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Rice-based ash in concrete: A review of past work and potential environmental sustainability

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Abstract:

The demand for concrete continues to grow with increases in population and increased urbanization. This demand, in turn, increases the need to reduce the environmental impacts of concrete while continuing to provide the same or better performance. Increasing population also creates increasing demand for food and energy resources. The cultivation of rice, the staple food of over half the world's inhabitants, results in the production of additional biomass. Rice biomass, such as hulls and straw, can be combusted as a renewable energy source and, under specific combustion conditions, the resulting ash can be used as a supplementary cementitious material or beneficial filler in the production of concrete, which can potentially lead to reduced environmental impacts in concrete.

This paper reviews rice-based ash and its influence on concrete properties to address current understanding of these ashes as an alternative mineral admixture. The review of the literature shows that under proper combustion conditions as well as through use of pre- and post-combustion treatments, highly pozzolanic (reactive) ash can be produced from rice hulls. These reactive ashes have the ability to improve several properties of concrete when used as a partial replacement for cement. Additionally, the production of rice-based ash can offer a lower greenhouse gas emitting pozzolan than portland cement and some conventional supplementary cementitious materials. Further research into the utilization of rice-straw ash, durability properties, and high performance concrete could lead to the production of rice ash as a cost- and environmentally-competitive alternative in the production of concrete.

Keywords:
Rice hull ash; Rice straw ash; Biomass ash; Pozzolans; Concrete; Supplementary cementitious materials

Declaration of interest: none.

Outline:

1. Introduction

The use of agricultural products as a source of energy and the subsequent use of the by-products as a constituent in concrete may provide a means to contribute to the meeting of sustainable development goals by reducing the environmental impacts of energy production and manufacture of concrete materials. Global energy demand has doubled in the past four decades and the use of bio-derived fuels has shown an 85% increase (IEA 2015b). This growth in energy demand is in large part a reflection of improved standards of living across the globe multiplied by population growth, which has also driven a large increase in the production of infrastructure materials (Monteiro *et al.,* 2017). The rate of increase of concrete production is outpacing most other infrastructure materials with notable resulting environmental impacts: concrete is responsible for 8-9% of anthropogenic greenhouse gas (GHG) emissions, 2-3% of the world's annual energy demand, and 9-10% of industrial water withdrawal (Monteiro *et al.,* 2017; Miller *et al.,* 2018a).

In the production of portland cement, both the fossil-fuel based combustion energy and the calcination process emit significant amounts of carbon dioxide into the atmosphere. Cement is only one of several constituents required in the production of concrete and mortar, and typically less than 10% of the mass of concrete (Mehta and Monteiro 2006), but it is responsible for the majority of GHG emissions from concrete production (Miller *et al.,* 2016). Use of mineral admixtures, such as pozzolans like fly ash or bio-derived ash, added as supplementary cementitious materials (SCMs) in concrete or pre-blended into the cement can reduce the demand for portland cement production. Pozzolans can also improve important material properties in concrete (Thomas 2007). Currently, SCMs are not being produced in large enough quantities to meet future impact reduction goals (WBCSD and IEA 2009; Miller *et al.,* 2018b).

Of the bio-derived ashes that have been studied for their potential to act as partial replacement of cement, rice-based ashes are among the most promising. Rice is the staple food of over half of the world's inhabitants (National Geographic Education Staff 2014). The USDA reports that 10.2 million metric tons were produced in the US in 2016 (USDA 2018). Similar to the combustion of fossil-derived energy sources, byproducts from combustion in the form of ash are formed during the combustion of bio-derived materials. Rice production in the US equates to a potential combustion product of approximately 2 million metric tons of combined rice hull ash (RHA) and rice straw ash (RSA) annually (Jenkins *et al.,* 1998; Mehta and Monteiro 2006; Binod *et al.,* 2010). Use of rice-based ash to fill the need for SCMs could offer an advantageous use of energy co-products (Aprianti *et al.,* 2015), thus increasing its economic and environmental value.

One of the most popular mineral admixtures used with cement in the production of concrete is coal fly ash. This fly ash is a by-product of coal combustion and possesses pozzolanic properties due to its high amorphous silica content, such that it acts as a binder when mixed with portland cement. While the use of an industrial by-product that reduces the demand for cement has appeal and a strong market, only 25-30% of fly ash produced is usable as a mineral admixture in concrete (FHWA 2012). The amount of useable fly ash is not keeping up with escalating demand and the availability of fly ash is questioned for the future (Stein *et al.,* 2015). Further, the availability of other mineral admixtures commonly used in concrete, such as ground granulated blast furnace slag, a byproduct from the iron production industry, has also been brought into question, which is pushing the cement and concrete industries to explore other material alternatives (Miller *et al.*, 2018b). These availability concerns have sparked interest in other ashes for use in cement-based materials (e.g., (Yin *et al.*, 2018)).

The availability of bio-derived ash, such as RHA and RSA, and the demand for pozzolonic materials is a strong motivator for expanding the understanding of use of such ashes in concrete. Research into the use of these ashes in the production of cement-based materials has grown by 10-fold in the past decade (Figure 1). This document presents an in-depth literature review that captures the current state of knowledge regarding use of RHA and RSA as SCMs. This includes consideration of the fundamental chemical and physical properties of the RHA and RSA, the performance of concrete mixtures containing RHA and RSA, and preliminary considerations for the environmental and economic impacts of RHA and RSA in concrete mixtures. A brief summary of the advantages and disadvantages of using RHA in concrete from this review is detailed in Table 1. While there is a rich set of literature on RHA in concrete, assessment of RSA in concrete is more limited (see Figure 1c).

