2O/NH_3$	Horiba Quantum-Cascade Laser Analyzer 1400 (QCL1400): NO: 0-100 ppm; NO_2: 0-50 ppm; N_2O: 0-100 ppm; NH_3: 0-50 ppm. Accuracy: ±1 ppm.
Horiba FMA-220		Emission: UHCs	Flame Ionization Magneto-Pneumatic Analyzer 220 (FMA220): UHC measurement range: 100 ppm. Accuracy: ±1% of the maximum range setting.
Extech-407762		Sound level	6 ranges from 30 to 130dB, high accuracy to ± 1.5dB and 0.1 dB resolution
FLIR-T640		Burner surface temperature	Temperature Range: -40 - 2000°C; 640 x 480 pixels
Nikon J1		Ignition time	1400 frames per second at slow motion recording mode
Nikon D90		Flame image	Image Resolution: 4288 x 2848

The experiment setup diagram is shown in Figure 23. All of the emission analyzers are synchronized to test different exhaust species from the same testing location at the same time. Each burner has its unique testing procedure and might be slightly different from each other.

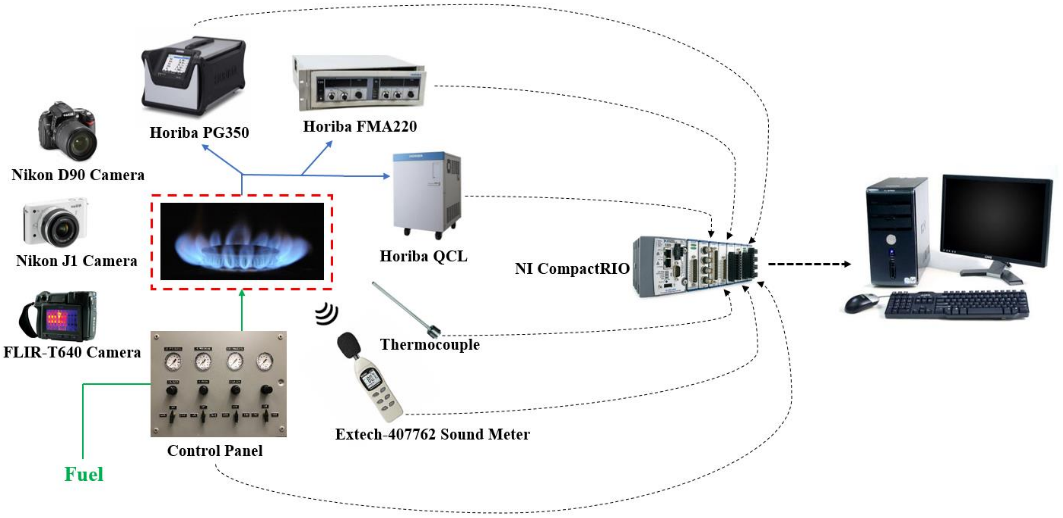


Figure 23: Residential burner performance testing setup

5.2 Cooking Section

Cooking with fire has a long history which is accompanied with human civilization development. Compared to other sections, cooking section has witnessed maybe the most variety of heat sources, from biomass (fuelwood, crop residuals, etc.) to coal, gas and electricity. Currently, the most dominant cooking technology is still through fuel combustion, which is believed to last a long time, maybe even after the fossil fuel depletion on earth.

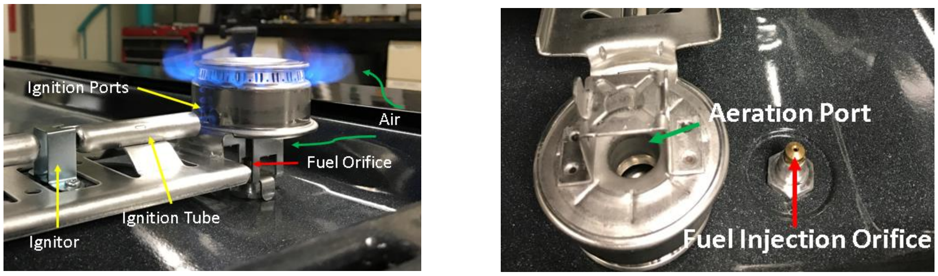
Compared with other residential aspects that are adopting combustion technologies, cooking is the most interactive combustion activity since the flames can be seen and even controlled by end users. Therefore, fuel interchangeability studies on the cooking section is of great interest and deserve a comprehensive investigation that goes beyond current regulations on cooking appliances.

5.2.1 *Cooktop burner*

A representative cooktop burner is selected to conduct the fuel interchangeability study in this section. Multiple combustion performances are investigated.

5.2.1.1 Burner configuration

The cooktop burner geometry is shown in Figure 24. The heating load of the cooktop burner is 9100 Btu/hr (~2.7 kW). As shown, fuel is injected into the burner head while entraining a small amount of surrounding air. Part of the mixture flows out of the ignition ports and arrive at the ignitor through the ignition tube. After ignition, the flame propagates to the burner head and a full flame will be established.



(a) Cooktop burner appearance *(b) Cooktop burner fuel injection configuration*

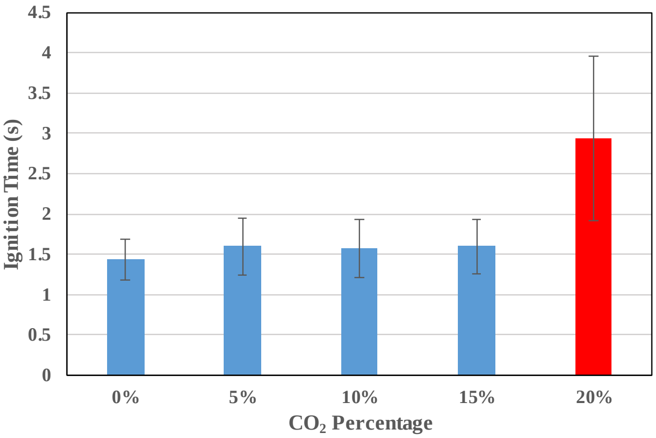
Figure 24: Cooktop burner configuration

The self-aspirating burner utilizes partially premixed combustion technology, which means part of the air (primary air) is mixed with fuel before combustion, and the rest of the air is supplied to the flame as secondary air in the combustion process. The primary air to fuel ratio is established such that the mixture is above the rich flammability limit for safety purpose. The burner configuration studied has a measured fuel concentration of around 18%, which means that the mixture exceeds the rich flammability limit of natural gas and therefore combustion cannot occur within the burner head. However, hydrogen has a flammability range of 4% - 75%, this means that if the burner operated on hydrogen-rich fuel, the hydrogen percentage within the burner head can fall within the flammability range very easily. Hence the risk of the flame flashback into the burner head for hydrogen is much higher than natural gas. As a result of the widely different combustion characteristics of hydrogen and natural gas, flashback is expected to occur at some point as the hydrogen level increases. In the present study, this flashback limit amount was considered as an important limiting factor to be determined.

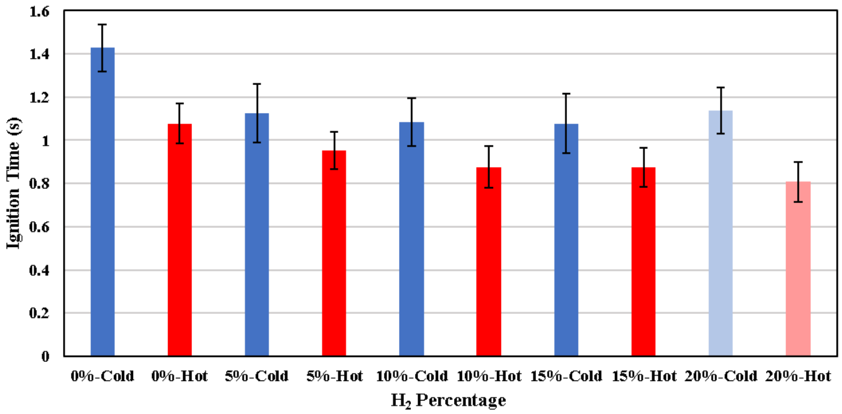
5.2.1.2 Ignition performance

American National Standards Institute-Z21.1 (ANSI-Z21.1) [153] regulates that eligible cooktop burners should ignite within 4 seconds from turning on the spark to stable flame establishment. In this study, the 4 cooktop burners (each with same heating rate specification) are tested. Each burner test is repeated 5 times under both cold and hot ignition conditions.

Figure 25 (a) shows the ignition time for different percentages of CO_2 addition. The average ignition time for natural gas is 1.43s, which is slightly shorter than 5%-15% CO_2 addition, but still within the variance range. With the increase of CO_2 percentage, the ignition time variance becomes larger. When the CO_2 percentage increases to 20%, the ignition time increases sharply and only 4 out of 20 ignition attempts are successful. The results show that CO_2 addition less than 15% will not increase the ignition time. Moreover, all the ignitions happen within 4 seconds. If the burners do not ignite within 4 seconds, they don’t ignite at all. This might be because of the compactness of cooktop burner ignition system; the relatively high flame speed will compensate the cooling effect of inert gas in the mixture. As long as the cooling effect of the inert gas will not quench the flame, it will propagate from the ignition spark to the burner head immediately. Otherwise, ignition will not happen.



(a) Ignition time of the cooktop burner on NG/CO_2 mixtures



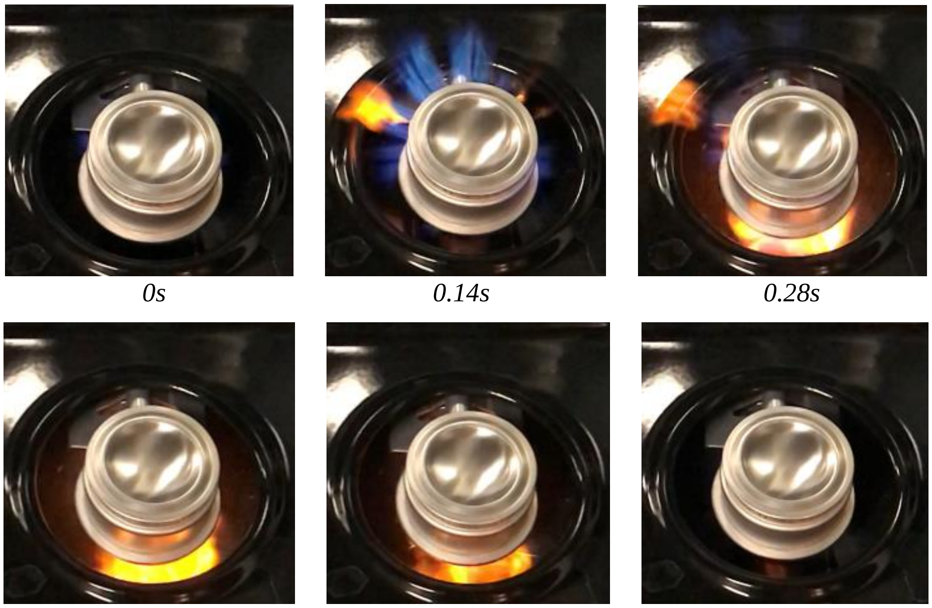
(b) Ignition time of the cooktop burner on NG/H_2 mixtures

Figure 25: Ignition time of the cooktop burner operating on different fuel mixtures

Figure 25 (b) shows the ignition time of the cooktop burner operating on natural gas/hydrogen mixtures. Blue and red bars stand for cold and hot ignition separately. Cold condition means the ignition test is conducted when the cooktop burner starts at room temperature. Hot condition represents a re-ignition test: the burner is operated for 5 minutes, and is then turned off and then turned on again. This re-ignition time is recorded as the hot ignition time. As shown, the ignition time under hot conditions is shorter than that under cold conditions. Figure 25 (b) demonstrates that ignition time decreases a little by increasing hydrogen percentage in the fuel. However, due to the unsteadiness of the ignition process, the variance of the ignition time is relatively large, which is shown by the 95% confidential interval error bars. Under cold ignition condition, 5% hydrogen addition in the fuel decreases the ignition time. However, under hot ignition condition, the ignition time difference between pure natural gas and 5% hydrogen addition is not significant, which is shown by the confidential interval overlap of these two conditions. The reason why this phenomenon appears is that the ignition time decrease caused by hydrogen addition is overshadowed by the high burner temperature.

From 5% to 15% H_2 addition, no significant effect upon ignition is observed. However, with 20% or more hydrogen addition an intermittent flashback starts to occur. Figure 26 shows the flashback process during ignition. After the control knob was turned to “Lite” position, the flame propagates from the igniter to the burner ports. However, before the flame is fully established at the burner ports, flashback occurs. The flame propagates through the burner ports and the ignition ports into the burner head. The fuel/air mixture in the burner head is within the flammable limit, therefore the mixture ignites immediately and flame propagates out of the burner through the bottom aeration port with a loud noise. At last, the flame extinguishes.

Based on these results, it is concluded that more than about 15% hydrogen added to natural gas will lead to operability issues on this type of cooktop burner.



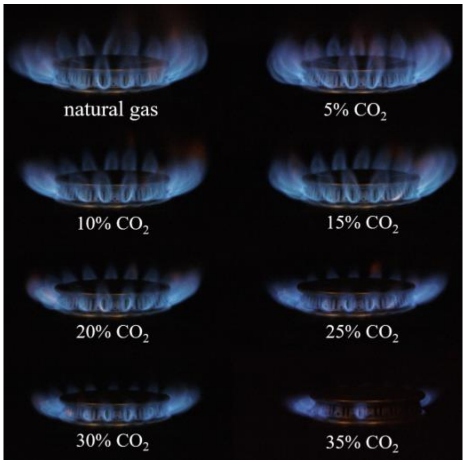
0s *0.14s* *0.28s*

0.42s *0.56s* *0.70s*

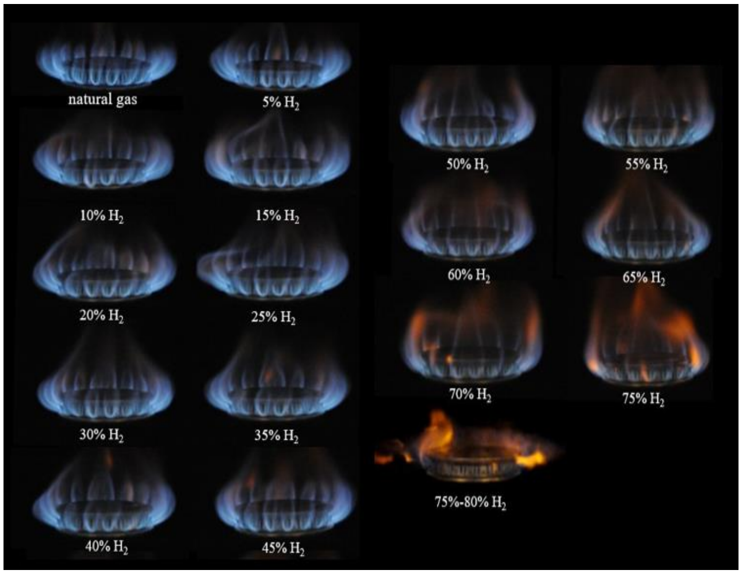
Figure 26: Cooktop burner flashback at 20% H_2 addition

5.2.1.3 Flame characteristics and operability

Flame appearance is an important and also a straightforward way to evaluate the combustion performance of a burner. The flame characteristics of the cooktop burner operating on natural gas/carbon dioxide mixtures are shown in Figure 27 (a). As can be observed, 5% addition of CO_2 doesn’t have much influence on flame structure. However, more than 5% of CO_2 addition will decrease the flame length, and this effect is even more pronounced after 15% CO_2 addition. When the CO_2 percentage is higher than 25%, flame tips will not impinge on the pot surface and starts to fluctuate at the burner ports. After 30% CO_2 addition, the cooktop burner flames can't sustain combustion over all burner ports. As shown in the picture, 35% CO_2 addition results in partial flame coverage over the ports, and the flame bundles hop among neighboring ports before blow-off. During this experiment, no combustion noise or yellow tips were observed.



(a) Flame appearance of the cooktop burner on NG/CO_2 mixtures



(b) Flame appearance of the cooktop burner on NG/H_2 mixtures

Figure 27: Flame appearance of the cooktop burner

Figure 27 (b) shows the flames of the cooktop burner operating on natural gas/hydrogen mixtures. As the hydrogen percentage increases, the flame shows increasing unsteadiness. When the hydrogen percentage increases above 75%, the flame flashes back into the burner head before the 80% hydrogen target is reached. Hence the burner port velocity and quenching characteristics are no longer able to prevent the flame from entering the burner head. It should be noted that this experiment was run under idle conditions, meaning no pot or pan is placed above the flame on the burner top.

Under actual pot heating conditions, an obvious interaction between the flame tip and the bottom surface of the cooking pot is observed, which adds uncertainty and unsteadiness to the flame. As shown in Figure 28, when heating the pot, flashback happens at 55% hydrogen addition, and the burner was destroyed within half a minute. Hence the behavior observed under idle conditions are significantly different from that when actually heating a pot. This observation leads to consideration of how tests are done should a standard method be needed.

Regarding the observed flashback behavior, it is noteworthy that Jones [105] also conducted the hydrogen addition research in 2018. A cooktop burner used in the UK was investigated. However, the only factor considered in Jones's study was flashback safety, compared to the larger spectrum of combustion performance tests conducted in the current study. Jones successfully injected 34.7% hydrogen into the cooktop burner, but the experiment details were not given; therefore, it is not clear whether that was the ignition limit, normal operation limit, cooking limit or the lowest limit of these three. From his analysis, Jones concluded that "It is thus proposed that up to 30 mol% of the natural gas supply may be replaced in the U.K. with guaranteed safety and reliability for the domestic end-user, without any modification of the appliance infrastructure." However, Jones's conclusion is in conflict with the current results which suggest a limit of less than 20% hydrogen addition. It is noted that the cooktop burner used in the present study is also available on the U.K. market. Therefore, studies should be conducted on more burners and multiple combustion performance factors should be considered, before drawing the final conclusion of how much hydrogen should be injected into the pipeline.

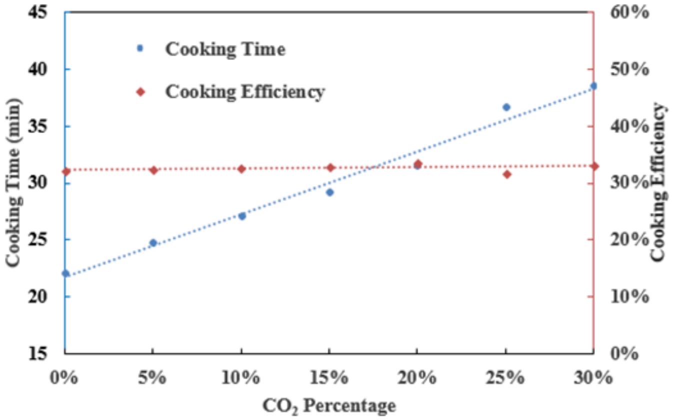
Figure 28: Cooktop burner flashback in the water boiling process

5.2.1.4 Cooking efficiency

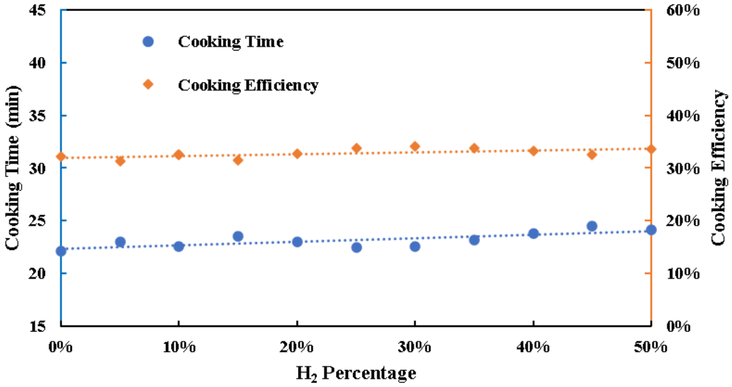
In this section, results associated with the actual heat transfer to a cooking pot are presented. The standard 1 gallon cooking pot recommended by ANSI, is utilized for this test. The pot is filled with water to 80% of the total 1 gallon (3.785 L) capacity.

Cooking time and cooking efficiency are recorded to compare the "cooking performance" of the cooktop burner operating on different fuel classes. Cooking time is defined as the time period of water temperature rising from the room temperature to the boiling point. Cooking efficiency is usually hard to measure, because it is related to a lot of factors including the food being cooked and utensils, etc.. In this research, cooking efficiency is defined as the ratio of heat absorbed by the water to the total heat supplied to the burner, as shown in Equation (1).

Figure 29 (a) shows the cooking time for the water to boil and the cooking efficiency for natural gas/carbon dioxide fuel blends. It takes around 22 minutes to boil 4 quarts of water. When the CO_2 percentage is increased to 30%, it needs 39 minutes for the water temperature to reach the boiling point. It is noteworthy that the cooking efficiency stays at 32% even with increasing CO_2 percentage in the fuel. The favorable scenario for a substitute fuel is to keep the cooking efficiency constant or to increase the cooking efficiency. Faster cooking time is unnecessary, because faster cooking usually means high heating load, which can result in overcooking in a short time. On the other hand, from the emissions view point, longer operating times may potentially lead to an overall increase in the mass emissions of pollutants and CO_2 from the natural gas portion of the fuel.



(a) Cooking time and efficiency of the cooktop burner on NG/CO_2 mixtures



(b) Cooking time and efficiency of the cooktop burner on H_2/CO_2 mixtures

Figure 29: Cooking time and efficiency of the cooktop burner

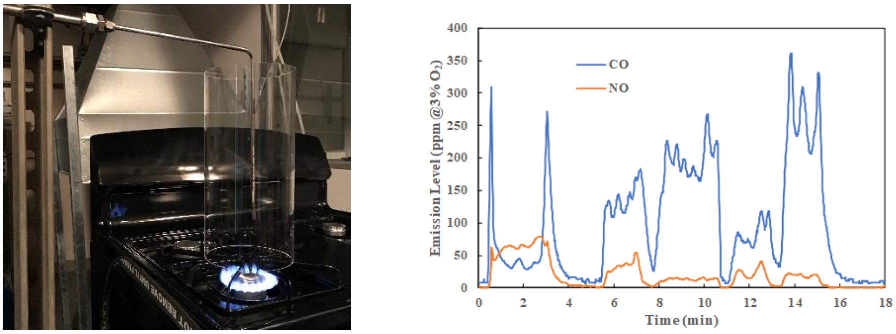
Due to the flashback limit of 55% hydrogen during pot heating, the limit for hydrogen addition is 50% for these tests. In fuel interchangeability study for cooktop burners, an increase in efficiency is desirable, but the case of cooking time is different. A situation resulting in longer cooking time is usually not desirable, and a situation that can dramatically reduce cooking time is also not ideal. This is because shorter cooking time means the cooking appliance is not working at its rated heating load. Therefore, the ideal situation of fuel interchangeability is increasing the cooking efficiency while keeping the cooking time constant.

Cooking time and efficiency variation with hydrogen percentage are shown in Figure 29 (b). Each data point in the figure is the average of 4 individual cooktop burners (each with same heat rate specification). As shown, both cooking time and cooking efficiency increase slightly with hydrogen addition. From 0% to 50% hydrogen addition, cooking time increases by 9% and efficiency increases by 4%. Many factors influence cooking time and cooking efficiency. For example, the adiabatic flame temperature of hydrogen is higher than that of natural gas for a given fuel/air ratio, but the heat input using hydrogen-rich fuel is lower than natural gas due to the slightly lower Wobbe Index of hydrogen. These two effects might compensate each other and result in small variation of cooking time and efficiency. Moreover, the flame characteristic change might also influence heat transfer by radiation and convection. The small variation of hydrogen-rich fuel cooking performance bodes well for adding hydrogen to natural gas as a means to displace carbon.

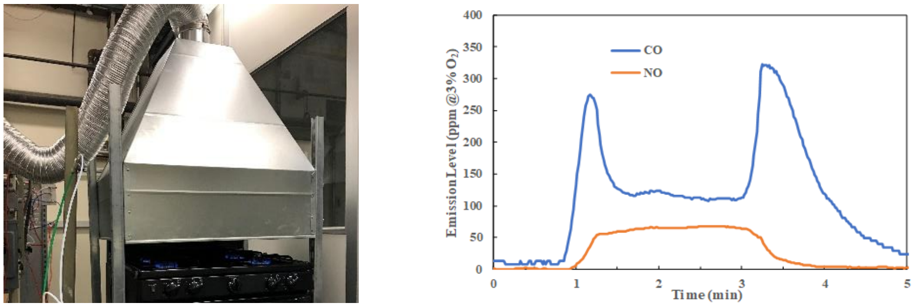
5.2.1.5 Emissions

Two different emission measurement methods are compared with each other in Figure 30. For the quartz tube collection method, a 1-foot long quartz tube with a diameter of 5 inches is used to collect exhaust. The sampling probe has 10 horizontal holes which ensures that we take exhaust from maximum locations in the tube cross section. The probe is located at a mid-location of the quartz tube length. Five horizontal positions within the quartz tube are chosen to take the emission samples: center and four corners of a cross. Emission sampling at each location starts from ignition to turn-off. As shown in Figure 30 (a), the emission levels fluctuate not only with locations but also with time. To use the quartz tube on open-air flames, longer tubes may be required to promote homogeneous exhaust and a star-shaped probe used to take emission samples from multiple locations simultaneously.

To compare with the quartz tube collection method, an exhaust hood was manufactured to collect the emissions of 4 cooktop burners. This method is recommended by ANSI-Z21. The hood should be around 5 inches above the range top surface- to ensure enough surrounding air entrainment into the combustion region. The emissions measured by the hood collection method is shown in Figure 30 (b). The two peaks of CO emission show the ignition and turn-off processes. It is apparent that the emissions are stabilized quickly after ignition. Therefore, in this study, the hood collection method is adopted.



(a) Quartz tube emissions collection



(b) Exhaust hood emissions collection

Figure 30: Emissions comparison of two measurement methods

To simulate the actual cooking process, ANSI Z21.1 recommends conducting water boiling experiments while measuring emissions. Therefore, four 5-quart pots with the diameter of 9 inches were chosen. During the experiment, the pots are filled with water up to 80% of the maximum volume. To investigate the influence of cooking utensil on the emissions, comparative experiments are conducted. In Figure 31, the solid lines are emissions obtained with the water boiling (i.e., with load) and the dashed lines are emissions without the pots (i.e., zero load). In the relatively steady period, CO emission is higher with load over the burners, due to the quenching of flames by the cooking utensils. For the relatively stabilized time period (2-11 min), the average CO emission is 176 ppm (@3% O_2) with load and 144 ppm (@3% O_2) without any load. However, NO_X ($NO+NO_2$) emissions show an opposite trend compared to CO. The average NO_X emission with heating load is 95 ppm (@3% O_2), while on idle burning it is 119 ppm (@3% O_2). The decrease of NO_X in the cooking process is because of the cooling effect of the pots which results in thermal NO_X reduction. In this study, the loaded system is used for the subsequent results.

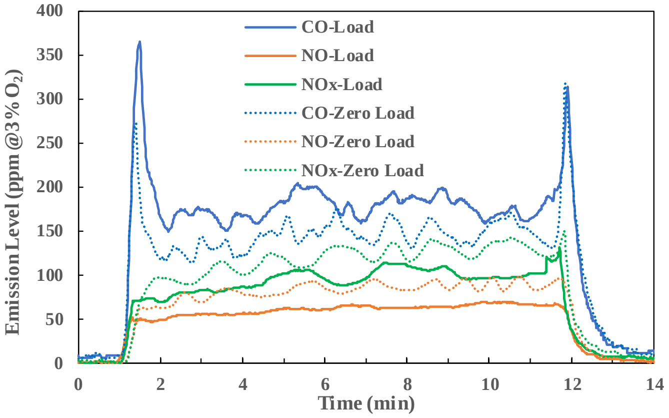


Figure 31: Cooktop burner emissions of load/zero load operating conditions

As concluded in Chapter 1, not many emission regulations on cooking appliances exist. For safety reasons, some regulations mentioned CO, however, no specific regulations for NO_X or UHC exists for cooktop burners. The existing NO_X emission regulations are more generally applied on combustion devices. Moreover, nearly every standard has its own way of measuring or evaluating emissions. Usually, emissions are not regulated by the absolute concentrations in the exhaust, that is because different combustion devices use different combustion technology and sampling methods. This might cause dilution in the exhaust. Dilution of the exhaust results in a reduction of the absolute emission concentrations. To account for this, some regulations correct to a specific oxygen level which is very common in industry. Others correct to a standard carbon dioxide level or use calorific corrections. In the current work, emissions of the cooktop burners are compared using different methodologies to give an insight on the emissions of cooktop burners and provide reference for future regulations.

Oxygen correction of emissions is widely adopted in gas turbine industry. Lean premixed combustion technology has been widely adopted in gas turbine industry, which will result in extra air in the exhaust. Therefore, emissions are usually corrected under a certain level of oxygen. Different oxygen references are used depending on specific situations, 3%, 6%, 15% are most common references in industry. The equation of absolute emission concentration corrected at 3% oxygen is shown.

$$[X]_{3\%\,O_2,ppm} = [X]_{raw,ppm}\frac{20.9-3}{20.9-[O_2]_{raw,\%}} \qquad \text{Equation (18)}$$

Carbon dioxide correction is usually adopted in light industry, commercial and residential combustion devices using diffusion or partially premixed combustion technology. The carbon dioxide concentration reference of 12% is most widely used, which is the carbon dioxide concentration (on a dry base) in the exhaust of methane combustion in air at stoichiometric condition. This correction method is also called air-free method.

$$[X]_{12\%\,CO_2,ppm} = [X]_{raw,ppm}\frac{12}{[CO_2]_{raw,\%}-[CO_2]_{air,\%}} \qquad \text{Equation (19)}$$

Calorific correction is usually adopted in relatively low heating load burners in commercial or residential applications. It describes how much emission is generated per unit energy consumption. HHV and LHV can both be utilized in this correction, but the emission concentration using HHV is relatively lower than using LHV. In the present work, LHV is adopted as it reflects the actual heating value extracted from the fuel for the cookstove. The correction equation is given below.

$$[X]_{ng/J} = \frac{0.1*[X]_{raw,ppm}}{[CO_2]_{raw,\%}-[CO_2]_{air,\%}} \frac{mol\ CO_2}{MJ\ Fuel} M_{raw,g/mol} \qquad \text{Equation (20)}$$

In the emission test experiment, loaded (i.e., water boiling) conditions are used. The emission data are recorded every second. Emission data are analyzed for a 15 min period after a 10 min warm up period. Emissions of cooktop burners operating on different gases are shown in Figure 32 based on different correction methods.

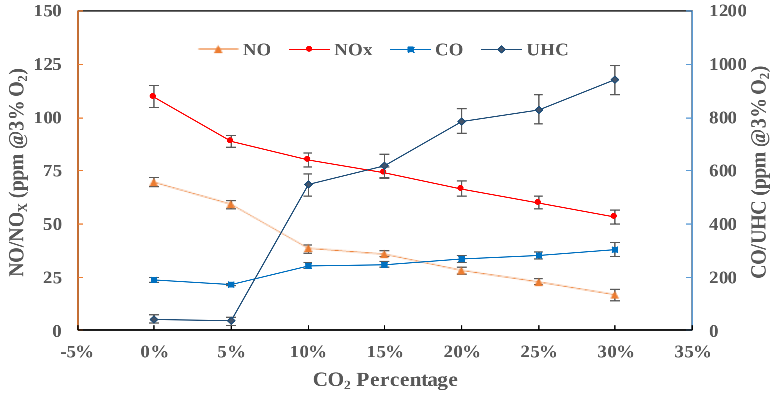
As can be seen, different correction methods don't change the trends of emissions but only the values. Generally speaking, increasing the CO_2 percentage in the fuel will decrease NO_X but increase CO and UHC. During the experiment, the absolute percentage of O_2 in the exhaust does not change much due to the open-air combustion, therefore, oxygen correction can be used as a reference.

Using carbon dioxide correction method doesn't change the emission levels at lower percentage of CO_2 added into the fuel condition. However, the CO_2 in the fuel starts to influence the corrected emissions after 10% CO_2 addition. Although CO and UHC increases significantly when CO_2 addition exceeding 10%, carbon dioxide correction method keeps them constant at around 200 ppm and 600 ppm separately. Therefore, carbon dioxide correction method is not a credible emission evaluation method for high CO_2 biofuels.

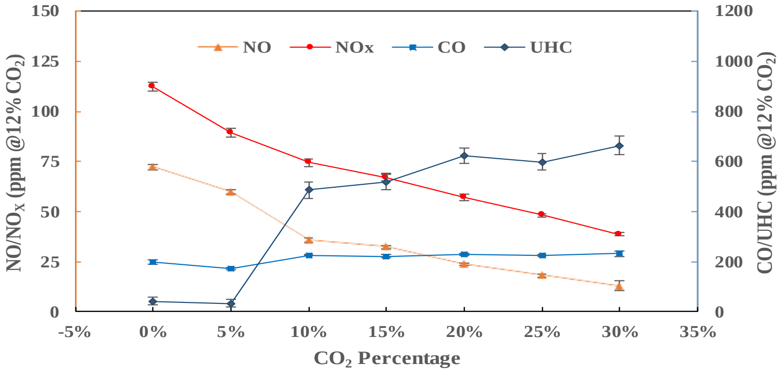
Calorific correction has similar tendency as oxygen correction method, which can also be used as a reliable emission correction method. In Figure 32 (c), NO_X emission is calculated based on the molecular weight NO_2, which makes it easier to match the regulations. When NO and NO_2 are calculated separately, the NO_X concentration is 41.8 ng/J, and CO is 57.9 ng/J when operating on natural gas.

As shown in Figure 32 (a), 5% CO_2 addition into the fuel doesn't have much influence on CO and UHC emissions, but it will help decrease the NO_X emission from 110 ppm (@3% O_2) by 19%

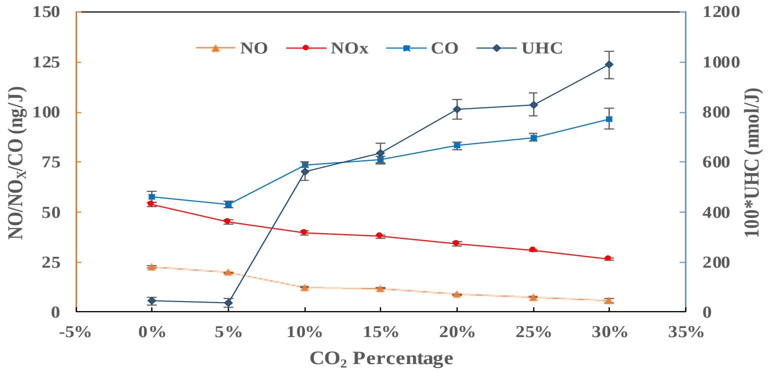
down to 89 ppm (@3% O_2). For pure natural gas and 5% CO_2 addition, CO is around 180 ppm (@3% O_2) and UHC is 40 ppm (@3% O_2). With the CO_2 addition exceeding 5%, NO_X sees a gradual decrease down to 53 ppm (@3% O_2) at 30% CO_2 addition. CO increase from around 180 ppm (@3% O_2) to 302 ppm (@3% O_2). UHC has a dramatic increase with CO_2 addition, especially from 5% to 10%. From pure natural gas to 30% CO_2 addition, UHC increases 22.5 times the initial value, up to 943 ppm (@3% O_2). With the increase of CO_2 percentage in the fuel, NO_X emission variance doesn't change much, but the variance of CO and UHC increase obviously. It is also worthy to note that, NO takes an average of 51% of NO_X during the experiment, which number doesn't vary much with CO_2 addition. This result show that open-air combustion has a high NO-NO_2 conversion rate, in which case NO_2 can't be ignored as some sealed chamber combustion devices do.



(a) 3% O_2 correction



(b) 12% CO_2 correction



(c) Calorific correction

Figure 32: Emissions of the cooktop burner operating on NG/CO_2 mixtures

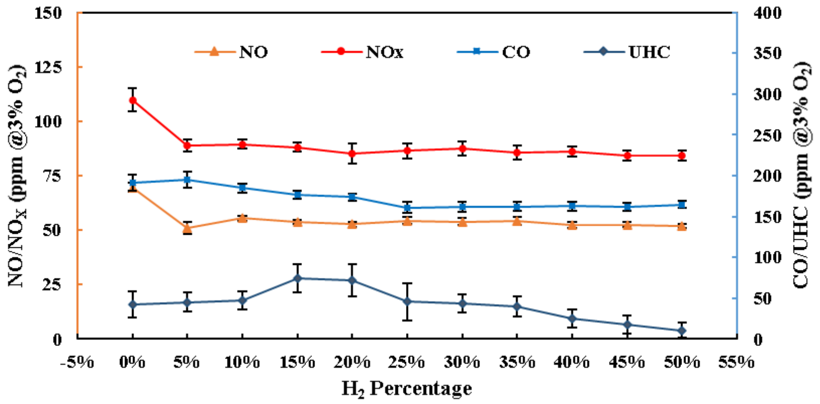
Figure 33 shows the emissions of the cooktop burner. As shown, with the addition of hydrogen, emissions decrease. From pure natural gas to 50% hydrogen, NO_X decreases from 110 ppm (3% O_2) to 84 ppm (3% O_2), and NO drops from 70 ppm (3% O_2) to 52 ppm (3% O_2). CO also decreases from 191 ppm (3% O_2) to 165 ppm (3% O_2). Although UHC emission has some variance, the general trend is that hydrogen addition reduces the UHC emission from 42 ppm (3% O_2) to 11 ppm (3% O_2). With the increase of hydrogen percentage in the fuel, CO and UHC emissions in the exhaust can be reduced as the carbon percentage goes down. In situations with added hydrogen, the higher adiabatic flame temperature contributes to the oxidation rate of the fuel in the combustion process. According to the Zeldovich mechanism, higher reaction temperature usually increases the NO_X emission; however, this study shows a trend of deceasing NO_X emission with more hydrogen addition, which is against the known theory. This effect might be caused by the operating condition. With a situation of 50% hydrogen addition, although hydrogen adiabatic flame temperature is 4% higher than the methane adiabatic flame temperature, the heating load of hydrogen/natural gas mixture is around 12% lower than natural gas. NO_X emissions are reduced because of the steeper heat release drop which overcomes the adiabatic flame temperature increase. It is not ideal to use the properties of the adiabatic flame temperature for open-air combustion analysis, because it is significantly different from the ideal adiabatic situations.

Another observation is that NO and NO_2 emission levels are very similar. For high temperature combustion (e.g., in gas turbines) [194], the formation rate of NO is much faster than NO_2. However, the NO and NO_2 emissions of the cooktop burner are very similar, which is caused by the relatively low temperature open-air flame property. Because of the significant heat loss caused by heating the surrounding air and radiating to the surroundings, the NO formation rate is not that different from the NO_2 formation rate. Moreover, the large amount of oxygen in the surrounding air is supplied to the flame, which helps speed up the oxidation of NO to NO_2.

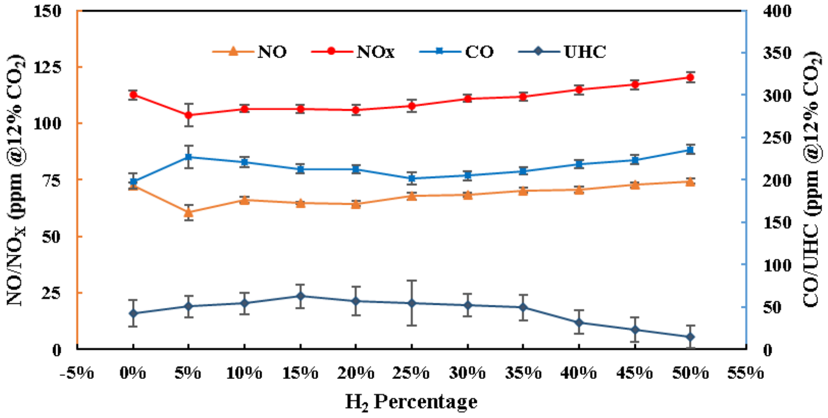
Finally, based on the MEXA 1400-QCL measurements, little to no NH_3 or N_2O emissions are observed (measurements are within the measurement accuracy). Usually, N_2O can result from lean combustion conditions, especially when the equivalence ratio is lower than 0.8 [182]. However, the fuel/air mixture equivalence ratio within the burner head is between 1.5 and 2, which makes the N_2O formation negligible.

In terms of the presentation basis for the emissions, as can be seen in Figure 33 (b), the emissions with added hydrogen content are higher when 12% CO_2 is used as the basis. The CO_2 correction equation has the CO_2 concentration in the denominator, which will become lower with the increase of hydrogen in the fuel mixture. Therefore, NO_X and CO increase as hydrogen addition increases. Compared to the oxygen correction, the CO_2 correction method might not be "fair" for hydrogen-rich fuel emission evaluation.

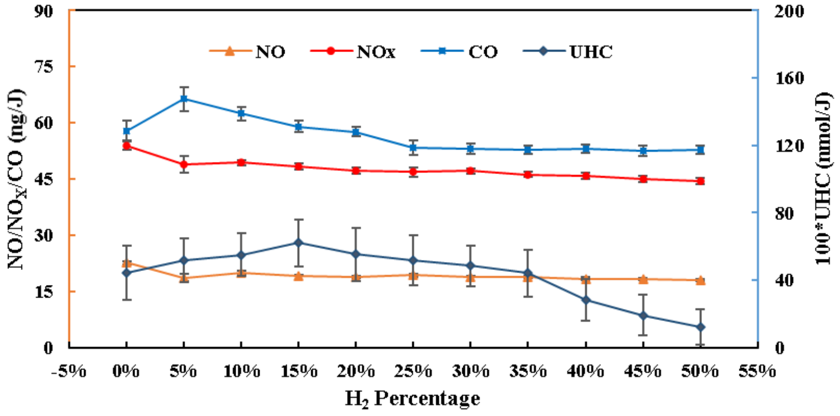
The calorific correction shows a similar trend as the oxygen corrected results. The UHC emission drops sharply at high amounts of added hydrogen. However, the variation of other emission species is smaller compared to oxygen correction. The noteworthy point is the difference of NO and NO_X emission based on calorific correction is larger than that based on O_2 and CO_2 corrections. This is because NO_X calorific correction converts NO to NO_2 on a mass basis.



(a) 3% O_2 correction



(b) 12% CO_2 correction



(c) Calorific correction

Figure 33: Emissions of the cooktop burner operating on NG/H_2 mixtures

5.2.1.6 Other performances

Combustion noise is measured by Extech-407762 at the burner operating condition. The background laboratory sound pressure level is 56 dB. When the burner is operating on pure natural gas, the sound level increases slightly to 56.8 dB. As shown in Figure 34, the combustion noise intensity doesn't change significantly for cases with hydrogen addition up to 40%. At hydrogen addition levels above 40%, however, an obvious "buzz" noise of the flame is evident which is associated with the flame dynamics at higher hydrogen percentages.

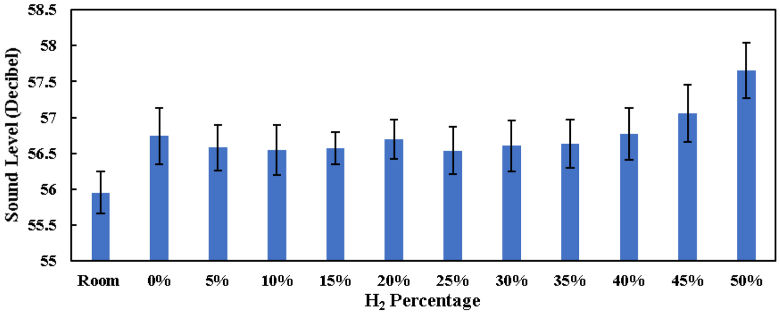
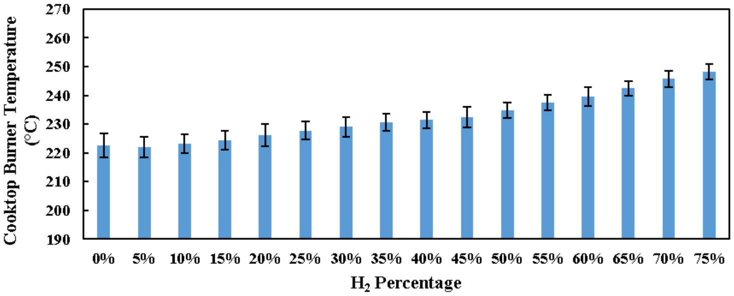


Figure 34: Combustion noise of the cooktop burner operating on NG/H_2 mixtures

Due to the difference between the hydrogen flame velocity and the adiabatic flame temperature, hydrogen addition might also influence the flame stabilization distance at the burner ports which can, in turn, influence the burner temperature. The temperature distribution image of the cooktop burner is shown in Figure 35. The highest temperature of the burner is the area near the injection ports. Therefore, this area is chosen as the representative area for burner temperature measurement. Figure 35 (b) shows that the hydrogen addition increases the burner temperature steadily. At pure natural gas operating condition, the burner temperature is around 223 °C, and it increases to 248 °C at 75% hydrogen addition. Burner life cycles may be reduced by temperature increase. However, the burner is very robust and it might take hundreds of hours of continuous running to find the influence of this temperature increase. If the real cooking procedure was 1-hour level operating, the influence of this temperature increase might be negligible.

(a) FLIR image of the cooktop burner



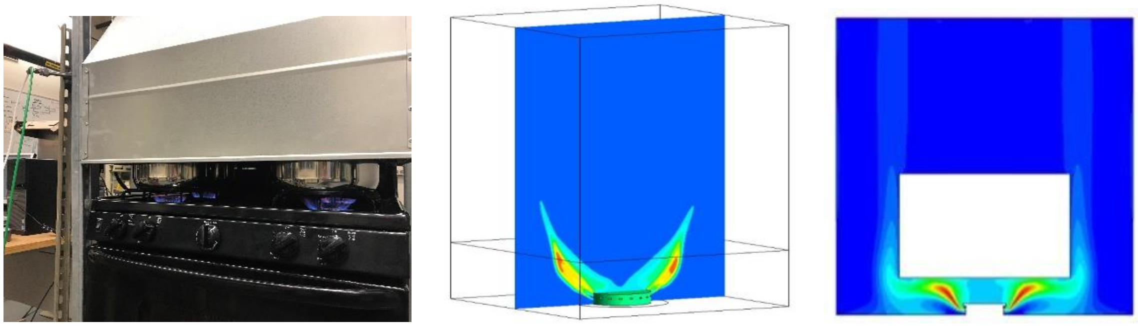
(b) Cooktop burner temperature

Figure 35: Cooktop burner temperature operating on NG/H_2 mixtures

5.2.1.7 Chemical reaction network

The cooktop burner flow-reaction field is modeled by ANSYS-Fluent. The mesh of the simulated burner is generated by ANSYS-ICEM. As shown in Figure 36, the 3-D model is extracted from the experiment setup. Fuel and primary air mixture flows through the burner flame ports and reacts with the surrounding air. In this experiment, the burners and cookpots are covered by the exhaust hood. Therefore, the secondary air is from two places: aeration bowl around the burner and the 12.7 cm (5 inches) gap between the exhaust hood and the cooking range surface.

Afterwards, the 3-D model is simplified to a 2-D model due to its symmetrical property. The number of the mesh nodes are decreased from over 300,000 to around 30,000, which requires less computing resources.