Table 1. Advantages and disadvantages of use of rice hull ash in cement-based materials (based on (Mehta and Pirtz 1978; Mehta and Folliard 1995; Jenkins *et al.*, 1998; Sugita *et al.*, 1999; de Sensale 2006; Salas *et al.*, 2009; de Sensale 2010; Harish *et al.*, 2010; Chao-Lung *et al.*, 2011; Nguyen *et al.*, 2011; Antiohos *et al.*, 2014; Gastaldini *et al.*, 2014; Rêgo *et al.*, 2015; Soltani *et al.*, 2015; Venkatanarayanan and Rangaraju 2015; Caltrans 2016; He *et al.*, 2017))

Advantages	**Disadvantages**
- Broadly available agricultural resource	- Variability as a function of particular cultivation practices and variation in rice types
- Potential for benefits of both energy production and pozzolanic material production	- Necessitates controlled combustion to produce amorphous silica with pozzolanic behavior
- Potential alternative to pozzolans like silica fume, a byproduct of producing silicon metal or ferrosilicon alloys, for the production of high performance concrete in developing regions	- Commonly reported increased water requirements and reduced workability
- Reduced heat of hydration	- Increased setting time
- Reduced air content	- Capital investment in combustion facilities that would produce ash with desired properties
- Increased long-term strength	- Low density ash is more expensive to ship
- Improved durability (reduced chloride ingress, permeability, and alkali-silica reaction)	
- Potential beneficial contributions to carbon footprint of concrete	

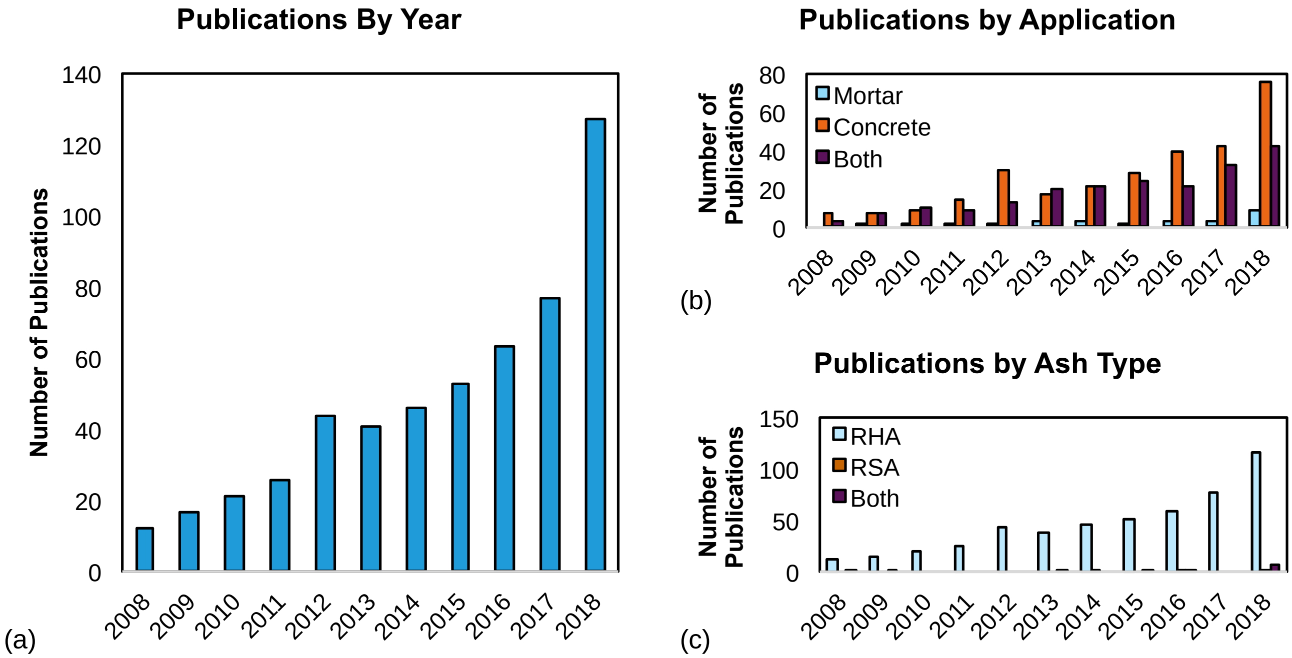


Figure 1. Number of publications annually on the use of rice-hull ash (RHA) and/or rice-straw ash (RSA) in concrete and mortar: (a) shows the total number of publications, (b) shows the breakdown in publications by application in a cement-based material, (c) shows the breakdown in publications by ash type. Data were obtained on February 26th, 2019 using the Web of Science Science Citation Index (SCI-EXPANDED) – 1900-present database with the keywords for the four topic searches being "Rice Husk Ash AND Concrete", "Rice Husk Ash AND Mortar", "Rice Straw Ash AND Concrete", and "Rice Straw Ash AND Mortar". the results were filtered to be "Materials Science", "Engineering", "Construction Building Technology", or "Science Technology Other Topics" research areas only.

Rice hulls can be used for a variety of other applications as well; however, not all are of high economic value (Brodt *et al.*, 2014). These applications range from use as bedding for livestock and as a soil amendment, to combusting this biomass as an energy source (Brodt *et al.*, 2014). It has also been suggested that the high silicate content of these ashes may make them suitable for the production of other materials, such as ceramic glazing, solar panels, and insulators (Sandhu and Siddique 2017).

2. Chemical and physical properties of rice–based ash

2.1. Rice ash composition

When appropriate processes have been implemented, RHA is highly reactive, more so than ordinary pozzolans (Mehta 1977). In fact, it can provide properties similar to those provided by use of silica fume

(Mehta and Monteiro 2006), which is often applied in the production of ultra-high performance concrete. Due to the similarities noted for RHA and silica fume, material property comparisons are often drawn between these two pozzolans. As a point of reference, the chemical analysis of several conventional SCMs, biomass ashes, as well as both RHA and RSA are shown in Table 2. As can be seen, the high silica content of the rice-based ash, especially the RHA, is similar to the chemical analysis of silica fume. The data used to obtain the averages for the RHA and RSA composition are plotted in Figure 2. This plot shows there is little variation in the silica content for the RHA (a range of 25% and standard deviation of 6%); however, a wider dispersion is present for RSA (a range of 52% and standard deviation of 22%) with higher levels of unwanted potassium oxide (up to 150 times more than that reported for RHA). The lower silica content and higher potassium content could lead to less favorable properties in concrete if not treated properly.

While the literature suggests several means to achieve desired pozzolanic reactivity from RHA, the literature is sparser with regard to RSA. Further, one of the potential benefits of use of biomass ash as a cementitious material is that it comes from the economically beneficial production of energy during combustion. However, to attain reactive silica in the rice-based ashes, low temperatures for combustion, which would be suboptimal for many energy production methods, are recommended. As such, developing means to both produce a viable pozzolanic material and energy resource from rice byproducts remains an open area of study.