(a) Experiment setup (b) 3-D cooktop burner (c) 2-D cooktop burner

Figure 36: CFD Model generation for the cooktop burner

Figure 37 shows the schematic of the 2-D model. The cooktop burner tested has a diameter of 6 cm and a height of 3.5 cm. There are 42 flame ports and each port is 4.5 *1 mm^2. The hood opening in the simulation model is 40 *40 cm^2. The gap between the hood and the cooking range surface is 12.7 cm (5 inches). The aeration bowl around the cooktop burner has a diameter of 10 cm.

During the experiments, emissions measurements were taken while boiling water. Therefore, a wall boundary is generated above the cooktop burner. In the simulation, this boundary is set as a constant temperature wall, whose temperature stays at 100 °C as the boiling water.

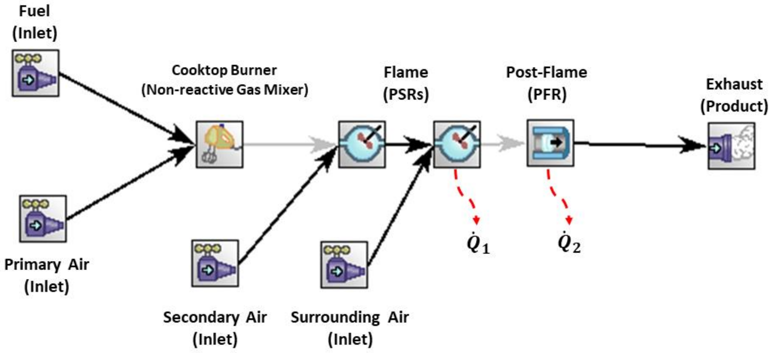
Figure 37: 2-D cooktop burner model

A chemical reaction network (CRN) model is generated based on the experiment and the CFD results, as shown in Figure 38.

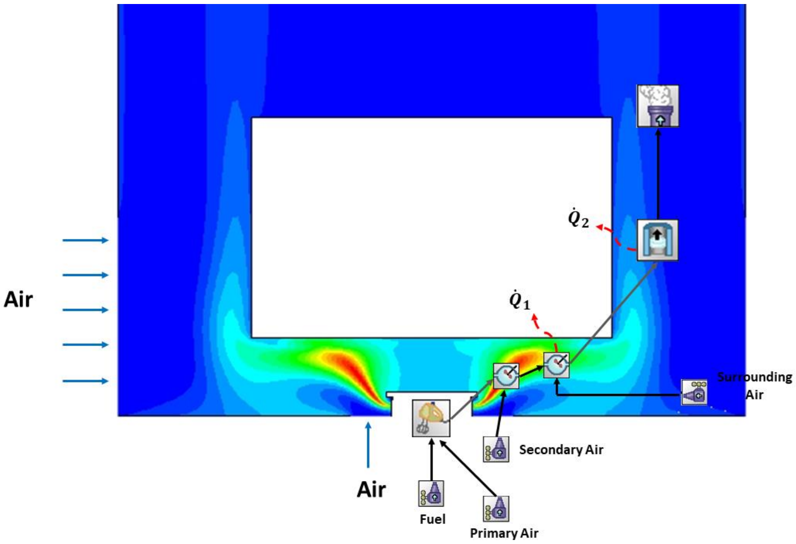
Fuel and primary air are first mixed together in a non-reactive gas mixer, which is used to simulate the cooktop burner head. At pure natural gas operating condition, the equivalence ratio of this mixture is 2.04 according to the experiment tests. Therefore, the mixture is out of the combustible range and the non-reactive gas mixture should be used.

Afterwards, two perfectly stirred reactors (PSRs) are adopted to simulate the flame. The secondary air injection into the first PSR is to simulate the primary air from the aeration bowl around the cooktop burner, and the surrounding air into the second PSR is to simulate the dilution air from the exhaust hood gap. The heat loss $\dot{Q}_1$ from the PSR is 0.33 kJ/s, which is calculated from the heat absorption rate of the heated water.

A plug flow reactor (PFR) is used to simulate the post-flame region. The heat loss $\dot{Q}_2$ is due to hot products heating the water pot side wall. The heat loss rate is 0.06 kJ/(cm*s). The GRI-Mech 3.0 reaction mechanism is adopted.



(a) CRN model of the cooktop burner



(b) CFD-CRN model of the cooktop burner

Figure 38: Numerical model of the cooktop burner

The CO/NO_X emission results of the cooktop burner operating on NG/H_2 mixtures are shown in Figure 39. Both experiment and simulation show that hydrogen injection can help decrease CO and NO_X emissions in a general trend. Experiments show a flat trend, but simulation curves are relatively steeper. However, after 30% hydrogen injection, the decreasing ratio of the CO simulation curve also goes down and NO_X emissions start to fluctuate at a relatively stable level.

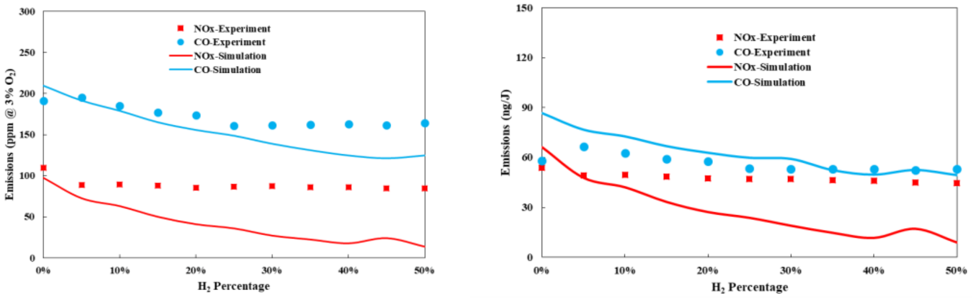


Figure 39: CO/NO_X emissions comparison between simulation and experiments of the cooktop burner

5.2.2 *Oven burner*

5.2.2.1 Burner configuration

A commercially available cooking range with 4 cooktop burners and a 4.8 ft^3 (~0.14 m^3)-capacity oven is selected as a representative residential appliance with which to conduct experiments. The oven burner operates at a rated heating load of 5.3 kW (18,000 Btu/hr) with the oven temperature range from 200 °F (93 °C) to 500 °F (260 °C). The oven burner investigated in this study is a self-aspirating burner, which is widely utilized in residential appliances. The oven burner is usually covered by a metal plate and located at the bottom of the oven. During the burner performance investigation, the plate is removed to facilitate better observation. As shown in Figure 40, fuel flows through an orifice and entrains some surrounding air (primary air) through the aeration port into the tube burner. The air entrainment is achieved only by the fuel momentum, without any forced draft assistance. In other words, the oven burner configuration studied is considered self-aspirating or self-aerated. When the fuel/air mixture flows out of the flame ports on the tube burner, it will be ignited by the ignition source, which, in this case, is an electrically heated surface. Because the fuel/air mixture is usually fuel-rich, the air inside the oven will serve as the secondary air in the combustion process to further oxidize the fuel. In the oven burner design, the amount of primary air can be controlled by adjusting the air shutter. The oven burner is commonly designed for "dual fuel" operation, usually natural gas and propane. When the oven burner is operating on higher hydrocarbons like propane, the air shutter opening is increased to ensure sufficient primary air is entrained to eliminate yellow tips (i.e., sooting).

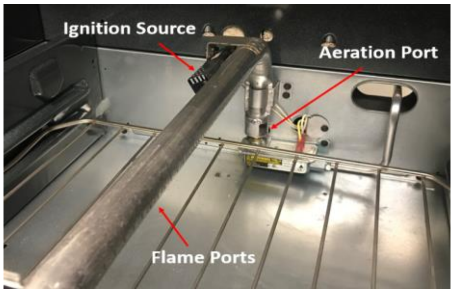
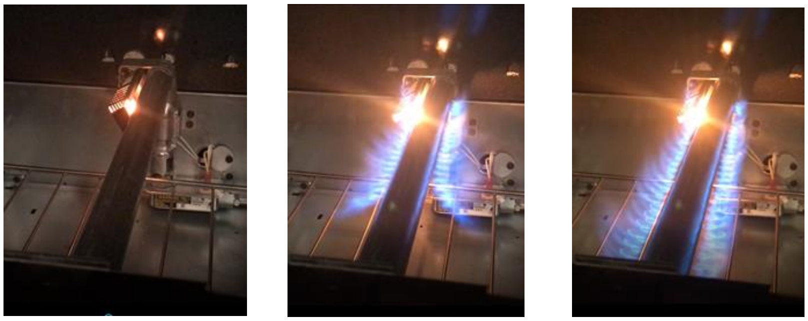


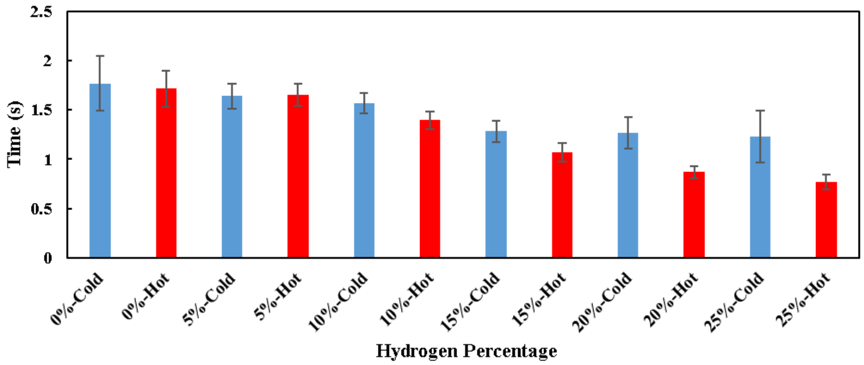
Figure 40: Oven burner configuration

5.2.2.2 Ignition performance

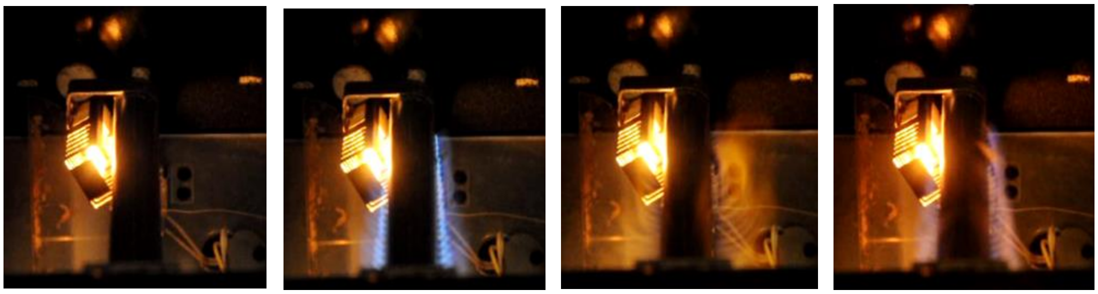
The oven burner ignition process is illustrated in Figure 41 (a). The igniter (electric hot surface) is turned on first, then the fuel is injected into the oven burner. After the fuel/primary air mixture flows out of the fuel ports on the tube burner, the flame will be established. The ignition time is defined as the time period from fuel injection to the time of full flame establishment (i.e., the flame is established along the entire length of the burner tube). ANSI Z21.1 requires that the oven burner ignition time should be within 4 seconds for safety reasons.



(a) Oven burner configuration and ignition process



(b) Oven burner ignition time



0s *1.75s* *2.36s* *4.22s*

(c) Oven burner flashback at 30% hydrogen addition

Figure 41: Ignition performance of the oven burner

The ignition time of the oven burner operating on different fuel mixtures is shown in Figure 41 (b). Following ANSI Z21.1, five ignition tests are conducted under both hot and cold conditions.

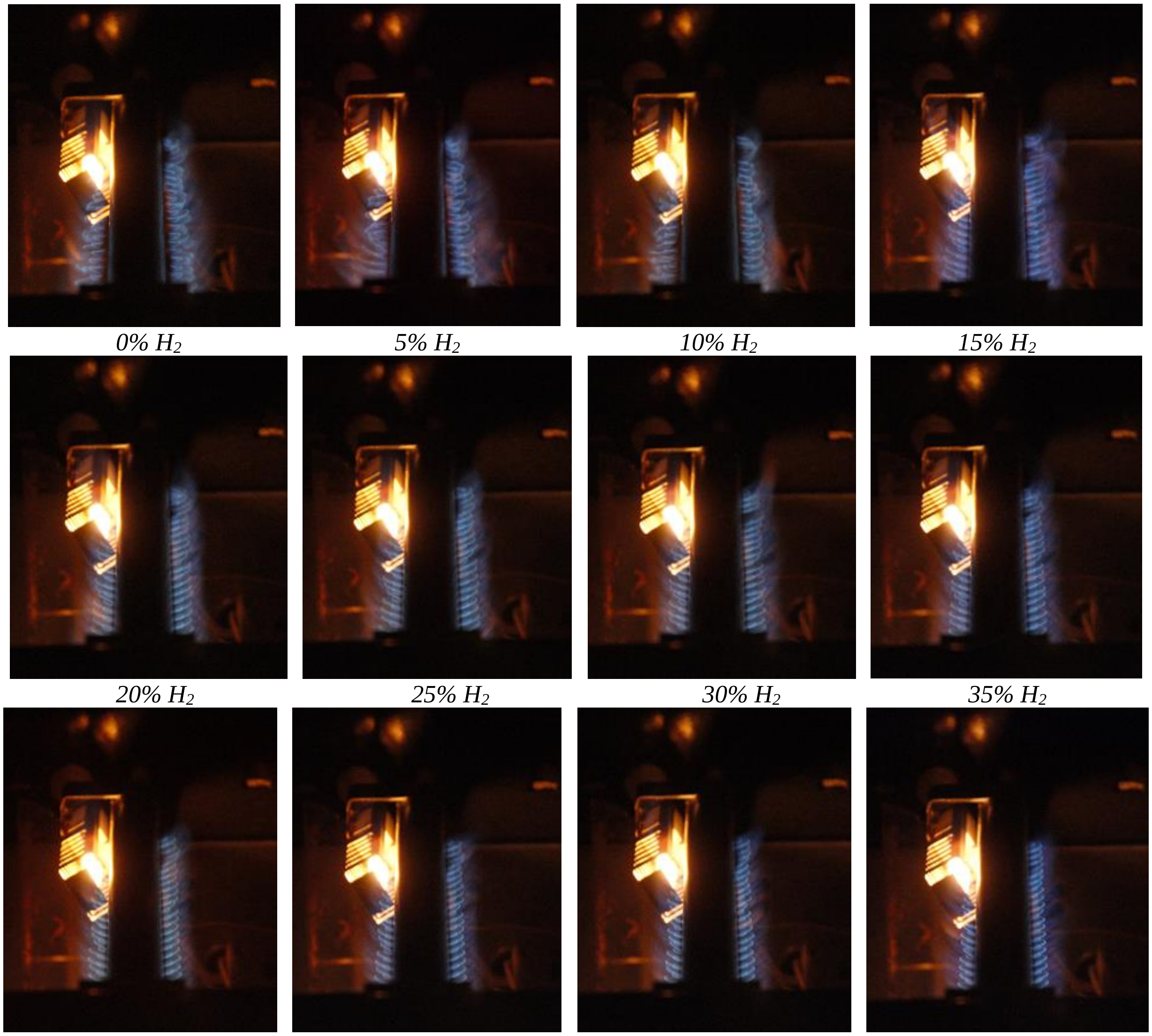
Cold ignition is conducted with the oven starting at room temperature. Hot ignition is the re-ignition after the burner has been operating for three minutes, shutdown, and then reignited. Reignition occurs many times during the cooking process as the burner turns on to maintain the set oven temperature. As shown in Figure 41 (b), with up to 10% hydrogen addition, the ignition time under both cold and hot conditions remains around 1.75s. With more than 10% hydrogen addition, the ignition time decreases. The decrease is greater under hot ignition conditions. Under cold ignition condition, the ignition time exhibits larger variation. Generally speaking, adding hydrogen into the fuel helps decrease the ignition time. However, flashback upon ignition occurs once the fuel contains more than about 25% hydrogen. This process is shown in a time sequence of images in Figure 41 (c). At first, the flame establishes very quickly, then the flame starts to flashback into the tube burner. Two seconds after the burner is ignited, the flame migrates from its original location and propagates into the tubular oven burner while producing a loud “pop” noise. Afterwards, the flame remains stabilized within the tube and emerges through the burner flame ports. An oscillating orange-blue color flame is established due to the soot formation from hydrocarbon rich combustion. Based on measured flashback limits, therefore, the upper limit for hydrogen addition is about 25% by volume.

There are two reasons causing the flashback of the oven burner: 1) the fuel/air mixture in the oven burner falls within the flammable range 2) The flame propagates faster than the fuel/air mixture flow velocity. In methane/air mixtures, if the volumetric percentage of methane is between 5% and 15%, the mixture will be flammable. However, this flammable range can be extended to 4% to 75% when it comes to hydrogen. Usually, the fuel/air mixture in appliance burners is fuel rich, so the equivalence ratio is either above or around the upper flammable limit. However, as more hydrogen is added into the fuel, the fuel/air mixture in the burner tends to fall into the flammable range. In the ignition process, the oven burner is filled with air before the fuel flows in. Therefore, the equivalence ratio can fall below the upper flammable limit easily and it is easier for flashback to occur at this situation. Moreover, the Reynolds number of the mixture flow in the oven burner is around 3500, which is in the transitional range between laminar and turbulent flow. The unsteadiness in the ignition procedure may induce extra turbulence in the flow field, and it might cause flame speed increase then induce flashback. Therefore, ignition process is more easily to cause flashback than steady operating conditions for the oven burner.

5.2.2.3 Flame characteristics and operability

Ignition is an unsteady process which adds uncertainty to the determination of the flashback limits. To investigate other performance features at higher hydrogen percentage conditions, ignition is completed with pure natural gas, then hydrogen is gradually added into the fuel. Under this steady operating condition more than 55% of hydrogen can be added into the fuel without inducing flashback.

Oven burner flames with different amounts of hydrogen added in the fuel are shown in Figure 42. The flame lift distance decreases as more hydrogen is added into the fuel. Without any hydrogen added, the flame is obviously lifted from the burner. With 10% hydrogen in the fuel, the flame shapes look similar to the natural gas flame with an obvious flame lift distance. However, with more than 10% hydrogen addition, the flame starts to attach to the tube burner and the flame length gets shorter. Figure 42 shows the flame images of 0% to 55% hydrogen flames.



$0\%\ H_2$ $5\%\ H_2$ $10\%\ H_2$ $15\%\ H_2$

$20\%\ H_2$ $25\%\ H_2$ $30\%\ H_2$ $35\%\ H_2$

$40\%\ H_2$ $45\%\ H_2$ $50\%\ H_2$ $55\%\ H_2$

Figure 42: Oven burner flame characteristics

As more hydrogen is added into the fuel, the flame stabilizes closer to the burner surface, and part of the burner glows red from overheating as shown in Figure 43. When the oven burner operates on natural gas, the burner surface temperature is much lower than the flame temperature because the flame is lifted from the burner surface. However, as more hydrogen added in the fuel, the high flame speed brings the flame closer to the burner, and burner parts in close physical proximity to the flame are over heated. The oven burner is not a standard burner, therefore, the flame ports are not identically manufactured. This results in uneven burner surface temperature distribution, which will likely shorten the life of the oven burner.

Figure 43: Over-heated oven burner part at 20% hydrogen addition

5.2.2.4 Emissions

Exhaust dilution has significant influence on reported emission measurements. If the exhaust is diluted before it is sampled for analysis, the absolute level of the emission species might drop significantly, which brings bias in the emission measurement. Therefore, oxygen correction is usually adopted to eliminate the influence of air dilution. However, this correction method of evaluating emission levels also has some resolution challenges. As shown in Equation (18), as the oxygen level in the exhaust approaching 20.9%, the uncertainty in this equation will increase (based on ability of the oxygen analyzer to report the oxygen level precisely) as the denominator approaches zero. Therefore, even if correction method can be used in the exhaust evaluation, dilution of the exhaust should be avoided. However, most of the residential appliances use open-air combustion technology, including the oven burner in this study. It is inevitable that the exhaust is going to be diluted in the combustion or sampling process due to the open-air combustion situation. Hence, it is necessary to avoid dilution in the sampling process.

Two main methods for sampling the exhaust of the oven: 1) using an exhaust hood to collect emissions and 2) taking the sample directly from the oven flue collar located at the range top surface. In this study, both sampling methods are used and compared with each other. The raw and corrected concentrations of NO and O_2 concentrations at different locations are shown in Figure 44. As expected, the oven burner operates in cycles. The oven burner turns on after it is given a temperature setting. After a while, around 6 minutes in this case, the temperature in the oven reaches the target temperature and the burner turns off automatically. Afterwards, the oven burner will turn on and off to keep the temperature in the oven at the target temperature, so the emissions are periodic in nature. It can be seen that NO emission level at the exhaust hood is around 10 ppm by the end of the first burn, and the O_2 concentration in the exhaust is around 19%. Under this condition, the exhaust is highly diluted. The red dashed line represents the NO emission sampled at the flue collar, without the dilution from the surrounding air. The emission level by the end of the first burn is around 35 ppm with O_2 concentration around 15%. In contrast, the open-air results have significantly different results. However, O_2 correction helps rectify this. As shown, the red and blue solid lines are the 3% O_2 corrected NO emission levels for the two sampling locations. The corrected emission levels are similar for the first burn, although the oxygen level is about 4% higher for the hood sample due to added dilution for that case. These results illustrate the importance of correcting the sample to a constant level of O_2.

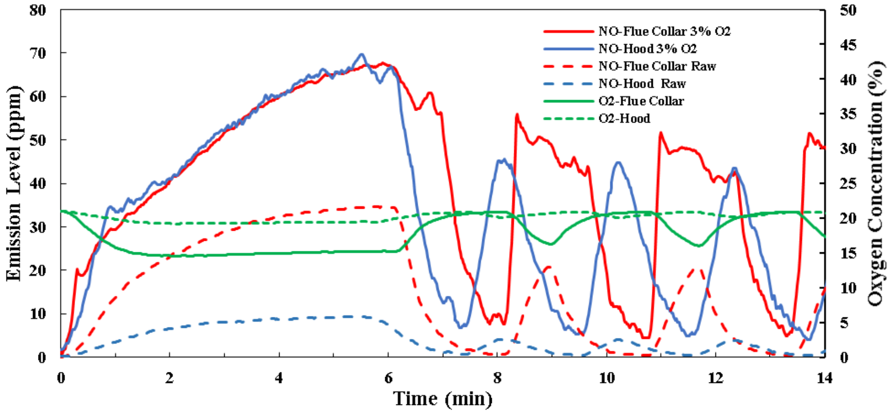
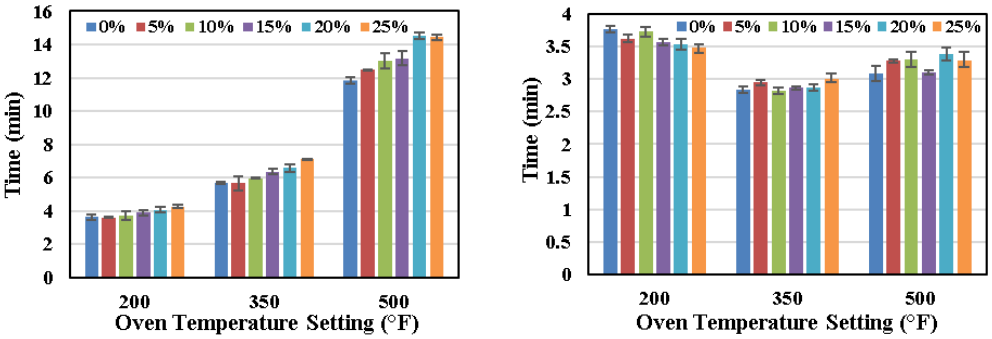


Figure 44: NO and O_2 concentrations at different sampling locations of the oven

The periodic behavior is an important attribute of the oven burner which adds significant dynamics to the emission measurement interpretation. This behavior is examined in more detail using the results from samples taken from the exhaust collar location. Figure 45 (a) shows the oven first burn operating time periods under different temperature settings and different fuel mixtures. To simulate the actual operating condition of an oven burner, the burner is covered by the metal plate and the oven door is closed. As a result, no visual access to the oven burner is available. Therefore, the burner "first burn operating time" is defined as the time from the start of the oven to the time NO emission reach their first peak level. As shown, as the oven burner temperature setting increases from 200 °F (93 °C) to 500 °F (260 °C), it takes longer time to reach the target temperature. For each temperature setting, as more hydrogen is added into the fuel, the first burn time period increases slightly due to the heating load decrease associated with the hydrogen. For example, when the temperature setting is 350 °F (177 °C), the first burner time period increases from 5.7 min to 7.1 min. Figure 45 (b) shows the cyclic burn time periods at different temperature settings. The cyclic time period is defined as the time period between two NO peaks under the cyclic operating condition. This time period includes the combustion and the non-combustion periods. This time period doesn't vary much under different conditions.



(a) First burn time periods *(b) Cyclic burn time periods*

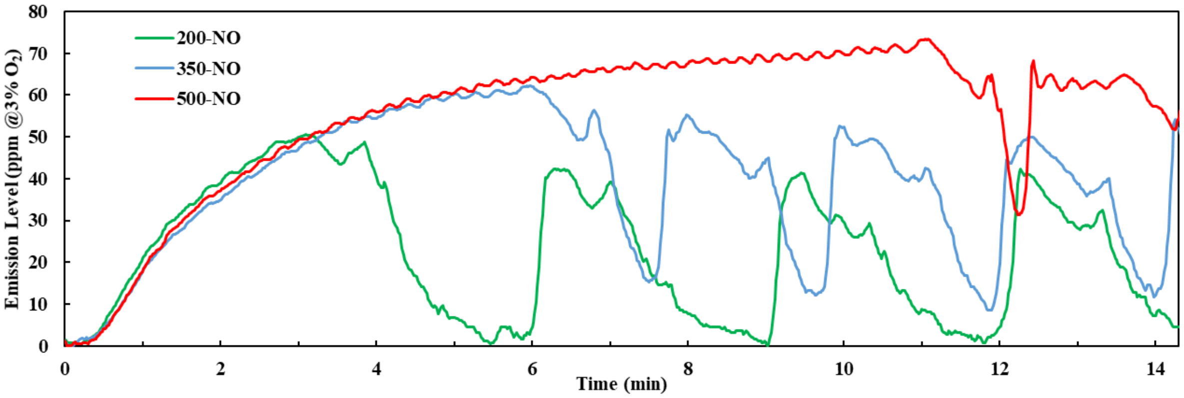
Figure 45: Oven operating time periods

Note that the emissions are also influenced by temperature settings. NO and CO emissions under different oven temperatures settings are shown in Figure 46. The temperature setting influences the emission test results in two ways:

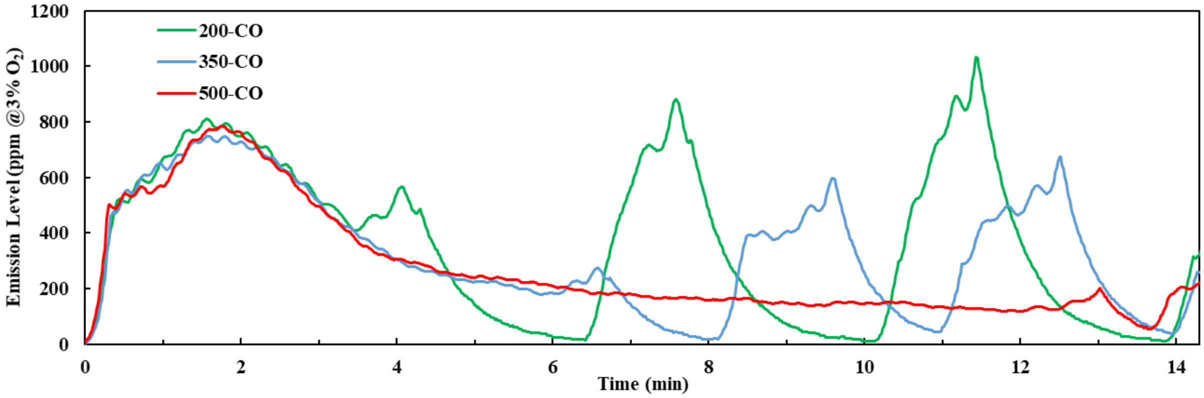
1) When the set temperature is relatively low, e.g., 200 °F (93 °C) in this study, the first burn ends before the combustion exhaust fills the oven capacity. Under this situation, the emissions are diluted by the existing air in the oven. Therefore, this emission level can’t be used to present the actual emission level of the oven burner.

2) When the oven temperature setting is relatively high, e.g., 500 °F (260 °C) in this study, the emission level reaches a relatively stable level, which means the gas volume within the oven (and exiting the flue sleeve) has reached steady dilution. However, as shown, the NO emissions continue to climb as CO emission continue to decrease with time. This is due to the temperature increase in the oven. With the temperature increasing, the thermal NO_X increases, however, high temperature helps oxidize CO into CO_2, so these two species see different trends.

It is noted that 350 °F (177 °C) is a very common oven temperature setting. As shown, the emission trend under this operating condition is between 200 °F (93 °C) and 500 °F (260 °C). It is not clear if the dilution effect has stabilized by end of the first burn for this set point. For the cyclic emissions, NO emissions are approaching the first burner level, but before that happens, the burner turns off. However, CO emissions of the cyclic periods are higher than the first burn due to the ignition and turn-off process. This type of periodic burner is widely adopted in residential appliances, including room furnaces, storage water heaters, etc.. Therefore, it is of great interest to understand its working principle to make reasonable emission measurement standard and regulations for policy makers.



(a) NO emissions under different temperature settings



(b) CO emissions under different temperature settings

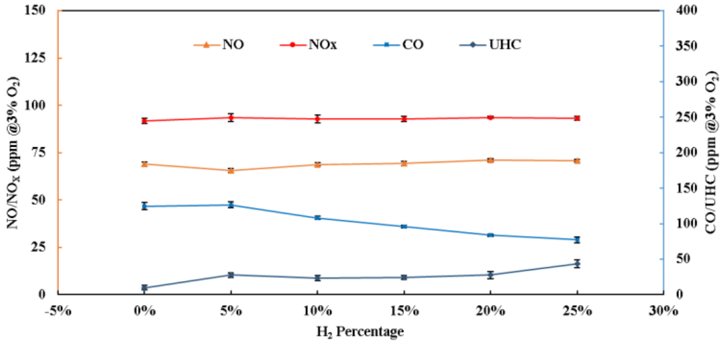
Figure 46: Oven emissions under different temperature settings

Due to the complex cyclic behavior, it is difficult to define an appropriate emission value. In the present study, to eliminate the dynamic associated with the oven burn period, the average emission level of last minute of the first burn under 500 °F (260 °C) is chosen to represent the oven emissions. For this temperature setting, the oven burner operates long enough for the combustion product to fill the whole oven capacity, and the analyzer readings also reach a relatively steady level.

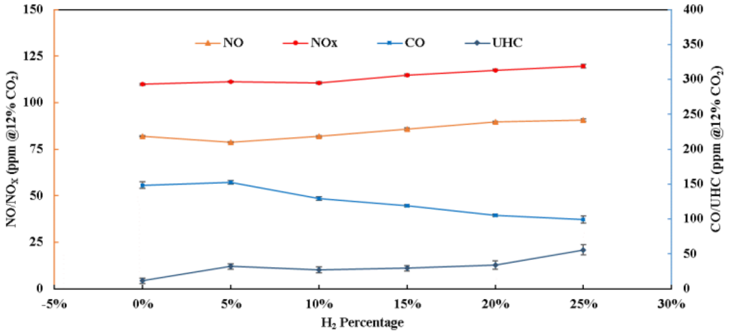
The emission results for the oven burner are presented in Figure 47. For pure natural gas, the total NO_X ($NO+NO_2$) emission is 92 ppm (3% O_2) and 75% of that is NO. NO_X emissions do not change much as hydrogen added into the fuel. CO emissions are 125 ppm (3% O_2) when the oven burner is operating on natural gas, and 5% hydrogen addition doesn’t change these levels significantly. When the hydrogen percentage in the fuel increases above 5%, the CO emissions steadily drop, reaching 77 ppm (3% O_2) with 25% hydrogen addition. UHC emissions are around 10 ppm (3% O_2) when the burner is operating on natural gas, and increase up to around 25 ppm (3% O_2) as more hydrogen added into the fuel. This level stays constant until hydrogen percentage approaches flashback conditions. At 25% hydrogen addition, UHCs reach 44 ppm (3% O_2). Figure 47 (c) presents the results on a calorific correction result, and these emission trends follow those exhibited by the oxygen correction basis as shown in Figure 47 (a).

As shown in Figure 47 (b), the emission levels based on carbon dioxide correction show some small differences compared to other cases. Due to the significant change in CO and UHC, these two species variations follow the same trend as the other two correction methods. However, NO and NO_X emissions show an increasing trend using the 12% CO_2 correction basis. This phenomenon is caused by the correction method itself. As can be seen in Equation (19), CO_2 concentration of the exhaust is in the denominator. This value decreases as more natural gas is replaced by hydrogen. Therefore, the CO_2 correction basis has an intrinsic property of increasing emission levels when using carbon displacing fuels such as hydrogen.

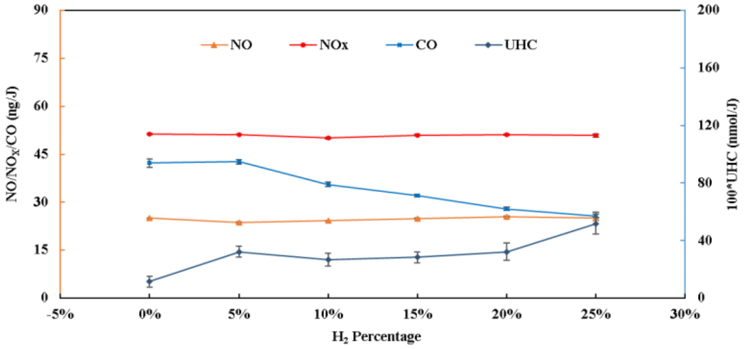
Finally, while not shown explicitly, it is noted that the measured N_2O and NH_3 emission levels where negligible in the context of the measurement accuracy of the MEXA 1400 QCL analyzer.



(a) 3% O_2 correction



(b) 12% CO_2 correction



(c) Calorific correction

Figure 47: Emissions of the oven burner operating on NG/H_2 mixtures

5.2.2.5 Primary air entrainment

Primary air entrainment behavior is an important property of self-aspirating flames including the oven burner flame in this study. Multiple air/fuel ratio correlations have been developed to predict the entrainment ability of a fuel stream thus guiding the burner design and optimization. Equation (21)-(23) [6] are examples which are used in the present study to compare with the experimental results. $\rho_{\text{fuel}}/\rho_{\text{air}}$ is the fuel specific density; A_p is the burner port (cross-section) area; A_j is the fuel jet orifice area; T_0 is the reference temperature (15 °C); T_1 is the fuel/air mixture temperature before combustion.

$$V_{air}/V_{fuel} = \sqrt{\frac{\rho_{fuel}}{\rho_{air}}}\left(\sqrt{\frac{A_p}{A_j}} - 1\right) \qquad \text{Equation (21)}$$

$$V_{air}/V_{fuel} = \sqrt{\frac{\rho_{fuel}}{\rho_{air}}}\left(\sqrt{\frac{A_p T_0}{A_j T_1}} - 1\right) \qquad \text{Equation (22)}$$

$$V_{air}/V_{fuel} = -\frac{\left(1+\frac{\rho_{fuel}}{\rho_{air}}\right)}{2} + \sqrt{\frac{\rho_{fuel}}{\rho_{air}}\frac{A_p}{A_j} * (0.52\sim0.65)} \qquad \text{Equation (23)}$$

The comparison of experimental data and correlations is shown in Figure 48. The experimental data points of primary air/fuel ratio fall on top of Jones's experimental correlation. Both of the theoretical correlations over-estimate the primary air/fuel flow ratio of different fuel mixtures. Although Prigg's theoretical correlation successfully predicts that the temperature increase of the mixture reduces the entrained air flowrate, it fails to predict the air entrainment behavior due to the theoretical assumption itself. In Jones's work, the entrainment equation is derived starting with the Bernoulli equation, and practical factors like friction loss are incorporated into an empirical correction factor. The value of the empirical factor varies from 0.52 to 0.65. In this study, because the air shutter controlling the aeration port area is not fully open, the lower value of 0.52 is chosen as the entrainment factor in this study. Therefore, it can be concluded that Jones's experimental correlation can more accurately predict the primary air entrainment behavior of the oven burner compared to other correlations as long as the correct coefficient is used.

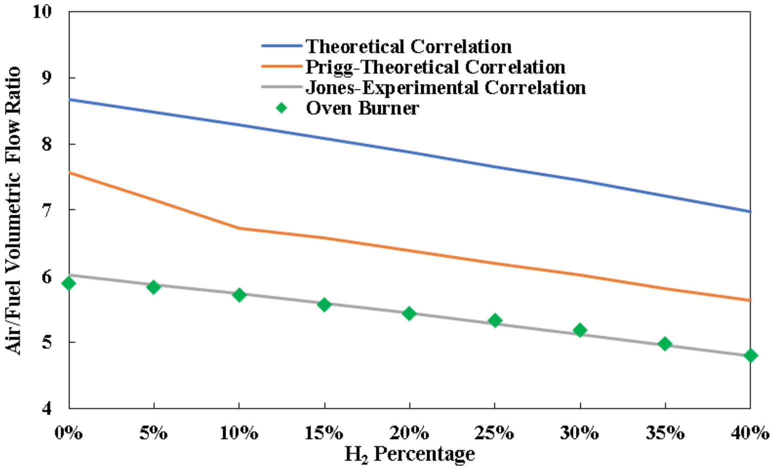


Figure 48: Oven burner primary air entrainment correlation

It is essential to understand the primary air entrainment behavior, especially for burner design or optimization. Extensive fundamental combustion studies have been conducted on various fuel/air mixtures, and all of them indicate that hydrogen addition can result in flame speed increase and flammable range extension. When the oven burner is operating on natural gas, the equivalence ratio of the fuel/air mixture is 1.55, which is around the upper limit of natural gas/air mixtures. As shown in Figure 48, the ability of the fuel mixture entraining primary air decreases as more hydrogen is added into the fuel, which should induce a richer fuel condition that helps decrease the flashback risk. However, as hydrogen percentages increases to 40%, the equivalence ratio decreases down to 1.34, which is within the flammable range. This effect is due to the lower air/fuel ratio at stoichiometric combustion condition of hydrogen. The low requirement of primary air to oxidize hydrogen and hydrogen’s wide flammability range bring challenges to decrease the flashback risk at hydrogen-rich conditions. It is also noteworthy that the flame ports on the oven burner are 4.5 mm by 1 mm rectangular slots. Between every two slots, there is a circular hole with the diameter of 1.5 mm working as the conjunction fuel port. The quenching effect of these small ports can help prevent flashback from happening, even if the fuel/air mixture in the burner is within the flammable range. The quenching distance of natural gas is 2.5 mm (φ=1, atmosphere pressure, room temperature). Therefore, when the oven burner is operating on natural gas, it is nearly impossible for the flame to propagate into the burner. However, the quenching distance of hydrogen is only 0.64 mm, which makes flashback easier to occur when the burner is operating on hydrogen-rich fuels.

5.2.2.6 Other performances

Combustion noise is measured using an Extech-407762 sound level meter. The sound meter is located at a fixed distance adjacent to the oven while keeping the oven door open. As shown in Figure 49, the background sound level in the lab is 60.7 dB. When the burner is turned on, the noise level fluctuates between 61 to 62 dB with up to 30% hydrogen in the fuel. However, the noise level exceeds 62 dB when more than 35% hydrogen is added into the fuel. It is noted that, with more than 40% hydrogen in the fuel, an obvious tonal “buzz” noise is emitted which is associated with the high volumetric flowrate of the fuel and the flame dynamics near the flame ports.

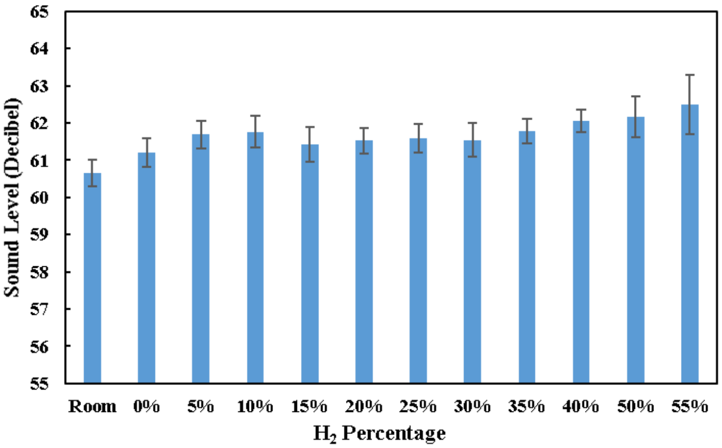
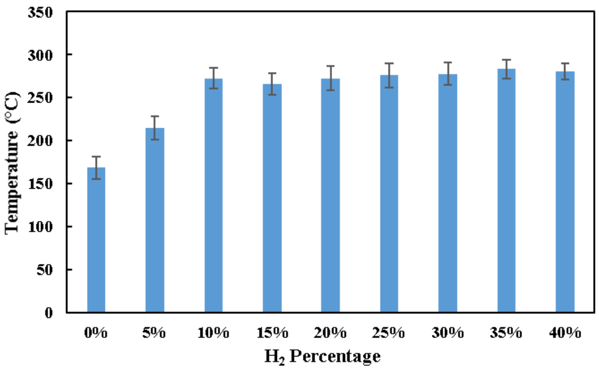


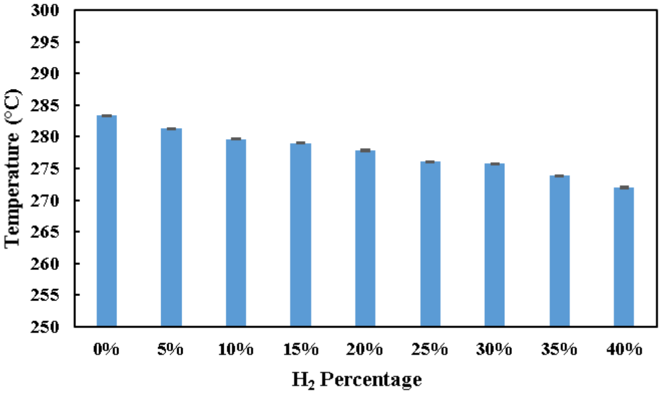
Figure 49: Combustion noise of the oven burner

To investigate the influence of the flame position on the burner, a FLIR-T640 infrared camera is used to measure the burner surface temperature. The area close to the burner flame ports is selected as the region of interest. The burner temperature results are shown in Figure 50 (a). Under pure natural gas operating condition, the burner temperature is around 169 °C (336 °F). With 5% hydrogen added into natural gas, the burner temperature reaches 215 °C (419 °F). As more hydrogen is added, the burner temperature stays at around 275 °C (527 °F). Although the adiabatic flame temperature of pure hydrogen is only around 5% higher than that of natural gas, the burner temperature increases by striking 63% at only 10% hydrogen addition into natural gas. This distinct temperature rise is mainly caused by the change in flame position, which may reduce the burner’s durability.

For remaining results reported, the oven bottom plate was reinstalled. As a first step, the plate average temperature was documented as a function of amount of hydrogen in the fuel. As shown in Figure 50 (b), the plate temperature decreases from 283 °C (541 °F) to 272 °C (521 °F) as the hydrogen percentage in the fuel increases from 0% to 40%. Two factors contribute to the temperature decrease: heating load decrease and the change in the flame position. Due to the Wobbe Index decrease, the heating load of the oven burner decreases from 5.3 kW (18,000 Btu/hr) to 4.8 kW (16,250 Btu/hr). The other factor influencing the metal plate temperature is the flame position. As shown in Figure 42, the flame position gets closer to the burner while hydrogen percentage in the fuel increasing. This effect results in the flame being located farther from the metal plate surface, which also contributes to the plate temperature decrease.



(a) Oven burner temperature at different hydrogen percentage



(b) Metal plate temperature at different hydrogen percentage

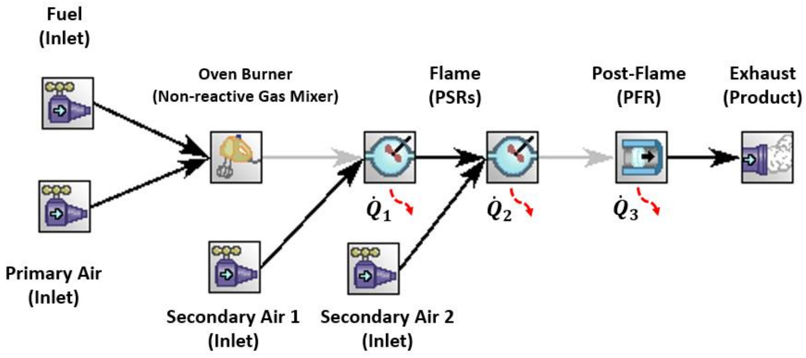
Figure 50: Oven burner and metal plate temperature

5.2.2.7 Chemical reaction network

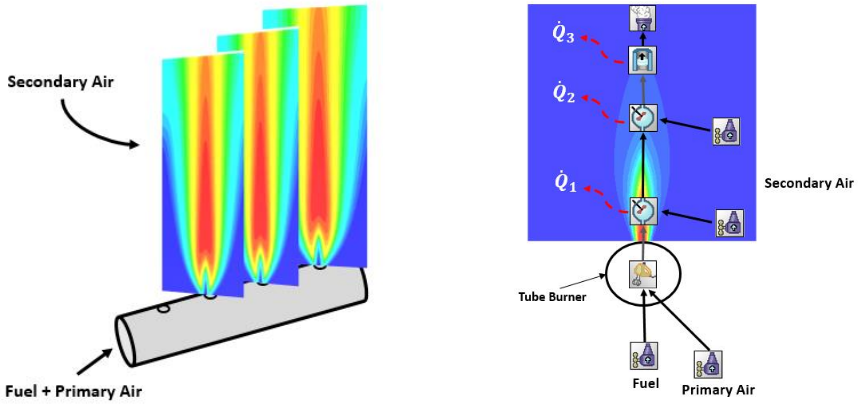
A chemical reaction network (CRN) model is generated based on the flame appearance, as shown in Figure 51. Fuel and primary air are first mixed together in a non-reactive gas mixer, which is used to simulate the oven tube burner. At pure natural gas operating condition, the equivalence ratio of this mixture is 1.55 according to the experiment tests. The combustion doesn't take place within the tube burner. Therefore, a non-reactive gas mixer is adopted before the reactors. Afterwards, two perfectly stirred reactors (PSRs) are adopted to simulate the flame. The first stage of the secondary air is to simulate the primary air from the area below the oven flame, and the second stage of the secondary air is to simulate the dilution air from the openings on the oven wall.

The heat loss $\dot{Q}_1$ from the PSR is 0.33 kJ/s, which is to simulate the heating process of the metal plate over the oven burner, the air in the oven and also the radiation loss. The smaller heat loss $\dot{Q}_2$ is 0.1 kJ/s, which is mainly to simulate the air heating process.