Table 2. Chemical analysis of representative supplementary cementitious materials, biomass ash, as well as ranges for rice straw ash and rice hull ash (based on data from (Jenkins *et al.*, 1998; Taylor *et al.*, 2007; Ramezanianpour *et al.*, 2011))

Ash Type	Silicon dioxide (SiO_2)	Aluminum oxide (Al_2O_3)	Titanium oxide (TiO_2)	Ferric oxide (Fe_2O_3)	Calcium oxide (CaO)	Magnesium oxide (MgO)	Sodium oxide (Na_2O)	Potassium oxide (K_2O)	Phosphorus oxide (P_2O_5)	Sulphur trioxide (SO_3)
Class F fly ash	52	23	-	11	5	-	1	2	-	0.8
Class C fly ash	35	18	-	6	21	-	5.8	0.7	-	4.1
Ground slag	35	12	-	1	40	-	0.3	0.4	-	9
Silica fume	90	0.4	-	0.4	1.6	-	0.5	2.2	-	0.4
Calcined clay	58	29	-	4	1	-	0.2	2	-	0.5
Tuff (a natural pozzolan)	65.74	12.24	0.29	2.05	2.87	0.96	1.92	2.02	0.03	0
Willow wood	2.35	1.41	0.05	0.73	41.2	2.47	0.94	15	7.4	1.83
Demol. wood	45.91	15.55	2.09	12.02	13.51	2.55	1.13	2.14	0.94	2.45
Wheat straw	55.32	1.88	0.08	0.73	6.14	1.06	1.71	25.6	1.26	4.4
Sugar cane bagasse	46.61	17.69	2.63	14.14	4.47	3.33	0.79	0.15	2.72	2.08
Rice straw *	60.78	1.82	0.12	1.16	4.10	3.37	0.62	17.61	7.08	2.69
Rice hull *	88.51	0.28	0.02	0.44	1.03	0.47	0.33	2.60	0.63	0.49

"-" indicates no value reported

* represents averages from data reported in Figure 2

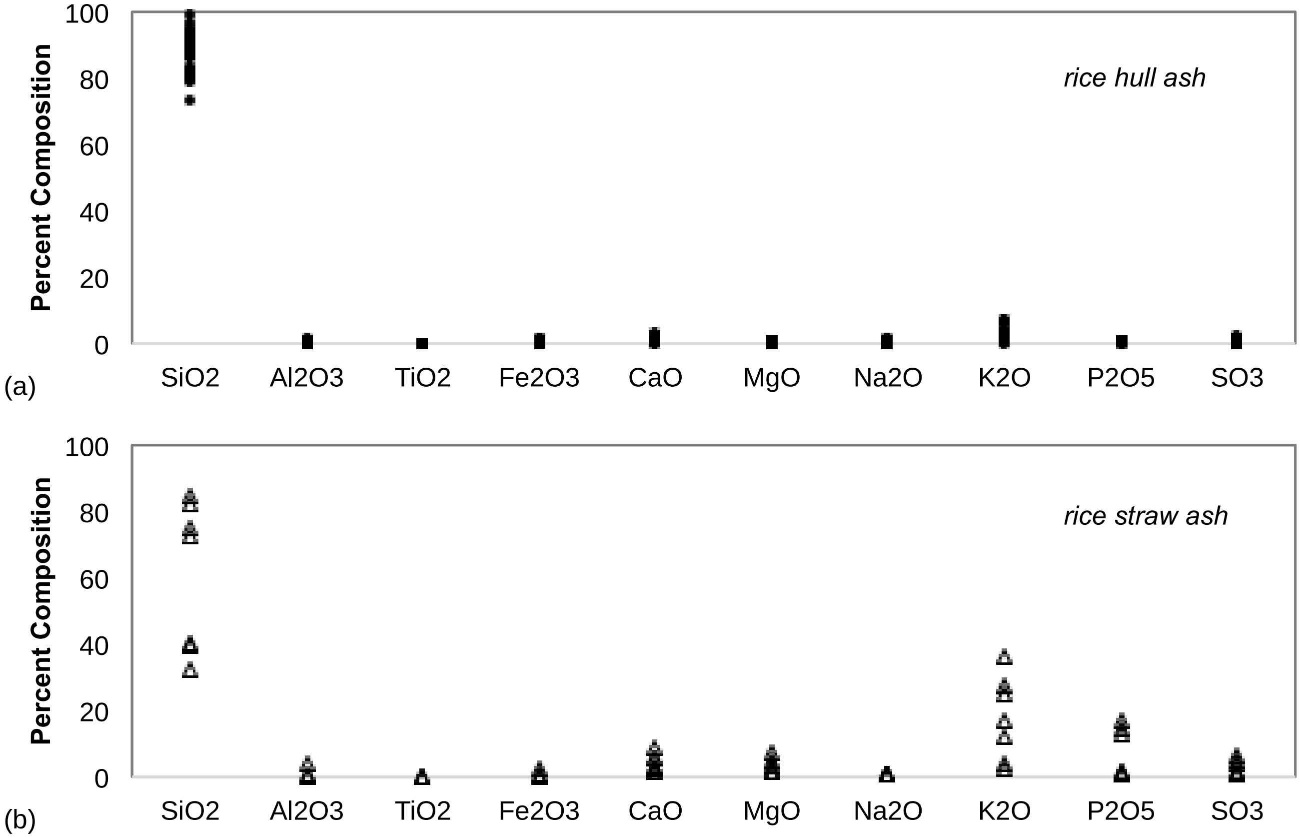


Figure 2. Ash compositions for (a) rice-hull ash as reported for 39 ashes and (b) rice straw ash as reported for 7 ashes (Jenkins *et al.*, 1998; Santos *et al.*, 1999; Hasparyk *et al.*, 2000; Bakker *et al.*, 2002; de Sensale 2006; Salas *et al.*, 2009; de Sensale 2010; Harish *et al.*, 2010; Chao-Lung *et al.*, 2011; Ramezanianpour *et al.*, 2011; Zain *et al.*, 2011; Xu *et al.*, 2012; Antiohos *et al.*, 2014; Gastaldini *et al.*, 2014; Bie *et al.*, 2015; Le *et al.*, 2015; Venkatanarayanan and Rangaraju 2015; Xu *et al.*, 2015; Fernandes *et al.*, 2016; Matalkah *et al.*, 2016; He *et al.*, 2017; Huang *et al.*, 2017; Roselló *et al.*, 2017; Zunino and Lopez 2017)