A plug flow reactor (PFR) is used to simulate the post-flame region. The heat loss $\dot{Q}_3$ is due to hot products heating the surrounding air and the oven itself. The heat loss rate is 0.07 kJ/(cm*s).



(a) CRN model for the oven burner



(b) CFD-CRN model for the oven burner

Figure 51: Numerical model of the oven burner

The CO/NO_X emission results comparison are shown in Figure 52. The blue color and red color present CO and NO_X emissions respectively. The dots stand for experimental results and the lines are for simulation. It should be noted that the calorific correction uses NO_2 as the base, which means NO is converted to NO_2 in the molecular weight calculation.

As more natural gas is replaced by H_2, experimentally measured CO emission decreases steadily from 125 ppm (@3% O_2) to 77 ppm (@3% O_2) with H_2 percentage increasing from 0% to 25%, which trend is captured by simulation very well.

The experiment shows that NO_X stays at a constant level around 93 ppm (@3% O_2). However, simulation result shows a steady decreasing trend of NO_X emission from 74 (@3% O_2) to 33 ppm (@3% O_2). This difference is believed to be caused by the heat loss deviation between simulation and experiment. The increase of hydrogen percentage usually causes the heating load decrease thus decreases thermal NO_X. However, the working principle of the oven is distinct from an open flame. The combustion process takes place in a confined space and won't stop until the oven is at the target temperature. Therefore, the flame temperature stays at a constant level even if the heat release rate is different.

As more H_2 is added into natural gas, the flame gets closer to the burner until the flame flashes back into the burner. For the oven burner tested in this study, flashback occurs at 30% H_2 injection according to the experiment result.

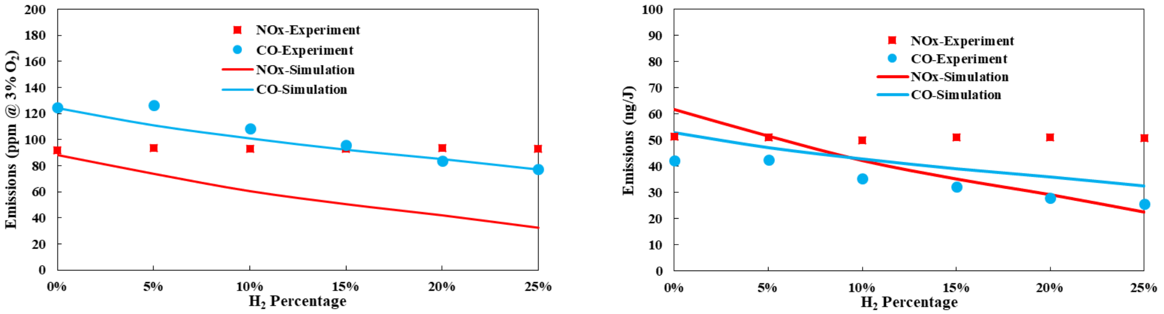


Figure 52: CO/NO_X emissions comparison between simulation and experiments of the oven burner

5.2.3 *Other applications*

Besides cooktop and oven burners, there are numerous other burner types on market utilizing self-aspirating technologies.

For example, broiler is another common burner type in residential houses, which has a similar or identical design as the oven burner. It is usually mounted at the top of the oven, which is to broil the exposed surface of the food. Most of the cooking burners are designed to operate on blue flames, since yellow flame tips or yellow flames indicate incomplete combustion which might produce noxious CO emissions and endanger end users. However, outdoor grill burners are slightly different than other burners and they are supposed to operate with yellow flame tips. The soot formation can help enhance the radiation heat transfer and might also add some extra flavor to the food.

Cooking is a complicated activity, which is more than heating the object (food) to a certain temperature. Sometimes people sacrifice the efficiency or emissions performance to achieve other purposes (smell, taste, texture, etc.). Therefore, incorporating renewable gases in cooking should also consider other perspectives besides combustion performance.

5.3 Heating Section

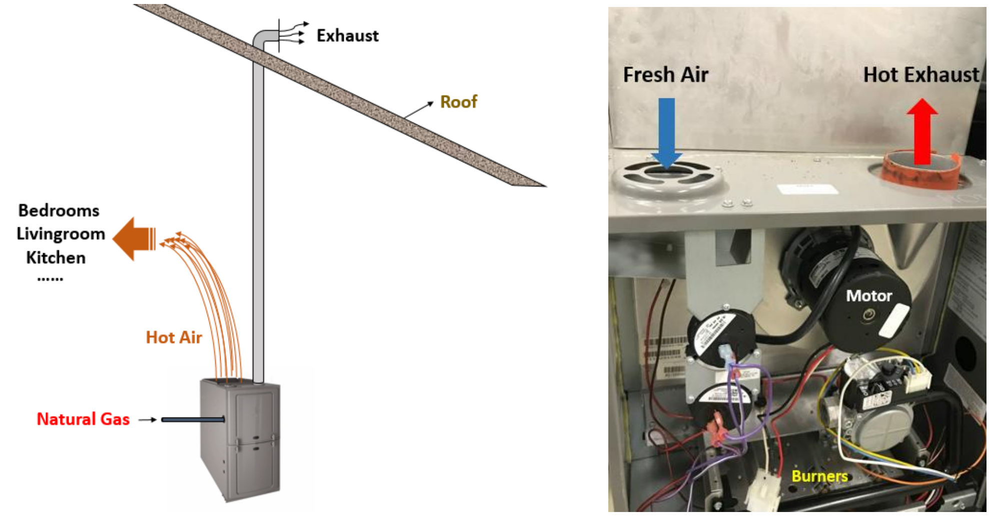
Heating is the major sector of the residential energy consumption. Space (air) heating and water heating take the highest percentage of natural gas usage in residential buildings. Therefore, understanding the working principle of the heating appliances are critical for future fuel advancement from natural gas to renewable gases.

5.3.1 *Room furnace*

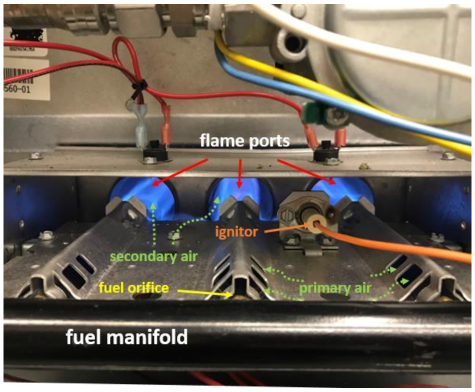
5.3.1.1 Burner configuration

A central air heating furnace is a common space heating appliance in residences. Figure 53 (a) demonstrates the installation diagram of a residential space heater furnace, which is regulated by ANSI Z21.86 [165]. The heater is usually located in the basement or a special cabinet for air heating devices and connected to the natural gas pipeline. As the natural gas is burning in the furnace, the hot exhaust goes through a non-contact heat exchanger and then is vented out of the house via and exhaust duct. The heated air from the furnace is distributed by air pumps to different spaces in the house, like bedrooms, living room, kitchen, etc..

In this study, multiple room furnaces in the North America market were investigated and their working mechanisms and burner configuration were compared with each other. Most of the room furnaces have very similar working modes and burner configurations. Therefore, a representative room furnace with the heating load ranging from 15.4 kW (52,500 Btu/hr) to 22.0 kW (75,000 Btu/hr) is selected as a representative space heating appliance to conduct experiments with. Figure 53 (b) shows the combustion system of the furnace. There are two openings on the top of the furnace, one is the combustion air inlet and the other one is the exhaust outlet. A fan forces fresh air to the combustion system and vents the exhaust into the atmosphere. The furnace burner is an aspirated burner, and the burner geometry is illustrated in Figure 53 (c). At the operating condition, natural gas first fills the manifold and then flows into the furnace burners through the orifice openings. As the natural gas flows within the furnace burners, it entrains some surrounding air into the burners through the openings on the burners. This part of the used is generally referred to as primary air. When the fuel/primary air mixture arrives at the burner exit, it is ignited by an electrical spark. As the furnace flame stabilized at the burner exit, half of the flame goes into the flame ports. Since the primary air is not sufficient to oxidize all the natural gas, secondary air is entrained into the flames at the burner exit and also into the flame ports.



(a) Room furnace setup *(b) Room furnace burner tested*



(c) Room furnace burner diagram

Figure 53: Room furnace combustion system configuration

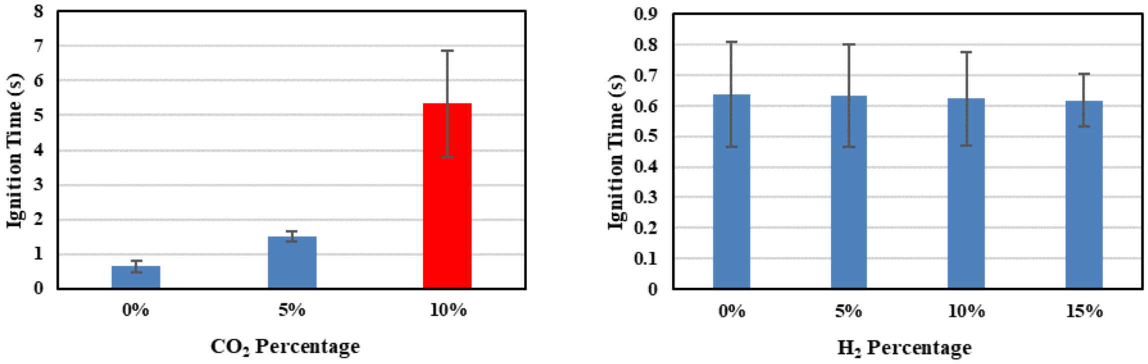
5.3.1.2 Ignition performance

Ignition time is one of the essential parameters considered in residential burner design and operation. Ignition failure can induce natural gas leakage into residential houses thus cause safety hazard. Ignition time is defined as the time from which fuel flows out of the flame ports to the time at which the flame for the room furnace burner is fully established. ANSI Z21.86 regulates that the room furnace ignition should be completed within 0.8 second. In the room furnace ignition process, the electric ignitor turns on first, and then natural gas will flow through the orifice and arrive at the ignitor. The far right burner adjacent to the ignitor will be ignited first and then the flame will propagate to the other two burners. To simulate the real-life operating condition, the flame ignition is conducted at room temperature. Under each operating condition, the ignition test is repeated five times.

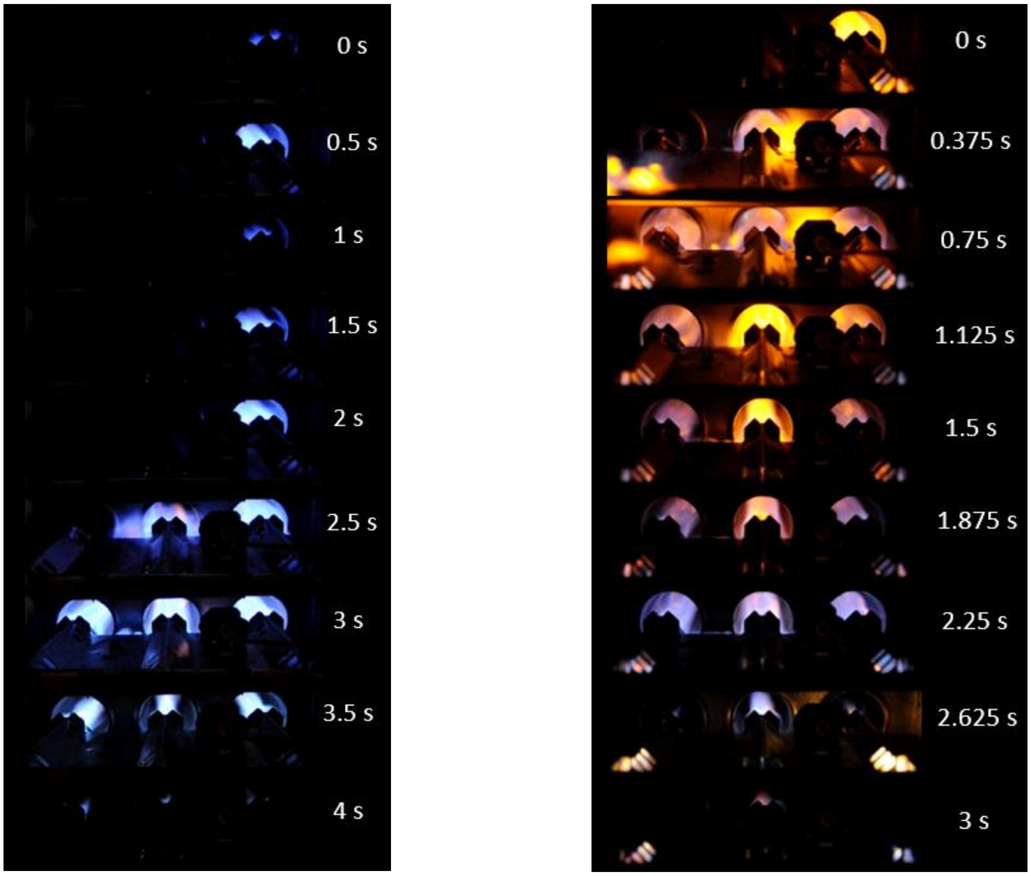
Figure 54 (a) shows the room furnace ignition time when it operates on natural gas/carbon dioxide mixtures. As the carbon dioxide percentage in the fuel mixture increases to 5%, the room furnace ignition time increases from 0.64s to 1.51s due to the inert property of carbon dioxide. When the carbon dioxide percentage in the fuel increases up to 10%, ignition failure occurs and the average ignition time goes up to 5.33s. An ignition failure process is shown in Figure 54 (c). It takes almost 2 seconds to ignite the burner at far right. Then the flame propagates to adjacent burners. However, the heat release of the carbon dioxide rich fuel is too low to maintain the flame at the burner exit, so the flame totally blows off after 4s.

For natural gas/hydrogen mixtures, flashback is the major concern as opposed to blow-off. Figure 54 (b) indicates the ignition time of the room furnace operating on natural gas/hydrogen mixtures. With up to 15% hydrogen addition, the ignition time doesn’t change much. However, the ignition time variation becomes smaller as more hydrogen is added into natural gas. However, flashback upon ignition starts to occur at 20% hydrogen addition. As shown in Figure 54 (d), the flame at the far right establishes very quickly after the fuel mixtures flows out of the manifold. However, due to the high flame speed of hydrogen rich fuel, the flame propagates within the burner all the way to the fuel orifice which can be seen from the primary air ports. Even if the flame establishes within 0.8s, part of the flame still stays within the burner which causes unsteadiness. Between 1s and 2s, the flame oscillates in and out of the burner then extinguishes after a major flashback into the burner. The flame in Figure 54 (d) appears yellowish due to the soot formation caused by the non-ideal flame location, where natural gas and primary air are not yet fully mixed. Moreover, this non-ideal flame can also destroy the burner or the whole combustion system due to overheating.

Ignition failure of the room furnace occurs at both carbon dioxide and hydrogen addition situations. It should be noted that the mechanisms of these two situations are different. Ignition failure for carbon dioxide rich fuel is because of the inert property of carbon dioxide causing the low heat release rate. However, the flashback-ignition failure for hydrogen rich fuel is caused by the fast flame propagation induced reaction field oscillation.



(a) Ignition time of NG/CO_2 mixtures *(b) Ignition time of NG/H_2 mixtures*



(c) Blow-off at 10% CO_2 addition *(d) Flashback at 20% H_2 addition*

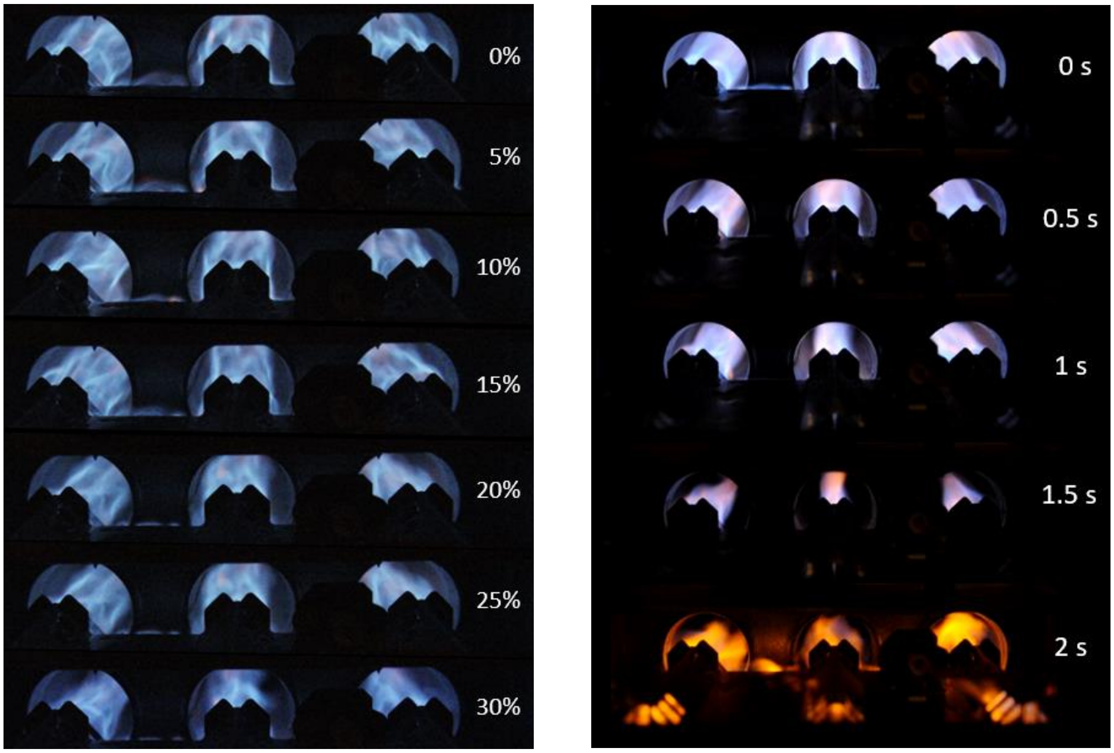
Figure 54: Ignition performance of the room furnace burner

5.3.1.3 Flame characteristics

Since ignition is an unsteady process, it adds uncertainties to the flashback behavior. To evaluate other performance attributes at higher levels of carbon dioxide or hydrogen, studies are conducted in which ignition is completed with pure natural gas and then carbon dioxide or hydrogen are gradually added into the fuel. This procedure is used to investigate burner temperature and emissions at higher carbon dioxide or hydrogen percentage conditions than those at which ignition blow-off of flashback occurs.

Images of the furnace flames with different carbon dioxide percentage are shown in Figure 55 (a). As more carbon dioxide added into natural gas, the flame diameter becomes smaller, and blow-off starts to occur at 30% carbon dioxide addition.

When the room furnace operates on natural gas/hydrogen mixtures, the flame characteristics do not change significantly. However, flashback occurs when ~45% hydrogen is added to natural gas. As the amount of hydrogen is increased, the flame stabilizes at the burner exit, and then propagates into the Venturi burner generating a notable “pop” noise.



(a) Characteristics of NG/CO_2 flames *(b) Operating flashback at 45% H_2 addition*

Figure 55: Room furnace burner flame characteristics

5.3.1.4 Emissions

The exhaust sampling probe is located at around 0.305m (1 foot) above the furnace exhaust outlet. Before the sampled exhaust is supplied to the emission analyzers, it first goes through a cooling system to remove the water. Each emission test lasts for 15 minutes after the emission level is stabilized. Since the emission analyzers output data every second, emission levels for each operating condition is based on an average of 900 individual measurements.

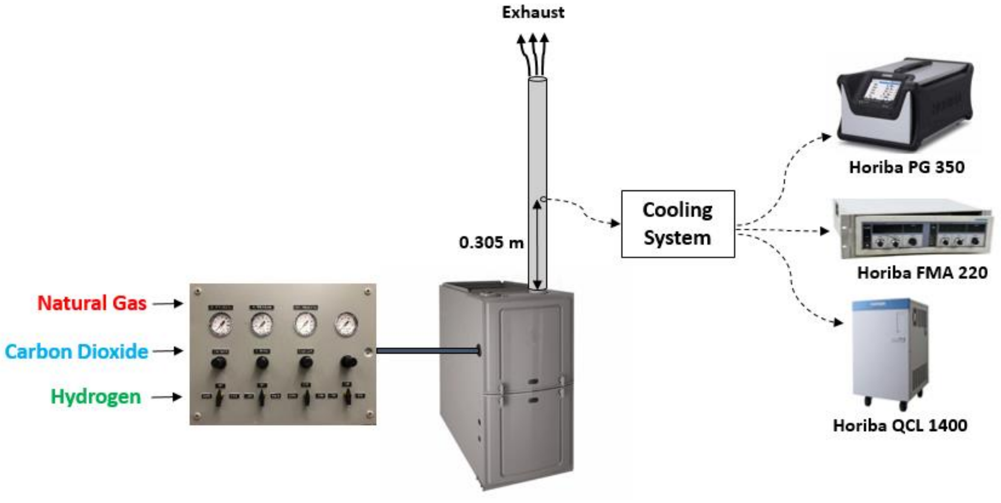
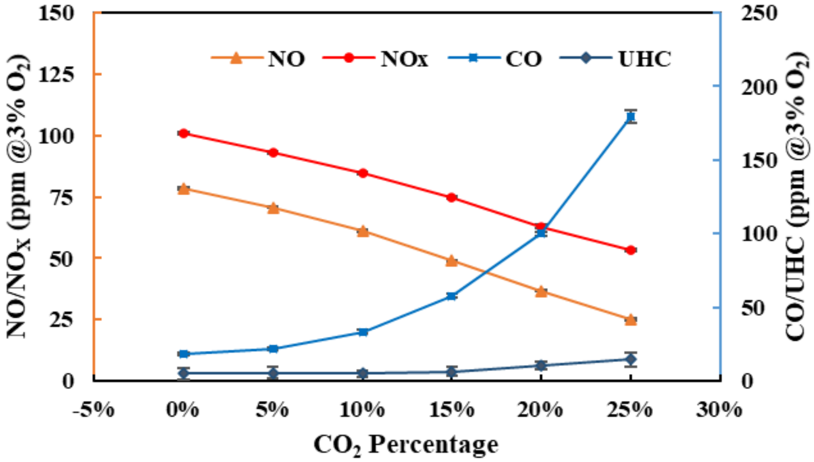


Figure 56: Room furnace emission testing setup

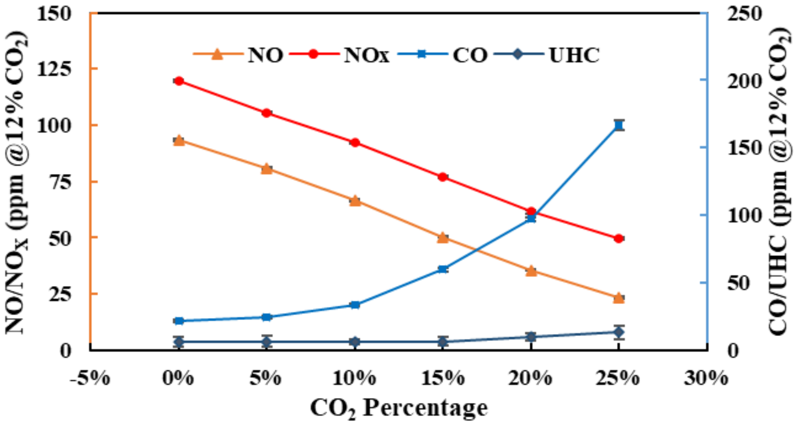
The emissions of the room furnace burner operated on various amount of CO_2 added to natural gas are presented in Figure 57. Figure 57 (a) shows the emissions under 3% O_2 reference level. At pure natural gas operating condition, the total NO_X ($NO+NO_2$) emission is 101 ppm (3% O_2) and 77% of that is NO. As more CO_2 added into the fuel, NO_X shows an approximately linear decrease. By 25% CO_2 addition, NO_X drops down to 53 ppm (3% O_2). Due to the flame temperature decrease, CO and UHC emissions increase as the CO_2 percentage in the fuel goes up. As the CO_2 addition increases from 0% to 25%, CO increases 10x from 18 ppm (3% O_2) to 180 ppm (3% O_2) and UHC emissions increase from 5 ppm (3% O_2) to 14 ppm (3% O_2).

Compared to the 3% O_2 correction method, the emission levels under other correction methods show a similar trend, which are presented in Figure 57 (b) and (c). Using the calorific correction, NO emission is converted to NO_2, which is also the reason why the difference between NO and NO_X emissions is larger than that based on O_2 and CO_2 correction methods. Note that the calorific correction in Figure 57 (c) is based on the fuel consumption (nanogram per joule of fuel consumed). However, Rule 4905 [166] and Rule 1111 [167] calorific correction methods use the heat output (nanogram per joule of heat output). To convert the calorific emission from fuel consumption-based to heat output-based, the Annual Fuel Utilization Efficiency (AFUE) needs to be incorporated. The AFUE of a room furnace is required to be measured according to ANSI/ASHRAE standard. The AFUE of a furnace or boiler stands represents its average operating efficiency which differs from the transient thermal efficiency of equipment. The room furnace tested in this study has an AFUE of 80%. When the room furnace is operating on pure natural gas, the NO_X emission level is at 39 ng per joule of heat output, which is slightly lower than the old Rule 1111 regulation.

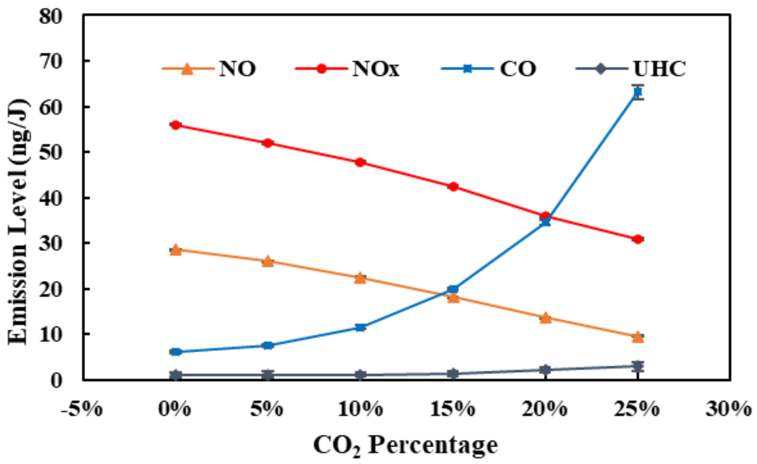
Finally, it is noted that the N_2O and NH_3 emission levels of the room furnace burner are below minimum detectable limits of the MEXA 1400 QCL analyzer measurement accuracy.



(a) 3% O_2 correction



(b) 12% CO_2 correction

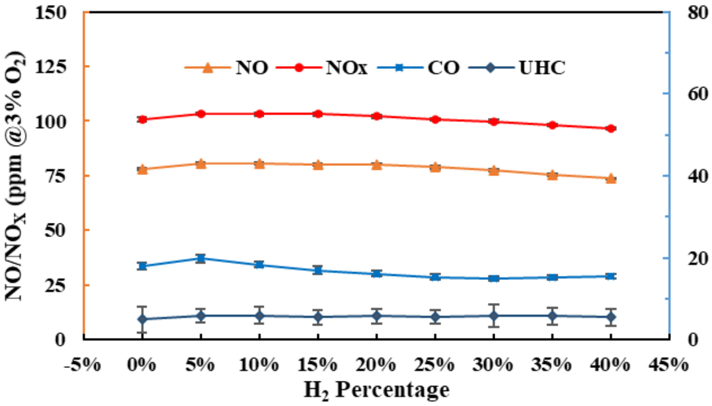


(c) Calorific correction

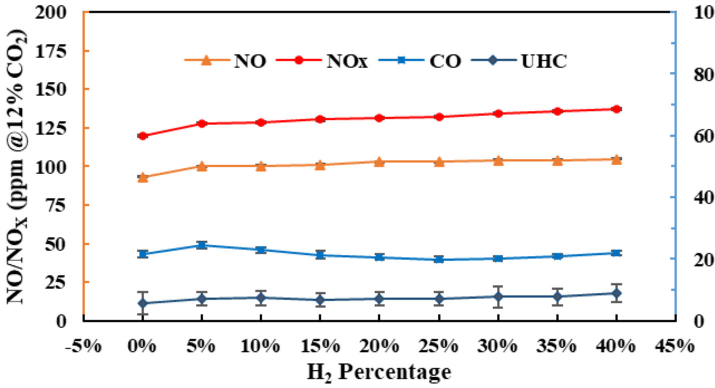
Figure 57: Room furnace burner emissions operating on NG/CO_2 mixtures

Room furnace emissions of natural gas/hydrogen mixtures are shown in Figure 58. Compared to the emissions of the room furnace operating on natural gas/carbon dioxide mixtures, hydrogen addition into natural gas doesn't change the emissions significantly. As shown in Figure 58 (a), NO_X emission drops slightly from 101 ppm (3% O_2) to 97 ppm (3% O_2) as hydrogen percentage in the fuel increases from 0% to 40%. CO decreases from 18 ppm (3% O_2) to 15 ppm (3% O_2) and UHC stays at a constant level around 5 ppm (3% O_2). The calorific correction in Figure 58 (c) shows a similar trend to the 3% O_2 correction of Figure 58 (a).

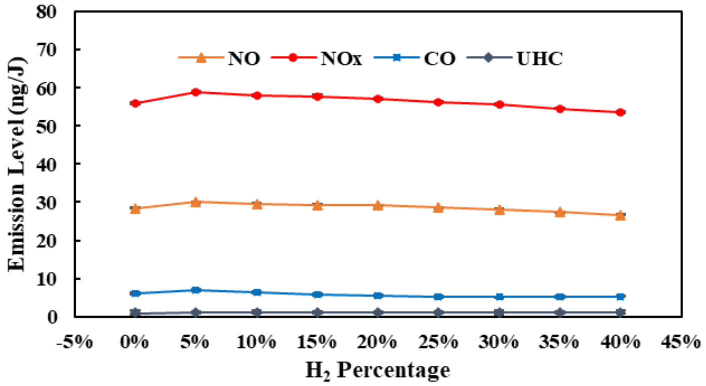
However, Figure 58 (b) shows a distinct increasing trend of several emission species. For example, NO_X emission increases from 120 ppm (12% CO_2) to 137 ppm (12% CO_2) with the hydrogen percentage in the fuel increasing up to 40%. This increasing trend is due to the intrinsic property of the CO_2 correction method. As shown in Equation (19), the CO_2 concentration is in the denominator. This value drops as more natural gas is replaced by hydrogen. Therefore, the emission level under CO_2 correction method shows a slight increasing trend. This should be considered when reporting emissions for hydrogen natural gas mixtures. In the extreme case, for example, as the hydrogen percentage in the fuel gets close to 100%, the CO_2 percentage in the exhaust will be close to the atmospheric level which results in the emission level going to infinity following Equation (19).



(a) 3% O_2 correction



(b) 12% CO_2 correction

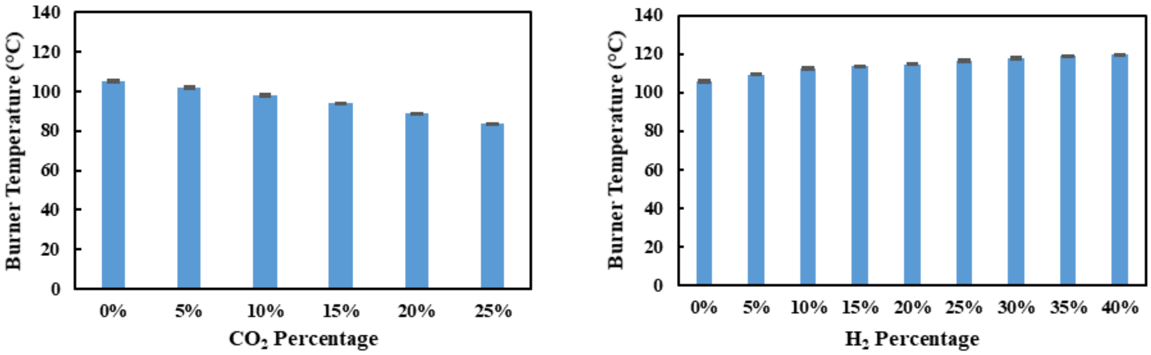


(c) Calorific correction

Figure 58: Room furnace burner emissions operating on NG/H_2 mixtures

5.3.1.5 Other performances

The burner tip temperature is measured by a FLIR-T640 camera. The burner tip temperature variation with different fuel species is shown in Figure 59. As shown in Figure 59 (a), the burner temperature decreases from 105 °C to 84 °C as the carbon dioxide percentage in the fuel increases from 0% to 25%. The heating load of the room furnace decreases from 19.0 kW to 12.0 kW due to the Wobbe Index decrease, as the carbon dioxide percentage in the fuel increases from 0% to 25%. Figure 59 (b) shows the burner temperature change when it is operating on natural gas/hydrogen mixtures. Although the heating load of the furnace decreases from 19.0 kW to 17.1 kW, the burner temperature increases from 105 °C to 120 °C. This is mainly caused by the flame getting closer to the burner exit, due to the higher flame speed of hydrogen. This temperature increase might cause burner material degrade when the room furnace operates on hydrogen rich fuels.



(a) Burner temperature of NG/CO_2 mixtures *(b) Burner temperature of NG/H_2 mixtures*

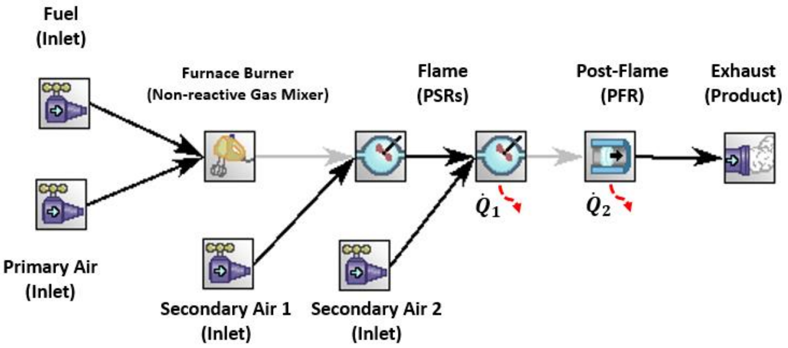
Figure 59: Room furnace burner temperature

5.3.1.6 Chemical reaction network

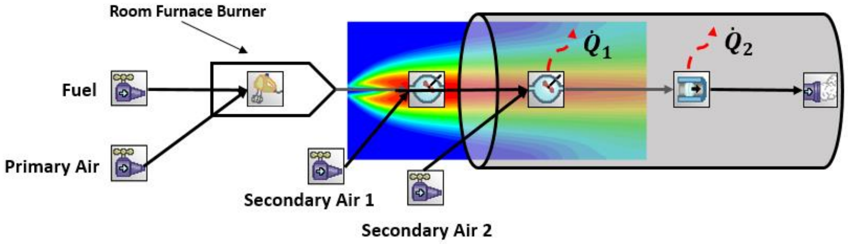
A chemical reaction network model is generated to simulate the room furnace burner flame based on the flame appearance, as shown in Figure 60. Fuel and primary air are first mixed together in a non-reactive gas mixer, which is used to simulate the furnace burner. In this case, the Venturi furnace burner works as the fuel/primary air container.

Afterwards, two perfectly stirred reactors (PSRs) are adopted to simulate the flame. The first stage of the secondary air is to simulate the air entrained at the burner exit, and the second stage of the secondary air is the air entrainment by the flame stream at the entrance of the flame tube.

The heat loss $\dot{Q}_1$ from the PSR is 0.1 kJ/s, which is to simulate the heat loss of the flame located at the flame tube entrance. The major heat loss $\dot{Q}_2$ is 0.07 kJ/(cm*s), which is to simulate the heat loss in the air heating process. The heat losses are estimated based on the exhaust temperature.



(a) CRN model of the room furnace burner



(b) CFD-CRN model of the room furnace burner

Figure 60: Numerical model of the room furnace burner

The CO/NO_X emission results comparison is shown in Figure 61. As shown, NO_X and CO emissions don't change much as more H_2 is injected into natural gas. Although the model under predicts NO_X emission using oxygen correction method, the experiment data fit the model prediction very well using calorific correction method.

The CO emission stays under 20 ppm at 3% oxygen correction and 10 ng/J of calorific correction. The NO_X emission stays around 100 ppm at 3% oxygen correction and 55 ng/J of calorific correction. The flashback occurs at 45% hydrogen addition into the fuel at the steady operating condition.

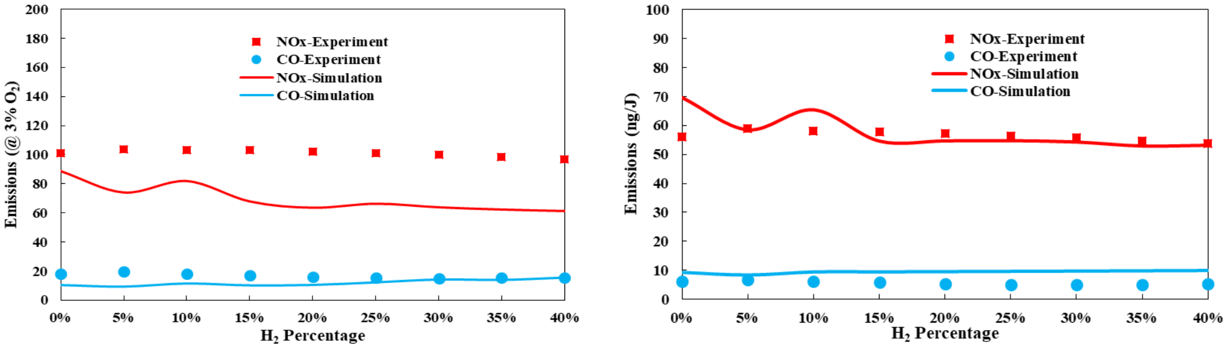


Figure 61: CO/NO$_X$ emissions comparison between simulation and experiments of the room furnace burner

5.3.2 *Tankless water heater*

Besides space heating, water heating is another important aspect of energy consumption in residential houses. Among different types of water heaters, tankless water heater is gaining significant popularity and government promotion due to its high energy efficiency and small space occupation.

5.3.2.1 Working principles

The inner structure of the investigated tankless water heater is shown in Figure 62, which has the highest rating of 199,000 Btu/hr (~ 58kW). As shown, the water heater has two heat transfer stages. After the hot exhaust goes through the primary heat exchanger, it will be directed to the secondary heat exchanger, in which the water content in the exhaust is going to be condensed. Therefore, this type of condensing water heater has a higher energy efficiency, which is usually higher than 90%.

The fuel control system of the tankless water heater keeps the heating load constant, which means it can adjust the fuel flowrate for different fuels based on the requirement of the heat output (hot water temperature setting & flowrate).

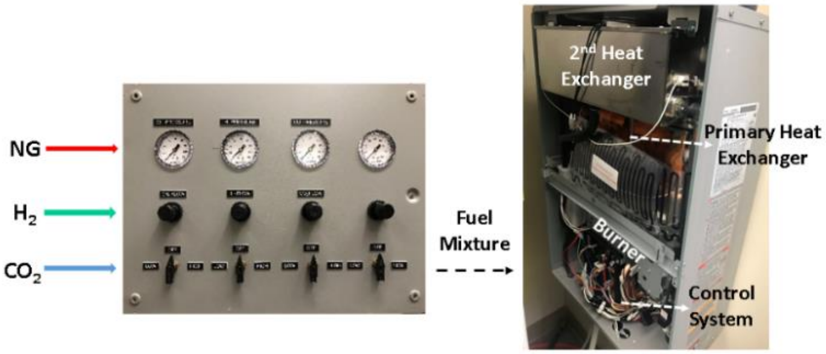


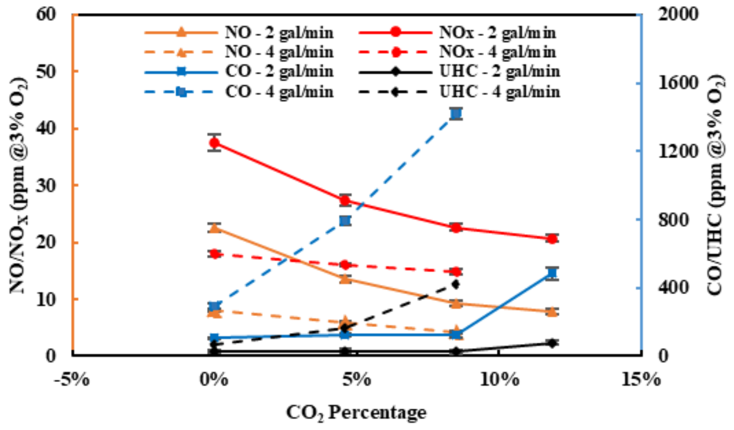
Figure 62: Tankless water heater inner structure

5.3.2.2 Emissions

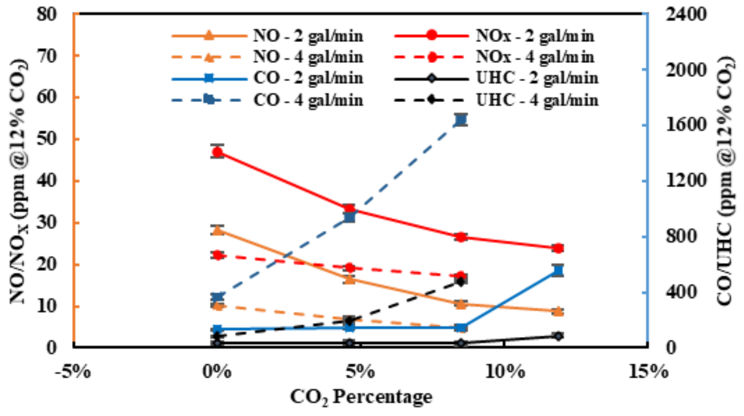
Figure 63 shows the emission levels of different water flowrates under CO_2/NG mixing situations, including NO, NO_X (NO+NO_2), CO and UHC. Generally speaking, CO_2 mixing into natural gas tends to decrease NO_X emission but increase CO/UHC emissions. This is caused by the inert property of CO_2, which can cause the flame temperature decrease. The lower flame temperature decreases the fuel oxidation rate, so the CO and UHC emissions increase. However, lower temperature of the reaction field slows down thermal NO_X formation, which results in NO_X concentration decrease in the exhaust.

As shown in Figure 63 (a), at 2 gal/min water flowrate condition, NO_X emission is around 37.5 ppm (@ 3% O_2) when the water heater operates on natural gas. When the CO_2 percentage in the fuel increases up to 12%, NO_X emission decreases down to 20.7 ppm (@ 3% O_2). To be noted, the NO emission is around half of the NO_X concentration. CO and UHC emissions do not change much when the CO_2 percentage is lower than 8%. CO emission stays at around 120 ppm (@ 3% O_2) and UHC is around 30 ppm (@ 3% O_2). After CO_2 percentage exceeds 8%, CO and UHC emissions increase significantly, and they are up to 484 ppm (@ 3% O_2) and 80 ppm (@ 3% O_2) separately when CO_2 percentage in the fuel reaches 12%. When the water flowrate increases up to 4 gal/min, at pure natural gas operating condition, NO_X emission reduces down to half of that at 2 gal/min. However, the decreasing rate is relatively low compared to 2 gal/min condition. The CO/UHC emissions of 4 gal/min are obviously higher than that of 2 gal/min, and the increasing trend of them with the CO_2 addition is also more obvious.

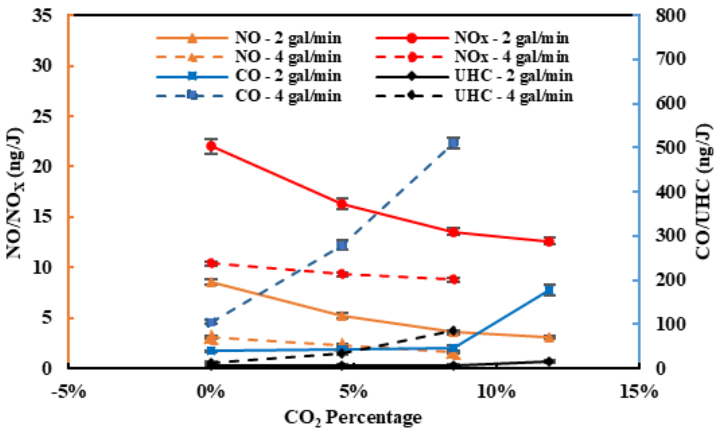
As shown in Figure 63 (b) and Figure 63 (c), CO_2 and calorific corrections show similar trend as O_2 correction. When the water flowrate is at 4 gal/min, NO_X emission of the tankless water heater is around 10 ng/J when operating on natural gas, which fits the strictest regulation of SDCAPCD Rule 69.5.1 [162].



(a) 3% O_2 correction



(b) 12% CO_2 correction

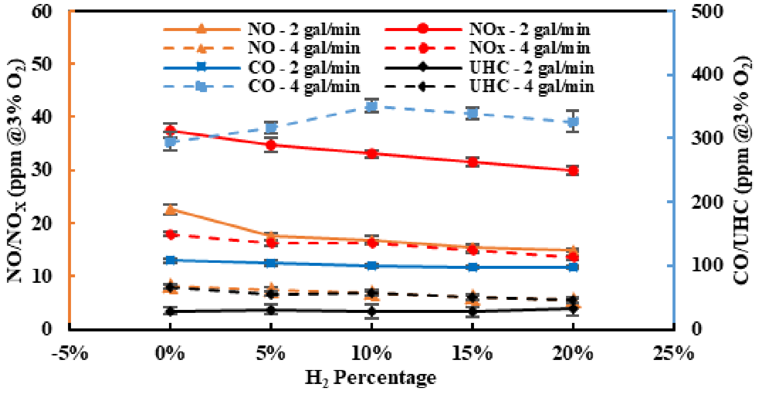


(c) Calorific correction

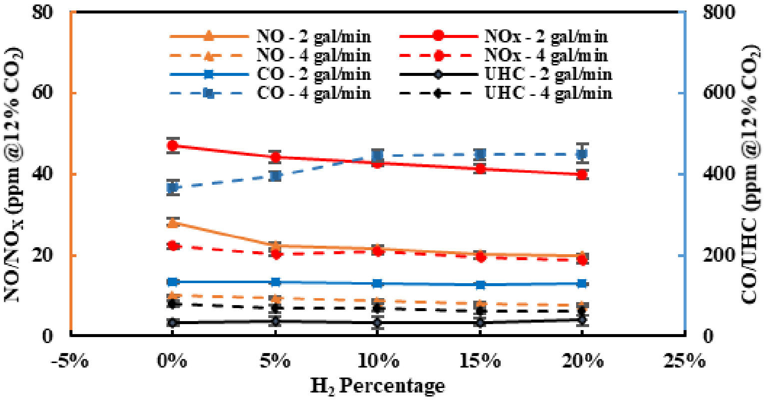
Figure 63: Tankless water heater emissions operating on NG/CO_2 mixtures

Figure 64 shows the emissions of the tankless water heater operating on H_2/NG mixtures. As can be seen in Figure 64 (a), compared to the emissions of operating on CO_2/NG mixtures, the emissions variance is relatively small. At 2 gal/min water flow condition, NO_X emissions deceases from 37.5 ppm (@ 3% O_2) to 29.9 ppm (@ 3% O_2) while the H_2 percentage in the fuel increasing from 0% to 20%. CO emission stays at around 100 ppm (@ 3% O_2) and UHC is around 30 ppm (@ 3% O_2). At 4 gal/min water flowrate, the NO_X emission decreases from 17.9 ppm (@ 3% O_2) to 13.5 ppm (@ 3% O_2), however, CO emission experiences some fluctuation.