RHA can display the appropriate chemical composition, specifically high amorphous silica content with minimal undesirable compounds, to contribute to the mechanical and durability properties of concrete (Gursel *et al.*, 2016). Under appropriate conditions, RHA can act as a pozzolan, reacting with calcium hydroxide in the cement hydration process. These traits are dissimilar to many other forms of biomass ash, for which the presence of alkali metals, such as sodium and potassium, or high carbon contents can lead to deleterious effects in the microstructure and durability of concrete (Rajamma *et al.*, 2009). While considered in the 1970s-1990s as a potential SCM (e.g., (Mehta 1977)), state-of-the-practice at that time often resulted in RHA that did not have the desired pozzolanic properties for use as an SCM. However, current understanding identifies factors that can affect the ability for these bio-derived ashes to provide an alternative to conventional binders (outlined in Figure 3) so that they can be controlled; these parameters are discussed in more detail in the subsequent subsections.

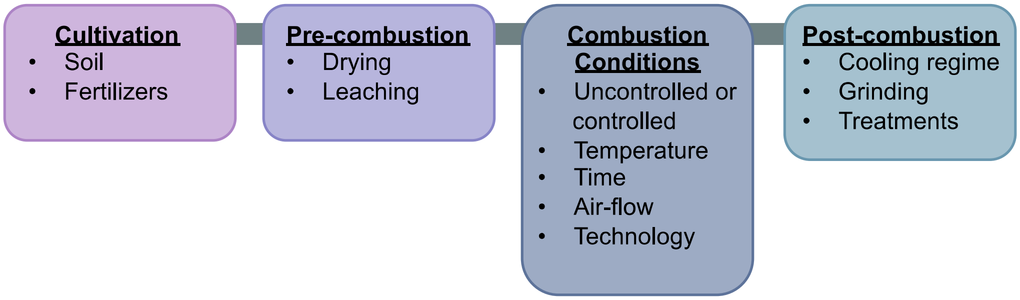


Figure 3. Flow diagram of parameters that can influence the reactivity of rice ash in cement-based materials

2.2. Rice cultivation

Cultivation practice for rice can have an influence on ash properties. The two primary additional biomass streams in rice cultivation beyond the food crop are rice hulls, the shells around rice kernels, and rice straw, the leaves and stems from the plants. In a typical crop, approximately 1-1.5 kg of rice straw and 0.2-0.28 kg of rice hull is produced for every 1 kg of rice paddy (Mehta and Monteiro 2006; Roselló *et al.*, 2017; Van Hung *et al.*, 2018). Both rice hulls and rice straw have relatively high ash contents: a reported 18% for rice straw and 20% for rice hulls compared to the 7% ash content for wheat straw, 2% for sugarcane bagasse, and 3% for hybrid poplar (Jenkins *et al.*, 1998). During rice cultivation, the plants absorb silica from the soil (Boateng and Skeete 1990), with higher silica soil leading to higher silica content ash (Khan *et al.,* 2018). Geographic and climate conditions can influence production and mineral contents of rice, and by association, their ashes. Further, the selection of fertilizers can alter the chemical composition of ashes from combustion, which can influence their usefulness in concrete (Zain *et al.,* 2011; Sandhu and Siddique 2017). For example, the addition of fertilizers with high potassium content can result in a higher level of potassium oxides in the rice ash (Zain *et al.,* 2011), which could contribute to deleterious effects in concrete durability if that ash is used as an SCM.

2.3. Pre-combustion treatment of rice biomass

The drying of biomass prior to combustion can improve certain properties of ash. These improvements include a reduction in remnant carbon (Zain *et al.,* 2011), which can influence demand for water and chemical admixtures in concrete batching (Harish *et al.,* 2010). Reductions in potassium content in biomass, and associated slag produced in combustion, can also be achieved through pre-heating treatments of feedstock (Thy *et al.,* 2005).

While leaching methods have predominantly been explored for their influence on energy production, they have the potential to play a role in the viability of rice-based ashes for use in cement-based materials because the presence of certain alkali metals can contribute to deleterious effects on concrete properties. Leaching biomass with water can reduce concentrations of alkali metals and chlorine (Jenkins *et al.*, 1998; Thy *et al.,* 2013). The leaching of these metals also pose potential benefits to the combustion process as their presence is often linked to the residual buildup of deposits in furnaces and boilers, leading to reduced efficiency over time (Thy *et al.*, 2006). Water leaching practice has been shown to increase the relative silicate content in rice straw ash by 20% (Jenkins *et al.,* 1998). While significant amounts of alkali metals can remain in the feedstock, in water leaching of rice straw, a reduction in unwanted sodium, chlorides, and potassium have been noted in the associated ash from combustion (Thy *et al.,* 2013). Leaching of biomass using acid solutions prior to their combustion has also been explored with researchers noting an increase in surface area and porosity of silica in ash (Bakar *et al.,* 2016) as well as improved stable pozzolanic activity (Salas *et al.,* 2009; Zunino and Lopez 2017). Further, leaching rice hulls with other acidic or alkali solutions has been reported as removing metallic impurities (Liu *et al.,* 2013; Shen 2017).