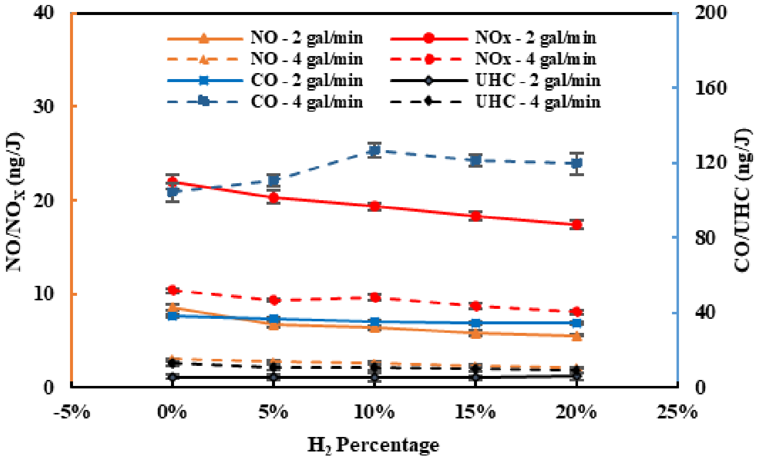
As shown in Figure 64 (c), using calorific correction does not change much of the trend of O_2 correction. However, CO_2 correction alleviates the decreasing trend of emissions as hydrogen percentage in the fuel increases. This is due to the property of the CO_2 correction methodology. The correction equation has the exhaust CO_2 percentage in the denominator. As more natural gas is replaced by H_2, the CO_2 percentage in the exhaust decreases, which will make the emission levels increase.



(a) 3% O_2 correction



(b) 12% CO_2 correction



(c) Calorific correction

Figure 64: Tankless water heater emissions operating on NG/H_2 mixtures

It should also be noted that NH_3 and N_2O emissions of the tankless water are lower than the detectable level of the emission analyzer. Therefore, it can be said that these two emissions can be negligible compared to the major emissions like NO_X/CO/UHC.

5.3.2.3 Efficiency

Efficiency is also one of the key factors to evaluate the performance of a water heater. The efficiency of a tankless water heater is defined as the heat absorbed by the water divided by the heat released from the fuel. Since the investigated water heater is a condensing water heater, a higher heating value is used to take the latent heat in the exhaust into account.

As mentioned in previous section, CO_2 injection into the fuel will decrease the Wobbe Index, therefore, the water heater takes more fuel to keep the water temperature constant until blow-off occurs. It can be concluded that CO_2 injection doesn't change the water heater efficiency that much before the combustion failure. When flame blow-off happens, the remote control reports error code of "Gas service has been interrupted".

H_2 addition increases the water heater efficiency. As shown in Figure 65, at 2 gal/min, the water heater efficiency increases from 92.4% to 97.2% when H_2 percentage increases from 0% to 20%. At 4 gal/min, the water heater efficiency increases from 89.5% to 94.1%. Another thing to be noted is the flowrate of condensing water from the exhaust, as it increases as more hydrogen is added.

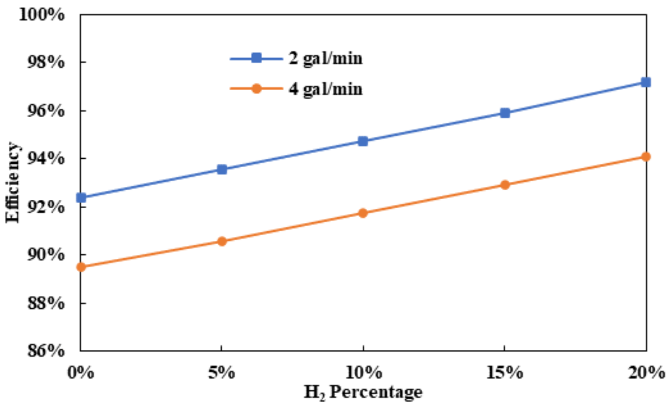


Figure 65: Tankless water heater efficiency operating on NG/H_2 mixtures

The increase of efficiency is mainly due to the decrease of heat input. However, the efficiency will not continue to go up to 100% as more hydrogen is added. As hydrogen exceeds a certain percentage, the tankless water heater will take more fuel from the pipeline and the efficiency will stop rising. However, before this situation happens, flashback might occur first due to the fast flame speed and wide flammability range of hydrogen, which might destroy the burner in the water heater.

5.3.3 *Other applications*

Heating is the largest energy consumption sector in residential buildings. Besides central room furnace, there are other options for space heating including fireplace, ventless space heater, etc.. However, due to their low efficiency and high emission levels, fireplace and space heater are getting depleted for indoor heating. However, fireplace might exist for a longer time for its outdoor heating effectiveness by radiation.

5.4 Summary

Existing residential appliances are mostly designed to operate on natural gas or propane. This chapter investigates on the combustion performance of representative residential self-aspirating burners operating on NG/CO_2 and NG/H_2 mixtures, which provides insights on conquering technical barriers for future residential fuel transition.

Here are the summary of the tested appliances performance operating on renewable gases:

- The cooktop burner can only operate on limited amount of CO_2 (~5 vol%), which tends to increase CO/UHC emissions before blow-off. The cooktop burner presented in this study has an ignition flashback limit of 20 vol% addition. Under steady-state operating condition, the hydrogen percentage in the fuel can be increased up to 75 vol%. However, when a cooking pot is placed over the burner, the flashback limit drops back to 55 vol% due to the interaction between the flame and the bottom of the cooking pot. Generally speaking, CO_2 addition tends to decease NO_X but to increase CO/UHC emissions. However, H_2 addition helps decrease the emissions slightly.
- The investigated oven burner has an ignition flashback limit of 30 vol% H_2 addition. However, the oven burner is over heated at around 20% H_2 added into natural gas. The H_2 addition doesn't change the NO_X emission levels significantly. At the same time, it decreases CO levels.
- The ignition flashback limit of the tested room furnace is 20% hydrogen addition in the pipeline natural gas. However, this limit increases up to 45% under steady operating conditions. The CO emissions stay at a stable level and NO_X emissions decrease slightly as H_2 is added into natural gas.
- The tested tankless water heater can operate on fuel mixtures up to 20 vol% H_2 without any influence on operability. H_2 addition helps decrease NO_X but doesn't change CO emissions that much. The addition of hydrogen helps slightly increase the water heater efficiency.

In conclusion, the residential self-aspirating flames don’t have a high tolerant level of CO_2. Therefore, before blending biogas into natural gas pipeline, the inert contents should be depleted to avoid unexpected CO formation in flames. However, low percentage (< 20 vol%) of H_2 addition can benefit CO or NO_X emissions reduction while maintaining other combustion performances, which shows a positive sign of incorporating renewable hydrogen in our existing natural gas infrastructures.

CHAPTER 6: Conclusions

6.1 Summary

Self-aspirating flames adopted in residential applications were studied to investigate the fuel interchangeability feasibility between natural gas and renewable gases. This study adopted both experimental methods and numerical simulation and went beyond the current regulations to explore the limits and technical barriers of adopting renewable fuels in existing fossil fuel facilities.

Existing gas quality regulations including heating value, Wobbe Index, etc. were investigated to examine the possible renewable gas blending percentage in natural gas. Current fuel flowrate control principles were discussed and the performance and technical challenges were predicted.

The influence of hydrogen addition on self-aspirating flame appearance was discussed and studied. Abnormal flame color of hydrogen-enriched fuels was investigated experimentally. Four groups of contrast tests were conducted to investigate the possible source of the flame color. Energy Dispersive X-Ray Spectroscopy (EDS) was adopted for analyzing the contaminant contents from the hydrogen storage facility.

Extensive combustion performance testing was applied to various residential self-aspirating flames. The investigated burners include cooktop burner, oven burner, central room furnace burner, water heater burner, etc.. Various fuel classes (NG/CO_2, NG/H_2) with varying renewable fuel percentages were supplied to the burners till the combustion failure occurred. The examined combustion performance include ignition behaviors, blow-off/flashback limits, emissions (CO, UHCs, NO, NO_2, N_2O, NH_3), efficiency, burner temperature, combustion noise, etc.. The primary air entrainment behavior of self-aspirating flames was also studied experimentally. CFD-CRN models were developed to capture the emissions variation tendencies of tested appliances, which is used to predict the performance of more self-aspirating flames operating on natural gas/renewable gas mixtures.

6.2 Conclusions

- **Gas combustion is so far the most efficient and reliable energy source in residential and commercial applications for providing heat.**

  Due to the low cost, high efficiency and reliability, gas combustion has a huge advantage in providing heat for residential and commercial sectors compared with electricity. Gas and electricity will co-exist a long time in the heating sector, especially when renewable gases can be adopted in existing natural gas storage and transmission facilities.

- **Heating value, Wobbe Index and other flame indices are limiting the adoption of renewable gases into existing natural gas infrastructures.**

  To incorporate lower hydrocarbons especially renewable hydrogen into existing natural gas infrastructures, the heating value limit should be lower than the existing regulation. For example, SoCalGas Company lowered the heating value limit from 990 Btu/scf to 970 Btu/scf, which allows the possible hydrogen injection percentage increase from 3 vol% to 6 vol%. These regulations should be coupled with each other to ensure safety and operability of gas consumption applications.

- **Fuel flow control systems have significant influence on renewable gases adoption in existing combustion applications.**

  Most combustion systems adopting self-aspirating flames lack delicate fuel flowrate control system. For systems without any flowrate control, Wobbe Index of the fuel is an indicator of the heat release rate. Fuel sensors coupled with the control systems are preferred for fuel flexible combustion applications. Secondary (condensing) heat transfer equipment can help increase the efficiency of the combustion application operating on hydrogen-enriched fuels.

- **Hydrogen cracking and embrittlement is not only a threat to storage and transport facilities but could also influence the flame characteristics.**

  Hydrogen carking and embrittlement not only degrade the material surface and cause reduction on storage and transmission facility lifespan, the particle residuals produced from these processes can also cause reddish color in flames. This abnormal flame color is similar to the flame yellow tips thus complicates the judgment on the combustion behavior. The misjudgment of the flame color can lead to faulty burner adjustment which might induce early flashback.

- **Existing residential self-aspirating flames have varying tolerant levels of renewable gases, and there is a positive sign of benefiting combustion performance of blending hydrogen into natural gas.**

  Different self-aspirating burner geometries have distinct tolerant levels on renewable gas contents. Carbon dioxide addition usually causes flame blow-off after 10 vol% addition, before which occurs, the carbon monoxide and unburned hydrocarbon emissions go up first. At relatively low percentage of hydrogen blending, self-aspirating flames performance generally improves regarding ignition time, emissions, efficiency, etc.. As hydrogen exceeds a certain limit, the flashback risk increases. Based on the test results, the existing residential self-aspirating flames can safely operate on natural gas/hydrogen mixtures, in which hydrogen percentage should be lower than 20%.

- **Most self-aspirating burners can be modified on site to incorporate more renewable gases. However, to further improve the combustion performance of burners operating on renewable gases, more research and development work needs to be done.**

  In previous energy transition from manufactured gases to natural gas, most of the self-aspirating burners in the U.K. were modified on site to accommodate the gas transition, which provides experience for the upcoming natural gas transition to renewable gases. A large number of today's gas appliances are designed interchangeable between natural gas and propane. These burners and appliance parts are already designed interchangeable, which gives more convenience to the on-site burner modification work.

  The heat output of self-aspirating flames operating on Wobbe Index principle is less easily affected by the replacement of natural gas with hydrogen due to WI similarity of these two fuels. Without modifying the fuel control system, the burners can be replaced in situ with modified hydrogen tolerant burners. The flame ports can be sized down to accommodate hydrogen-enriched fuel which needs smaller quenching distance to arrest flames before flashback occurs. The primary air aeration configuration could also be optimized to decrease the amount of air entrainment into the burner, which helps prevent flashback into the burner.

- **Most existing standards and regulations on residential self-aspirating burners are obsolete and incomprehensive.**

  Most self-aspirating burners operate at open-air conditions, which bring challenges to experimental measurements and combustion performance quantification. Therefore, universal testing standards and regulations on these burners should be developed. For example, the CO emission levels of tested burners in this dissertation are far below the ANSI regulations.

Moreover, the corrected emissions under a certain reference should be adopted in regulations instead of the raw emission levels.

6.3 Recommendations

In the past energy transitions, fuel interchangeability is essential to ensure a smooth transition from previous energy sources to the new fuel type. The fuel interchangeability studies for this upcoming or already happening fuel transition from fossil fuel to renewable energy forms need more comprehensive and thorough than ever. Since there are not any abundant ready-to-use fuel sources like oil or natural gas on earth anymore, the renewable fuels must be generated from different technologies.

The fuel interchangeability from natural gas to renewable gases in self-aspirating flames needs much more effort than previous energy transitions. The last systematic fuel transition was from manufactured gas to natural gas starting in the early 20th century. At that time, safety operation was nearly the only factor to be considered. Nowadays, much more aspects need to be paid attention to in terms of burner optimization to adopt renewable fuels. These aspects include efficiency, ease of operation and maintenance, emissions species, the decency of outlook, etc.. Although this dissertation has conducted relatively extensive analysis and testing, there are still a lot more challenges need to be dealt with:

- The influence of hydrogen-enriched fuel on the self-aspirating burner lifespan is still not clear. Whether hydrogen has a slow embrittlement effect on the burner material at combustion conditions needs further investigation.
- There is a need for the reaction mechanism accurately predicting emissions of partially premixed flames operating on hydrogen-enriched natural gas.

- Extensive self-aspirating burner optimization studies need to be conducted to increase their hydrogen tolerant levels.

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APPENDIX A

There is a mistake regarding hydrogen flammability limits in the Table 8.4 (Page 291) of the book < An Introduction to Combustion: Concepts and Applications, Third Edition>.

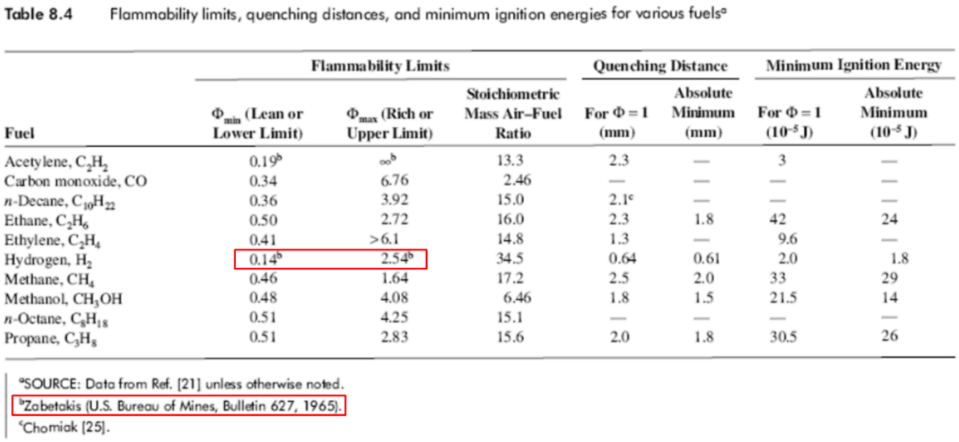


Table 8.4 Flammability limits, quenching distances, and minimum ignition energies for various fuels[a]

	Flammability Limits			Quenching Distance		Minimum Ignition Energy	
Fuel	Φ_{min} (Lean or Lower Limit)	Φ_{max} (Rich or Upper Limit)	Stoichiometric Mass Air–Fuel Ratio	For $\Phi = 1$ (mm)	Absolute Minimum (mm)	For $\Phi = 1$ (10^{-5} J)	Absolute Minimum (10^{-5} J)
Acetylene, C_2H_2	0.19[b]	∞[b]	13.3	2.3	—	3	—
Carbon monoxide, CO	0.34	6.76	2.46	—	—	—	—
n-Decane, $C_{10}H_{22}$	0.36	3.92	15.0	2.1[c]	—	—	—
Ethane, C_2H_6	0.50	2.72	16.0	2.3	1.8	42	24
Ethylene, C_2H_4	0.41	>6.1	14.8	1.3	—	9.6	—
Hydrogen, H_2	0.14[b]	2.54[b]	34.5	0.64	0.61	2.0	1.8
Methane, CH_4	0.46	1.64	17.2	2.5	2.0	33	29
Methanol, CH_3OH	0.48	4.08	6.46	1.8	1.5	21.5	14
n-Octane, C_8H_{18}	0.51	4.25	15.1	—	—	—	—
Propane, C_3H_8	0.51	2.83	15.6	2.0	1.8	30.5	26

[a] SOURCE: Data from Ref. [21] unless otherwise noted.

[b] Zabetakis (U.S. Bureau of Mines, Bulletin 627, 1965).

[c] Chomiak [25].

The table indicates that hydrogen has an equivalence ratio range of φ=0.14~2.54. Therefore, this equivalence ratio flammability range is used to calculate the volume percentage flammability range of hydrogen in air.

Keeping the coefficient before the term (O_2+3.76N_2) as 0.5, the coefficient before H_2 can represent the equivalence ratio.

$$\varphi H_2 + 0.5(O_2 + 3.76N_2) \rightarrow Products$$

Then the volume percentage flammability range can be calculated by the following equations:

$$V_{min,\%} = \frac{0.14}{0.14+0.5*4.76} \approx 5.6\%,\ V_{max,\%} = \frac{2.54}{2.54+0.5*4.76} \approx 51.6\%$$

Therefore, the volume percentage flammability range hydrogen is 5.6%-51.6% based on the equivalence ratio range provided in the book. However, a lot of papers are showing the volumetric flammability range of hydrogen is 4%-75%, which is different from Prof. Turns's book.

Therefore, the original report cited in the table: Zabetakis (U.S. Bureau of Mines, Bulletin 627, 1965) is found for analysis. The original data source is from Table 18 on Page 77 of the report.

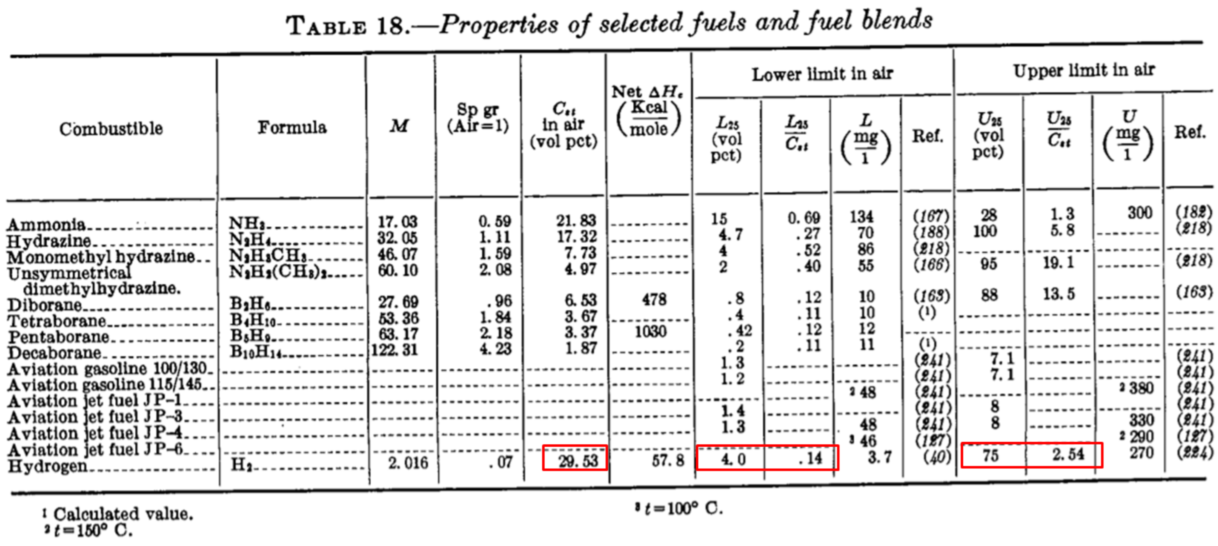


TABLE 18.—*Properties of selected fuels and fuel blends*

Combustible	Formula	M	Sp gr (Air=1)	C_{st} in air (vol pct)	Net ΔH_c $\left(\frac{\text{Kcal}}{\text{mole}}\right)$	Lower limit in air				Upper limit in air			
						L_{25} (vol pct)	$\frac{L_{25}}{C_{st}}$	L $\left(\frac{\text{mg}}{\text{l}}\right)$	Ref.	U_{25} (vol pct)	$\frac{U_{25}}{C_{st}}$	U $\left(\frac{\text{mg}}{\text{l}}\right)$	Ref.
Ammonia	NH_3	17.03	0.59	21.83	---	15	0.69	134	(*167*)	28	1.3	300	(*182*)
Hydrazine	N_2H_4	32.05	1.11	17.32	---	4.7	.27	70	(*188*)	100	5.8	---	(*218*)
Monomethyl hydrazine	$N_2H_3CH_3$	46.07	1.59	7.73	---	4	.52	86	(*218*)	---	---	---	---
Unsymmetrical dimethylhydrazine.	$N_2H_2(CH_3)_2$	60.10	2.08	4.97	---	2	.40	55	(*166*)	95	19.1	---	(*218*)
Diborane	B_2H_6	27.69	.96	6.53	478	.8	.12	10	(*163*)	88	13.5	---	(*163*)
Tetraborane	B_4H_{10}	53.36	1.84	3.67	---	.4	.11	10	[1]	---	---	---	---
Pentaborane	B_5H_9	63.17	2.18	3.37	1030	.42	.12	12	---	---	---	---	---
Decaborane	$B_{10}H_{14}$	122.31	4.23	1.87	---	.2	.11	11	[1]	---	---	---	---
Aviation gasoline 100/130	---	---	---	---	---	1.3	---	---	(*241*)	7.1	---	---	(*241*)
Aviation gasoline 115/145	---	---	---	---	---	1.2	---	---	(*241*)	7.1	---	---	(*241*)
Aviation jet fuel JP-1	---	---	---	---	---	---	---	[2] 48	(*241*)	---	---	[2] 380	(*241*)
Aviation jet fuel JP-3	---	---	---	---	---	1.4	---	---	(*241*)	8	---	---	(*241*)
Aviation jet fuel JP-4	---	---	---	---	---	1.3	---	48	(*241*)	8	---	330	(*241*)
Aviation jet fuel JP-6	---	---	---	---	---	---	---	[3] 46	(*127*)			[2] 290	(*127*)
Hydrogen	H_2	2.016	.07	29.53	57.8	4.0	.14	3.7	(*40*)	75	2.54	270	(*224*)

[1] Calculated value.
[2] $t=150°$ C.
[3] $t=100°$ C.

In the table, the volumetric flammability range of hydrogen is also 4% to 75%. However, the range of 0.14 to 2.54 is not the equivalence ratio, instead, it is the range of (hydrogen volume percentage at limit conditions/hydrogen volume percentage at stoichiometric condition).