2.4. Combustion conditions

In the production of biomass ash for use as a pozzolanic material, the combustion practice (e.g., controlled or uncontrolled combustion, combustion temperature, combustion technology) can play a large role in the properties of the ash (Mehta and Monteiro 2006; Rajamma *et al.,* 2009; Chao-Lung *et al.,* 2011). As was noted, rice-based ashes have notably high silica content (Jenkins *et al.,* 1998; Binod *et al.,* 2010) which is desirable, but the combustion conditions can determine whether these silicates will be reactive in cementitious blends (Fernandes *et al.,* 2016). Large amorphous (non-crystalline) fractions of silicates are needed for ash to be reactive. In open-field combustion or uncontrolled combustion, higher residual carbon and glassy silica phases can occur in the ash (Antiohos *et al.,* 2014; Rêgo *et al.,* 2015); neither property is desirable for a pozzolanic material (Sugita *et al.,* 1999; Harish *et al.,* 2010). In these uncontrolled conditions ashes may need to be ground to a very fine particle size to achieve reactivity. However, under controlled conditions, it is easier to produce silica that retains a non-crystalline form and a cellular microstructure, with a large surface area, which facilitates pozzolanic reactions (Mehta and Monteiro 2006).

The production of high reactivity ash is a function of incineration temperature, time, and cooling regime, as well as the properties of the raw materials (Nair *et al.,* 2008). To create a reactive ash, typically, high surface area and high amorphous silica content are sought. Technology and processes used for combustion can further influence parameters of ash such as specific surface area and the structure of silica produced (e.g., glassy amorphous or partially crystalline) (Fernandes *et al.,* 2016). For example, several studies have suggested that production of RHA from suspension fired technology is favorable for the production of high amorphous silica content when compared to methods such as stoker and fluidized bed boilers (Gursel *et al.,* 2016; Prasara-A and Gheewala 2017), and most sources suggest that use of controlled combustion in boilers leads to more reactive ash than uncontrolled fire.

Reports on the most desirable temperature for combustion typically fall in the 500-700°C range (Nair *et al.,* 2008; Zain *et al.*, 2011; Xu *et al.*, 2012; Fernandes *et al.,* 2016; Roselló *et al.*, 2017), with 500-600°C being cited as the most desirable to form reactive amorphous silica (Nair *et al.,* 2008). At the upper temperatures in this range, i.e., at 700°C, some evidence of the formation of unwanted crystalline structures has been shown (Xu *et al.*, 2012). At higher temperatures, silica would be expected to become primarily crystalline, commonly forming cristobalite and tridymite (Sugita *et al.,* 1999; Mehta and Monteiro 2006). With such crystalline structures, additional post-processing, such as grinding, would be required to form reactive ash (Mehta and Monteiro 2006; Zain *et al.*, 2011). At temperatures that are too low (i.e., below ~400°C), there is a potential for remnant carbon (Sugita *et al.,* 1999; Nair *et al.*, 2008). It has been reported that an appropriate time for combustion would be over 12 hours if low temperatures are used (e.g., 500°C) (Nair *et al.,* 2008). For up to 12 hours of combustion time at these temperatures, there is an increasing surface area and pore volume in the ash. There is nearly equivalent surface area and pore volume reported for combustion times between 12-24 hours at these low temperatures. Yet, it must be noted that at short combustion times and low temperatures, there is the potential for remnant carbon content (Nair *et al.*, 2008). At even higher temperatures (e.g., over 1000°C), however, at combustion times as short as 5 minutes, the formation of crystalline silica can occur (see Table 3). The supply of air during combustion has been noted as a useful condition during burning (Boateng and Skeete 1990; Nair *et al.*, 2008). However, high air flow rates could lead to higher temperatures at the center of a fixed bed and contribute to increased crystalline silica formation (Hamad and Khattab 1981).

Table 3. Summary of the effects of combustion temperatures based on (Nair *et al.,* 2008)

Burn temperature	**Associated burn times**	**Desirable/undesirable properties**	**Additional considerations**
300 to 500°C	Over 12 hours	Remnant carbon	
500 to 700°C	12 to 24 hours	Predominantly amorphous silica	
700°C and over	Less than one hour	Crystal forms of silica	Need to grind to improve reactivity

2.5. Post-combustion treatments of rice-based ash

Treatments of rice-ash collected after combustion can additionally improve ash reactivity. Grinding is the most commonly discussed post-combustion treatment. It has been noted that increased grinding time results in no significant variations in chemical composition or crystalline mineralogy (Xu *et al.*, 2015). The literature suggests appropriate levels of grinding of ash can result in a more desirable SCM in concrete, and grinding and fineness can have a strong influence on RHA reactivity (Harish *et al.*, 2010; Antiohos *et al.*, 2014; Alex *et al.*, 2016). Xu *et al.* (2015) showed that a 30 minute grinding time for the ashes they tested resulted in high levels of hydration and improved dispersion of ash, which then led to reduced porosity of the paste. Zerbino *et al.* (2012) noted a more dense, homogenous paste with ground RHA than when unground RHA was used. Rego *et al.* (2015) found that crystalline RHA would typically remain unreactive in cement hydration and act as a filler in concrete and mortar, but that grinding could increase its pozzolanic activity. Venkatanarayanan and Rangaraju (2015) found that grinding of RHA improved bulk density for use in concrete, improved uniformity, and typically improved compressive strength in concrete. Yet, prolonged grinding times may not consistently lead to high fineness as particle aggregation can occur (Xu *et al.*, 2015).

The chilling process of ash can influence reactivity (Alex *et al.*, 2016). At slow cooling temperatures condensation reactions can occur, leading to a network with fewer reactive surface sites than would occur with rapid cooling (Nair *et al.*, 2008). While the larger particle size associated with slow cooling does not necessarily change the percent crystallinity or surface area, because of the very high meso- and micro-porosity, slow cooling does result in lower amounts of silanol groups resulting in a lower pozzolanic reactivity (Nair *et al.*, 2008). To a lesser extent, other post-combustion treatment methods have been explored such as the use of sodium hydroxide solutions to chemically modify RHA for use as a partial replacement for cement with promising results, including increased early-age compressive strength and reduced porosity (Prasittisopin and Trejo 2017).

3. Early-age properties of concrete using rice ash

A summary of the changes to the fresh properties of concrete by incorporating RHA is shown in Figure 4. Work on the fresh properties of concrete and mortar containing RHA have suggested some trends for the water requirements, workability, air content, heat of hydration, and setting time. Often, RHA is reported as increasing water demand and reducing concrete workability, which could lead to requirements for increased use of water or plasticizers to get concrete to place properly. Work has also suggested RHA contributes to a drop in air content and a rise in setting time; depending on application these factors may be undesirable due to effects such as durability properties and changes in constructability. However, studies have suggested RHA may contribute to reduced heat of hydration, which is often a desirable benefit of pozzolanic materials. Yet, studies verifying changes to these properties using RSA remain limited as do studies investigating the effects of RHA other fresh properties such as pumpability, and bleeding and segregation.

SCM Type	Class F fly ash	Class C fly ash	Ground slag	Silica fume	Calcined shale	Calcined clay	Metakaolin	Rice Hull Ash
Water requirements	⬇	⬇	⬇	⬆	▬	▬	⬆	⬆
Workability	⬆	⬆	⬆	⬇	⬆	⬆	⬇	⬇ *
Air content	⬇	⬇	⬇	⬇	▬	▬	⬇	⬇
Heat of hydration	⬇	⬍	⬇	▬	⬇	⬇	⬇	⬇
Setting time	⬆	⬍	⬆	▬	⬆	⬆	▬	⬆ **

⬆ Increased
⬇ Reduced
⬍ Change varies
▬ No significant change

Notes:
* Harish 2010 noted variability for workability when using ground RHA
** de Sensale 2018 noted variability

Figure 4. Changes to fresh properties of concrete with conventional mineral admixtures and with rice hull ash (figure based on (Taylor *et al.*, 2007); trends for RHA based on data and descriptions of individual findings in Appendix A)

3.1. Workability

The literature notes the use of RHA can change workability of concrete mixtures as a function of several parameters. In several cases, an increase in water demand has been shown for RHA concretes (Mehta 1977; Salas *et al.,* 2009; Chao-Lung *et al.,* 2011; Zain *et al.,* 2011; Antiohos *et al.,* 2014; Venkatanarayanan and Rangaraju 2015), a factor noted for both unground and ground RHA (Chao-Lung *et al.,* 2011; Venkatanarayanan and Rangaraju 2015). For example, Sousa Coutinho (2003) noted that use of the same quantity of water and superplasticizer for mortars with and without RHA led to over an 85% reduction in slump when RHA was used as a partial replacement of cement. This increase in water demand has been attributed to several factors such as the cellular structure of the ash (Zain *et al.*, 2011), which contributes to a high specific surface area, and the hydrophilic nature of the RHA (Antiohos *et al.*, 2014). Some sources do report a reduced water demand with the inclusion of RHA (e.g., (Givi *et al.,* 2010)), which could be a function of improved particle packing and flow characteristics of the RHA concrete (Antiohos *et al.*, 2014). Mehta and Pirtz (1978) reported that the slump test might not be a good indicator of the workability of the RHA concrete. They noted that even with very low slump, the RHA concretes displayed cohesiveness and excellent workability. Additionally, the literature suggests that the amount of additional water or superplasticizer that might be used to alter flow of RHA concretes is less than what would be needed for silica fume concrete (Santos *et al.*, 1999; Venkatanarayanan and Rangaraju 2015). While it is frequently a less economically desirable use, rice-based ashes can also be utilized as a partial aggregate replacement (as opposed to a partial replacement for cement). However, an even greater water demand has been noted for use of RHA as a partial replacement of aggregates (Antiohos *et al.,* 2014).

3.2. Hydration

The literature commonly indicates there is an increased setting time when RHA is incorporated (Venkatanarayanan and Rangaraju 2015; Arel and Aydin 2018). While some work has suggested that the level of grinding does not influence setting time (Venkatanarayanan and Rangaraju 2015), other studies have shown that RHA from uncontrolled combustion may delay setting whereas RHA from controlled combustion can shorten the setting time (de Sensale and Rodríguez Viacava 2018). While there is less literature on the use of RSA, the use of RSA from uncontrolled combustion has also been shown to delay setting times with higher amounts of RSA use (Munshi *et al.*, 2013).

The hydration process of RHA concrete mixtures has several characteristics that vary from typical portland cement hydration. Similar to other pozzolans, the use of RHA has been shown to reduce head of hydration (Mehta and Pirtz 1978). Research indicates the use of RHA can lead to a dilution effect, a chemical effect, and absorption of water that would not be present as such in typical portland cement. Additionally, RHA particles can act as nucleation sites, accelerating cement hydration (Park *et al.,* 2016). Pozzolanic RHA is reactive in the alkaline environment of cement paste (Antiohos *et al.*, 2014; Rêgo *et al.*, 2015; Xu *et al.,* 2015; Zunino and Lopez 2017). Its advantageous ability to deplete available calcium hydroxide has been shown to be equivalent or superior to that of conventional SCMs, such as fly ash and silica fume (Antiohos *et al.*, 2014; Venkatanarayanan and Rangaraju 2015; Xu *et al.*, 2015), with an increased level of RHA corresponding to an increased depletion of calcium hydroxide (de Sensale and Rodríguez Viacava 2018). Some authors have suggested that there is some internal curing as a function of the porous structure of RHA releasing some water during the curing process (e.g., (de Sensale *et al.*, 2008; Nguyen *et al.*, 2010)).

4. Mechanical properties of concrete with rice ash

4.1. Compressive strength

Unlike woody-biomass ash, which has been shown to lead to either no notable increase in concrete strength or deteriorating strength (Rajamma *et al.*, 2009), the use of RHA as a partial replacement for portland cement has been shown to increase concrete compressive strength (Mehta 1977; Mehta and Pirtz 1978; Santos *et al.*, 1999; Sugita *et al.*, 1999; de Sensale 2006; Salas *et al.*, 2009; Chao-Lung *et al.*, 2011; Gastaldini *et al.,* 2014; Bie *et al.,* 2015; Rêgo *et al.*, 2015; Venkatanarayanan and Rangaraju 2015; He *et al.,* 2017; Zareei *et al.,* 2017; Arel and Aydin 2018). While the majority of the literature suggests higher compressive strength of RHA concrete compared to portland cement concretes at later ages, the literature on early-age strength is more varied, with some sources suggesting increased early age strength (e.g., (Mehta 1977; Mehta and Pirtz 1978; de Sensale 2006; Venkatanarayanan and Rangaraju 2015)) and others reduced early-age strength (e.g., (Santos *et al.,* 1999)), as shown in Figure 5. Using data from Chao-Lung *et al.* (2011), the lower strengths sometimes exhibited from partial replacement portland cement at early ages are displayed, namely, a 5-15% reduction at 7 day strength in this case. However, the lower replacement ratios (i.e., 10% and 20%) exhibit a 5-10% higher 28-day strength and 0-3% higher 90-day strength. Several authors have noted an optimal level of RHA use in the cementitious binder, typically ranging between 7.5-20% (Harish *et al.,* 2010; Antiohos *et al.,* 2014; Gastaldini *et al.,* 2014; Bie *et al.,* 2015; Venkatanarayanan and Rangaraju 2015; He *et al.,* 2017; Zareei *et al.,* 2017), as can be seen in Figure 5. While there is greater variability in the literature, concrete mixtures from three authors are plotted in Figure 5b and these mixtures display an optimized 28-day strength with a 10% RHA replacement of portland cement. At this replacement level, the authors show a 5 to 8 MPa increase in strength through use of RHA relative to use of portland cement as the sole binder. However, the optimal use of the RHA can vary depending on parameters such as the RHA characteristics and the type of cement (Jamil *et al.,* 2013).