Here are the calculations based on the table in U.S. Bureau of Mines, Bulletin 627, 1965:

$$\varphi H_2 + 0.5(O_2 + 3.76N_2) \rightarrow Products$$

At stoichiometric condition

$$H_2 + 0.5(O_2 + 3.76N_2) \rightarrow H_2O + 1.88N_2$$

At lower limit condition, $\varphi_L = 0.10$ can be derived from the following equation

$$\frac{L}{C_{st}} = \frac{\frac{\varphi_L}{\varphi_L + 0.5 * 4.76}}{29.53\%} = 0.14$$

At higher limit condition, $\varphi_U = 7.14$ can be derived from the following equation

$$\frac{U}{C_{st}} = \frac{\frac{\varphi_U}{\varphi_U + 0.5 * 4.76}}{29.53\%} = 2.54$$

Therefore, the flammability of hydrogen in air show be $0.10 < \varphi < 7.14$. To double check with the volumetric range from 4% to 75%:

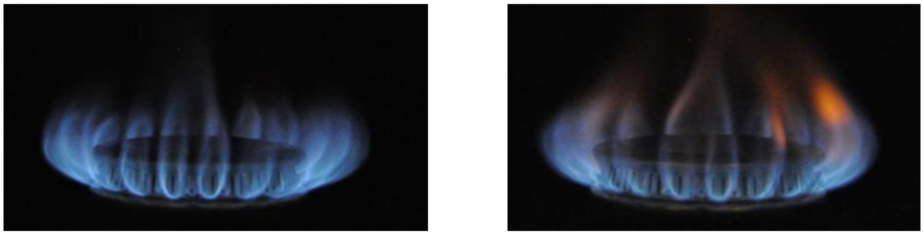
$$V_{min,\%} = \frac{0.1}{0.1+0.5*4.76} \approx 4\% \,, \quad V_{max,\%} = \frac{7.2}{7.2+0.5*4.76} = 75\%$$

To conclusion, in the book < An Introduction to Combustion: Concepts and Applications, Third Edition >, the hydrogen flammability range of φ=0.14~2.54 should be corrected to $0.1 < \varphi < 7.14$.

APPENDIX B

Flame appearance is an important factor that needs to be considered in fuel interchangeability studies, especially for open-air self-aspirating flames in residential appliances. For appliance end users without advancement measurement tools (thermocouples, emission analyzers, etc.), flame appearance might be the only index for people to evaluate whether the burner is operating appropriately or not. Therefore, the flame appearance study of natural gas mixed with renewable gases is a crucial aspect in fuel interchangeability study.

This chapter aims to explain the reason of the unusual reddish flame color of hydrogen enriched natural gas observed in the experimental study of a cooktop burner operated on different mixtures, as shown in Figure 66.



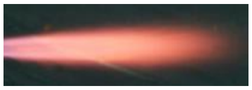
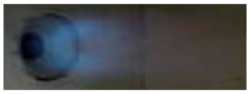
(a) 100% natural gas *(b) 55% natural gas + 45% hydrogen*

Figure 66: Cooktop burner flames operating on different fuels

Beyond the observation in this study, the reddish color of hydrogen-rich fuel has been observed in several labs around the world (USA, UK, Netherland, China, Korea, Japan, etc.) that have been actively studying high hydrogen content combustion applications, as shown in Table 20.

Fundamental studies show that pure hydrogen flames should be invisible [195][196][197][198] or pale blue due to reactions involving hydroxyl radicals [199][200]. Yet reddish flame colors are occasionally observed in hydrogen flames, as shown in Table 20. Before 2009, some researchers mentioned this phenomenon but didn’t provide details [201]. The reddish color was usually attributed to impurities in the fuel or air [202][203][204]. In 2009, Schefer [199] noted that the red color might be caused by excited water molecules. After this paper appeared in the literature, many hydrogen combustion studies cited Schefer’s work in explaining the reddish glow in hydrogen flame images.

Table 20: Reddish color observation of hydrogen flames

Time	Authors/Institution	Description	Operating Condition	Explanation
1997, 2000	Choudhuri, et al., University of Oklahoma, USA [202][203]	…100% Hydrogen… a reddish glow in the far-burner region….	Single jet burner; Diffusion flame; 100% H_2	Although pure Hydrogen flame should be invisible, the luminosity and colors observed are caused by the impurities in the fuel and the surrounding air.
2008	Wu, et al., Chinese Academy of Sciences, China [204]		Industrial jet burner; Diffusion flame; 100% H_2	Although pure hydrogen should be invisible, the luminosity and colors observed here may be caused by the impurity of fuel or air.
2009	Cho, Delft University of Technology, Netherlands; Chung, Seoul National University, Korea [201]		Gas turbine swirl burner; Premixed flame ($\phi=0.87$); 29% H_2+71% NG	The flame color became somewhat reddish with hydrogen addition.
2009	Schefer, et al., Sandia National Laboratories, USA [199]		Laminar jet burner; Diffusion flame; 100% H_2	The emission bands due to water…While visible emissions from hydrogen flames are considerably weaker than those from comparable hydrocarbon flames, they are visible at reduced light levels in most cases.
2013	Chaudhari, Jadavpur University, India; Sahu, University of Illinois at Chicago, USA; Virginia Polytechnic Institute and State University, USA [205]		Industrial swirl burner; Premixed flame (ϕ~1); 100% H_2	The reddish appearance of the flame is due to emission from non-hydrocarbon molecules… a possible source of the reddish color of the flame and spectral peak is the emission from the vibrationally excited H_2O molecules.
2018	Kalantari, et al., University of California, Irvine, USA [206]		Gas turbine jet burner; Premixed flame ($\phi=0.4$); 100% H_2	No explanation.
2018	Liu, et al., Chinese Academy of Sciences, China [207]		Industrial jet burner; Premixed flame ($\phi=0.73$); 50% H_2+50% NG	No explanation.
2018	Jones, et al., Swansea University, UK [208]		Cooktop burner; Partially premixed flame ($0.8<\phi<2.1$); 30% H_2+70% NG	No explanation.

Fundamental spectroscopy analysis [209] indicate that excited water molecules emit strongly at 717 nm and 810 nm, and that the peak band of excited molecular oxygen occurs at 768nm. These wavelength bands fall into the infrared region, meaning the radiation is nearly invisible to the human eye. Hence, this red-light emission should be quite weak or only visible under greatly reduced light conditions [199]. However, some studies show that this reddish radiation is very obvious even under normal light condition. Jones [105][208] from Swansea University of UK presented reddish flame tails from a burner operating on hydrogen/natural gas mixtures. Liu [207] from Chinese Academy of Sciences also found obvious reddish flame color as more hydrogen is added to natural gas. Kalantari [206] studied flashback behavior of hydrogen flames in a gas turbine burner, and observed obvious reddish flame color. In this last study, after conducting experiments, a layer of reddish powder was coated on the quartz tube confining the burner.

The underlying reason for the reddish color observed in hydrogen flames is still controversial. In the present work, analysis of potential reasons causing this abnormal hydrogen flame color are discussed. Combustion experiments are conducted comparing different situations, and electron microscopy analysis is adopted to investigate the contents of the reddish residual after hydrogen combustion.

Assumption 1: impurities in the pipeline or mixing station

One possibility is that the reddish color shown in Figure 66 is caused by impurities associated with the fuel mixing station or from the natural gas pipeline. The materials used in this mixing station are stainless steel (tube), Teflon (tube), and brass (valve). Stainless steel and Teflon won’t induce any flame color, but a large excess of cupric chloride could cause a diffuse red color in a hydrogen flame. However, this situation requires significant levels of cupric chloride or hydrogen with copper particles burning in chloride. In the study, no chloride content was detected in hydrogen to react with the brass material of the valve. Moreover, pure natural gas also flowed through the mixing station before supplied to the burners, and there was no reddish glow observed in natural gas flame. Therefore, the abnormal flame color is not caused by the impurities in the natural gas pipeline or mixing station. The possibility that the entrained dust from the surrounding was radiating the reddish emission is also dismissed, because all the flames studied burn in the same environment and the pure natural gas flame emits only the normal blue radiation.

Assumption 2: soot formation

Soot formation and radiation is a common source of reddish light emission in hydrocarbon flames. Therefore, emission measurements were obtained to assess any incomplete combustion that might cause soot formation. NO_X and CO emissions were measured by Horiba Portable Gas Analyzer 350 (PG 350), which has an accuracy of ±1 ppm. The UHC emission was measured by Horiba Flame Ionization Magneto-Pneumatic Analyzer 220 (FMA 220), which has an accuracy of ±1% of the measurement range. The CO/UHCs emissions decrease as more hydrogen is added into natural gas [107]. The emission sampling methodology can be found in Reference [106][107].

To further verify that the flame color is not from the radiation of soot, a pure hydrogen flame is used as a comparison. As shown in Figure 67, hydrogen flows through a stainless tube and is ignited by the cooktop burner, which is operating on pure natural gas. As shown, the pure hydrogen flame has an obvious reddish color, which also verifies that the color is not from soot formation, because hydrogen contains no carbon.

Figure 67: Natural gas and pure hydrogen flame color comparison

Therefore, it can be concluded that the reddish flame color is not associated with soot or hydrocarbons. The reddish/orangish color is associated with the hydrogen.

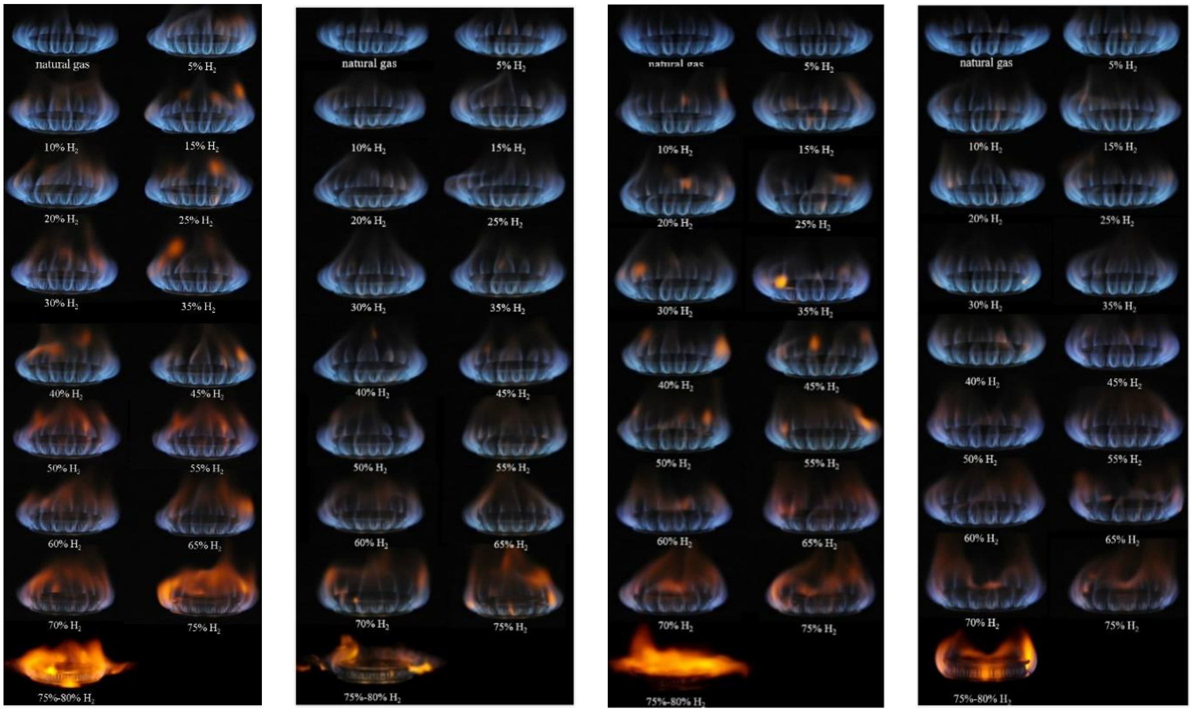
Assumption 3: hydrogen grades

The impurities in hydrogen can be gas contaminants, liquid droplets, or fine solid particles. Industrial grade (IG) hydrogen was used in tests mentioned above and it is a cost-effective test fuel used by many of the studies summarized in Table 20. In the present study, to assess the influence of gas purity on flame characteristics, ultra-high purity (UHP) hydrogen is also tested. Table 21 shows the composition specifications of the two hydrogen grades. Although the industrial grade hydrogen has more trace gas contaminants, those species are not associated with any reddish light emission.

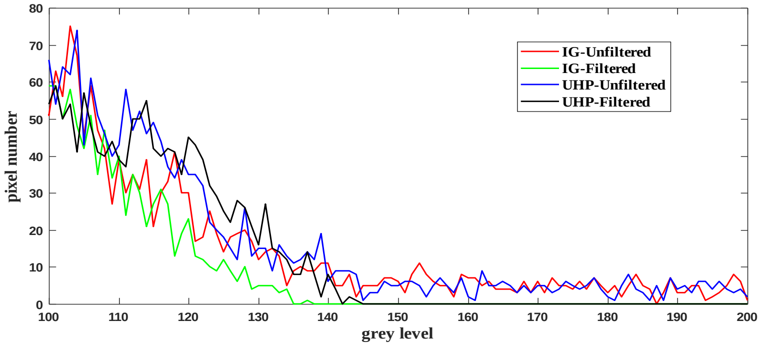
Table 21: Comparison of two different hydrogen grades

	Industrial Hydrogen	Ultra-high Purity Hydrogen
H_2	99.95%	99.999%
O_2	10 ppm	1 ppm
H_2O	Dew Point: -60 °C	2 ppm
HC	10 ppm	0.5 ppm
CO	10 ppm	1 ppm
CO_2	10 ppm	1 ppm
N_2	34 ppm	5 ppm
Relative Cost	1	>2
Cylinder Material	steel	steel

To investigate whether hydrogen has fine particles influencing the flame color, studies were conducted with and without a 0.5-micrometer filter installed on the fuel line. In effect, the two hydrogen grades and the application of a filter forms a 2 by 2 experiment. Figure 68 shows the cooktop burner flame colors with hydrogen percentage increasing from 0% to around 80%. Figure 68 (a) and (b) show the flame appearance of industrial grade (IG) hydrogen/natural gas mixtures with and without particle filtration. Figure 68 (c) and (d) show flame images for ultra-high purity (UHP) hydrogen/natural gas mixtures. As shown in Figure 68 (a) and (c), the IG and UHP hydrogen flame colors are very similar, and both of them have reddish emissions. However, with the 0.5 μm particle filter installed, the reddish light emission is reduced significantly. It should be noted that, when the hydrogen percentage in the fuel increases from 75% to 80%, flashback occurs due to the high flame speed of the fuel mixture. Under this circumstance, fuel/air mixture in the burner head is ignited before it flows out of the burner and mixes with the surrounding air. This fuel-rich combustion within the burner head causes soot formation, which results in a bright orange-yellow flame at flashback conditions.



(a) IG *(b) Filtered IG* *(c) UHP* *(d) Filtered UHP*



(e) Red color grey channel comparison of 45% hydrogen addition

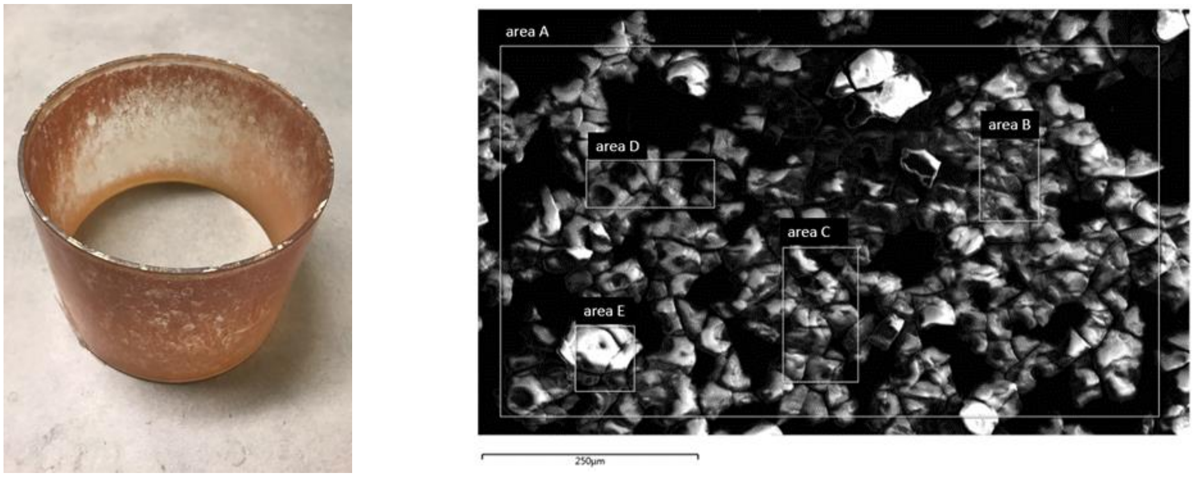
Figure 68: Flame color of different fuel grades

The grey maps of red color of the two images are compared. The flame images are 8-bit digital images, which means each pixel is represented by 28 (256) colors. Each pixel has three color channels (Red, Green, and Blue). The red channel in grey scale is analyzed to compare the flame red color of 45% hydrogen addition under four different situations, which is shown in Figure 68 (e). For the X axis, lower grey level represents “darker” color and higher grey level represents “brighter” color. Y axis is the number of pixels of each grey level. From the full grey map, we can see that the dark region, which is the black background, has the most pixels. For our analysis, we should mainly focus on the flame, which is the brighter region. Therefore, grey level higher than 100 is plotted in (e). As can be seen, there is not that much difference of the red color map at lower grey level, which means dark region, however, as the grey level going higher to the bright region, the unfiltered flame has more pixels containing high percentage of the red color. Moreover, there is not much difference between UHP hydrogen and IG hydrogen red color may in the bright region. Obviously, the application of filter can reduce the reddish color instead of using higher purity hydrogen. Therefore, it can be concluded that reddish/orangish color is not because of the gaseous impurities in the fuel, but from the particles in the hydrogen. As shown, none of the combinations of filter and gas quality impact the flashback propensity—the basic combustion performance behavior remains similar.

To assess the red color quantitatively, the red channel is isolated to compare four cases shown in Figure 68. A Design Expert-ANOVA tool is adopted to conduct t-test. The results indicate the red channel values decrease after filtration with more than 99.99% confidence. Further, the analysis indicates the difference is associated only with filtration levels, not with the purity of the hydrogen (IG vs UHP). Therefore, it can be concluded that reddish color in the flame has nothing to do with the hydrogen fuel grade but is associated with filterable particles in the hydrogen.

Assumption 4: hydrogen embrittlement

Based on the experiment results, it is concluded that the reddish light emission is caused by fine particles in the hydrogen source (in this case steel cylinders). To evaluate these particles, it was first attempted to collect them by flowing the hydrogen through filter paper. However, due to the low flow rate of the hydrogen supplied to the cooktop burner, the filter didn't collect enough particles to conduct electron microscopy analysis. Therefore, a quartz tube from Kalantari's work [206] shown in Figure 69 (a) is used to extract samples, which are analyzed by electron microscopy analysis using MAGELLAN 400L.



(a) Contaminated quartz tube *(b) Electron microscope image of the contaminant*

Figure 69: Quartz tube after hydrogen combustion and electron microscope image of the sample

The microscope image is shown in Figure 69 (b). Five characteristic areas are selected to analyze the elements in each area, and the percentage of each element is shown in Table 22. The elements of highest percentage in the sample are O and Si, the rest are Fe, Na and Ca. The reason for O and Si in the sample is due to the contaminant sample being taken off the inner surface of the quartz tube by a carbon tape. As a result, the sample contains compounds from the quartz tube itself.

Table 22: Elements of the quartz tube contaminant

	Area A	Area B	Area C	Area D	Area E
O	54.17%	53.02%	53.18%	53.73%	54.83%
Si	44.71%	46.07%	45.38%	44.82%	44.7%
Fe	0.86%	0.69%	1%	1.17%	0.47%
Na	0.26%	0.22%	0.27%	0.28%	/
Ca	/	/	0.17%	/	/

Many studies involving hydrogen use gases provided in similar relatively low-cost steel cylinders, although the gas suppliers might be different. It is believed that the reddish color is caused by the metal particles in the cylinder produced from reactions between hydrogen and the cylinder material or the storage and transport facilities before hydrogen was injected into steel cylinders. Hydrogen cracking and embrittlement is one of the major concerns hindering utilizing hydrogen as a pipeline fuel. Due to the small size of hydrogen molecule compared to other gas molecules, hydrogen is more likely to diffuse into the crystal lattice of the metal, creating pressure within the metal, then create cracks on the material surface. This phenomenon is called hydrogen attack or hydrogen-assisted cracking.

Not only high pressure can increase the rate of hydrogen embrittlement, high temperature can also amplify the embrittlement effect, especially for carbon steel materials [210][211][212]. Figure 70 shows the operating limits of different materials exposed to hydrogen environments. The solid lines note the hydrogen cracking (or fissuring) limits for various steels. As can be seen, carbon steel (most common steel for gas cylinders) is one of the most vulnerable materials for hydrogen attack. For a lab-use hydrogen gas cylinder, the pressure is usually above 2000 psi which means hydrogen cracking is taking place under room temperature. Sandia National Laboratories [213] also found there is no hydrogen percentage threshold in a mixture below which hydrogen effect can be ignored. That means even small amounts of hydrogen have large effects on materials.

This hydrogen cracking phenomenon on different steel materials were investigated by Dadfarnia et al. [214]. This study shows that under cyclic loading, the cracking effect of hydrogen is more significant which might shorten the life of the material. The pressure in hydrogen cylinders are generally cycled between atmosphere pressure and 10 MPa due to the end user consumption and gas supplier refill. This pressure cycle behavior exacerbates the hydrogen cracking behavior and may result in metal particle deposition in hydrogen cylinders. Moreover, chemical reactions between hydrogen and the metal itself or with some impurity or alloy addition in the metal can also cause damage to the material. This phenomenon is called embrittlement, which is associated with the chemical property of hydrogen [215].

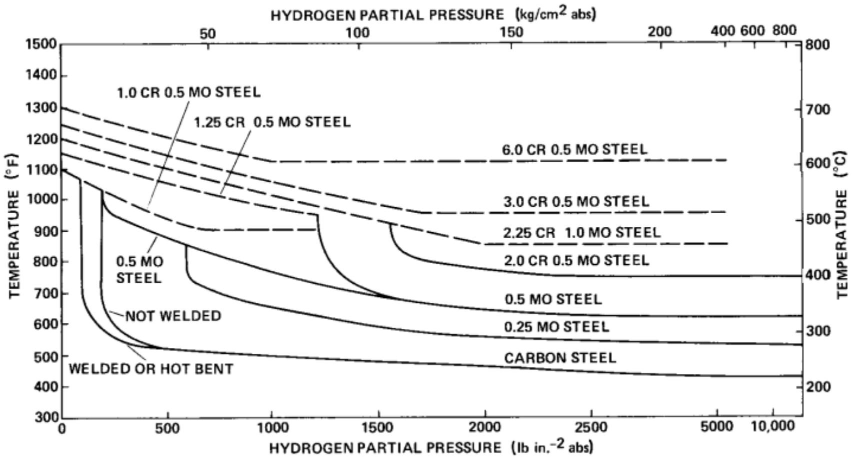


Figure 70: Operating limits developed from industrial experience for various steels exposed to hydrogen-containing experiments [211]

The dash lines in Figure 70 are embrittlement (decarburization) limits, which involve chemical reactions between hydrogen and the material. There are many possible reactions in the embrittlement process. The reaction between hydrogen and carbon in the metal is one of the most common reactions [216]:

$$Fe_3C + 2H_2 \rightarrow 3Fe + CH_4$$

The reddish-brown particles on the quartz tube is believed to be Fe_2O_3. It is also believed that the iron ions in the flame are originally formed from the iron metal particles produced by hydrogen embrittlement process, which are then further oxidized into Fe_2O_3 by both the slow oxidation in the storage facility (Table 21 shows a small amount of oxygen existing in hydrogen cylinder) and in the combustion process. In a flame, Fe^{2+} can cause gold-brownish yellow color, Fe^{3+} can cause orange color. Sodium results in bright yellow color, and calcium appears orange to orange-red color. Based on the electron microscopy results, the cause of the reddish color contaminant is believed to be the combination of $Fe^{2+}/Fe^{3+}/Na^{+}/Ca^{2+}$, mainly iron ions. In fact, the method of adding iron or steel filings in flames to achieve reddish color is used extensively in fireworks chemistry [217]. These metal particles emit reddish light whose wavelength usually falls between 620 and 700 nm [218].