While many properties of RHA are similar to the conventionally used silica fume, this optimal replacement level of portland cement is higher than what is typically seen for silica fume (Harish *et al.,* 2010; Gastaldini *et al.,* 2014). This variability, as well as variability noted in strength values, could be attributed to variation in RHA properties, particularly the reactive amorphous silica content ranges found between literature sources (Venkatanarayanan and Rangaraju 2015). Such variability in the biomass ash can result from the factors discussed in Section 2. For example, Khan *et al.* (2012) showed that more crystalline RHA acted more as a filler than a reactant in the hydration process; it still contributed to compressive strength, but not to the same extent that would be expected from a pozzolanic reaction. Further, the size of RHA makes it an exceptional filler in concrete, filling in micro-voids and improving concrete strength (Soltani *et al.*, 2015).

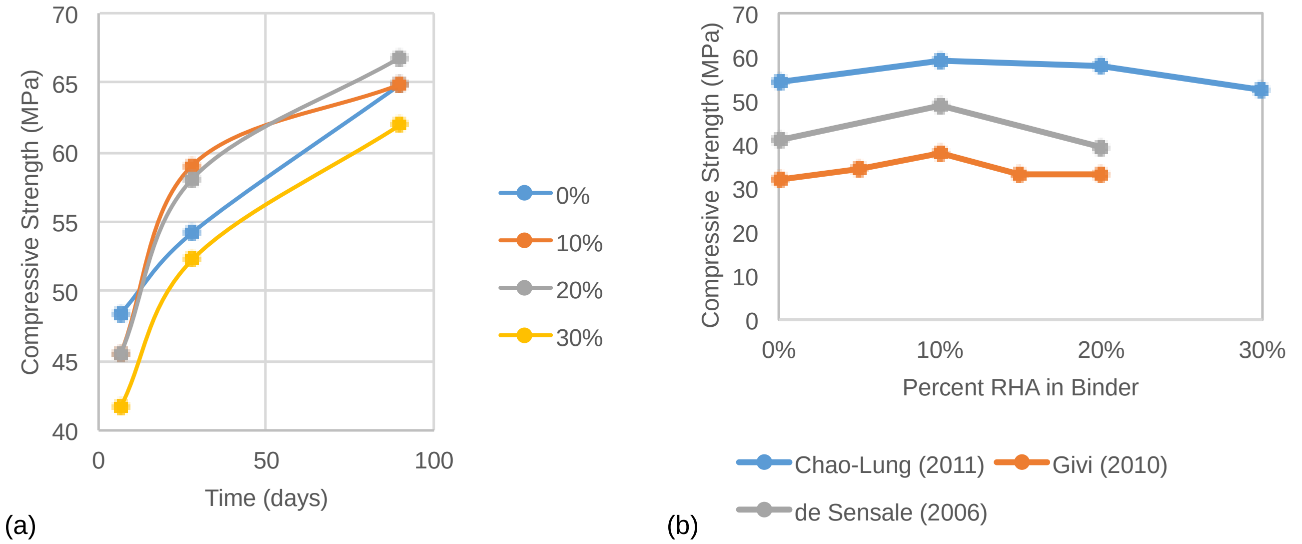
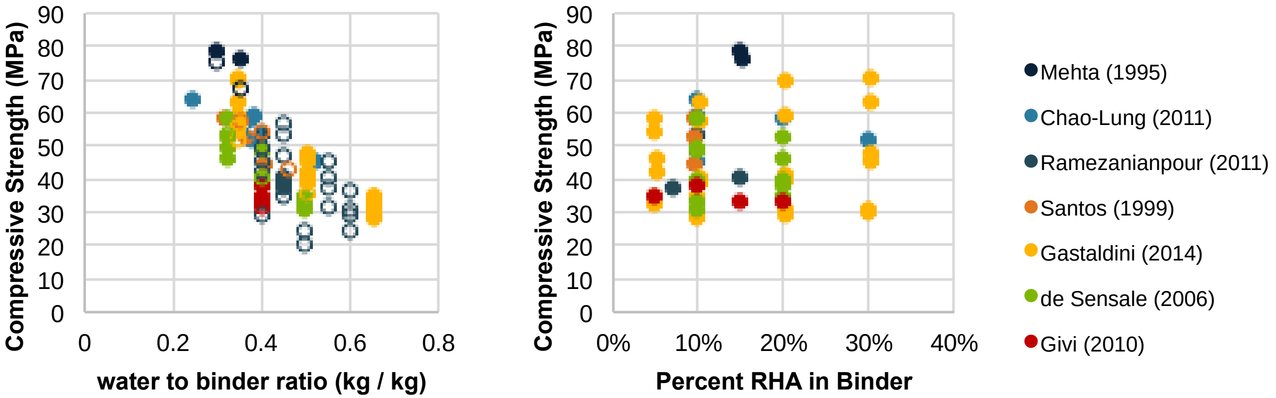


Figure 5. Compressive strength as a function of (a) curing age for 0 to 30% RHA replacement of cement (data from (Chao-Lung *et al.*, 2011)) and as a function of (b) percent rice hull ash (RHA) as a partial replacement for cement in ~0.40 water to binder ratio concrete mixtures at 28 days (de Sensale 2006; Givi *et al.*, 2010; Chao-Lung *et al.*, 2011). (Note: for mixtures presented, strengths are adjusted to account for differences in specimen dimensions using equations from (Yi *et al.*, 2006)).

The combustion conditions, pre-combustion treatments, and post-combustion treatments applied in ash preparation can influence the compressive strength obtained in RHA-concrete. While controlled combustion has been shown to contribute to improved ash reactivity, authors still report some favorable use of RHA in concrete from uncontrolled conditions (Rêgo *et al.*, 2015; Zunino and Lopez 2017). At combustion temperatures that facilitate the formation of amorphous silicates, there is a beneficial effect of RHA on concrete strength, but this is not consistently the case for ash from high, crystalline producing, temperatures for which different or no replacement levels may be desirable (Xu *et al.*, 2012; Bie *et al.*, 2015). Pre-combustion treatments, such as acid leaching, can help contribute to desirable ash properties, leading to ash with similar contributions to strength as silica fume (Salas *et al.*, 2009). Post-combustion treatments, such as alkali treatment can improve properties such as early-age strength development (Prasittisopin and Trejo 2017). The post-combustion treatment of grinding ash is reported to influence concrete compressive strength; often a decrease RHA particle size is associated with production of higher strength concrete (Zain *et al.*, 2011; Antiohos *et al.*, 2014; Venkatanarayanan and Rangaraju 2015; Alex *et al.*, 2016; Ambedkar *et al.*, 2017). Appropriate grinding, as with appropriate pre-combustion techniques, has been reported as resulting in RHA with similar contributions to strength as highly desirable silica fume at equivalent replacement levels (Venkatanarayanan and Rangaraju 2015). However, it has also been noted that over-grinding of RHA can lead to particle aggregation and a depreciating benefit to compressive strength (Xu *et al.*, 2015).

Additional considerations must be made for batching decisions and their influence on the favorability of RHA in concrete. Mehta (1977) established that there was no difference in concrete mechanical behavior whether RHA was blended or interground with cement. However, the water-to-binder ratio can influence the ratio of strength gain with increasing RHA content (Sugita *et al.*, 1999) and can influence the optimal level of RHA replacement (Gastaldini *et al.*, 2014). The effects on concrete strength when RHA is used as an SCM are shown in Figure 6. It can be seen from these mixtures that achieving equivalent or higher strength concrete with partial replacement of portland cement by RHA is achievable at various water to binder ratios. As Figure 6 also shows, a wide range of compressive strengths can be achieved at various replacement levels by tailoring factors such as water to binder ratio, moist-curing period, and ash properties, among others.

While not as well reported in the literature, RSA has also shown the ability to improve strength of cement-based materials. Again, an optimal level is reported for this ash type (Munshi *et al.*, 2013; Roselló *et al.*, 2017).



(a) (b)

Figure 6. Compressive strength of (a) concrete mixtures with (●) and without rice-hull ash (RHA) (○) as a function of varying water-to-binder ratios and (b) concrete mixtures with RHA as a function of percent RHA in the cementitious binder (Data from: (Mehta and Folliard 1995; Santos *et al.*, 1999; de Sensale 2006; Givi *et al.*, 2010; Chao-Lung *et al.*, 2011; Ramezanianpour *et al.*, 2011; Gastaldini *et al.*, 2014); strengths adjusted to reflect differences in test specimen size using equations presented by (Yi *et al.*, 2006))

4.2. Tensile strength

In the assessment of split cylinder tensile strengths, similar trends are present for the incorporation of RHA as were present for compressive strength. Some authors note an increase in early age strength (e.g., (Zareei *et al.*, 2017)), whereas others show a beneficial contribution to strength only at later ages (e.g., (Alex *et al.*, 2016)). Optimal levels of RHA use are noted (Zareei *et al.*, 2017). Controlled combustion leads to improved tensile strength compared to use of ash made from uncontrolled combustion (de Sensale 2006). The split cylinder tensile strengths are dependent on water-to-binder ratios, for which, at low water-to-binder ratios, the use of ash from uncontrolled combustion was shown to have the ability to lead to reduced strength, whereas ash from controlled combustion led to equivalent or improved tensile strength at varying water to binder ratios (de Sensale 2006). Additionally, grinding of ash can improve properties: while use of either ground or unground RHA was shown to improve tensile strength over a mixture containing solely portland cement as the binder, only the use of ground RHA was similar to that of silica fume (Venkatanarayanan and Rangaraju 2015). Again, limited data are available on the use of RSA in the assessment of changes to tensile strength, but increases in compressive strength in cement-based materials through use of RSA may be indicative of potential for their ability to contribute to tensile strength.

4.3. Flexural strength

The literature suggests flexural strength of concrete prepared with RHA is dependent on many of the same parameters as were noted for compressive strength. Bie *et al.* (2015) noted a temperature dependence for ash production and flexural strength achieved: at 600°C, below the temperature at which crystalline structures would be expected to form, all levels of RHA led to increased flexural strength; at 700°C, use of RHA content at 5% led to reduced strength, although this loss was not noted at other replacement levels. Venkatanarayanan and Rangaraju (2015) noted that use of RHA increased flexural strength compared to a mixture with only portland cement as the binder whether or not the RHA was ground after combustion; however, only the ground RHA led to similar properties as silica fume at similar replacement levels, which was used as a reference material. Also, Salas *et al.* (2009) showed the use of an acid pre-combustion treatment lead to RHA contributing similar flexural strength properties as silica fume (again, used as a reference material).

4.4. Modulus of elasticity

The literature suggests a positive relationship between the use of RHA in concrete and an increase in modulus of elasticity. Trends in benefits to modulus of elasticity are reported as similar to changes noted in compressive strength (Venkatanarayanan and Rangaraju 2015; He *et al.*, 2017; Zareei *et al.*, 2017). Again, there is a dependency on processing conditions, such as grinding, (Venkatanarayanan and Rangaraju 2015), dependency on water-to-binder ratio (He *et al.*, 2017), and suggestion of optimal ratios of RHA use (Zareei *et al.*, 2017).

The mechanical properties of concrete and mortar containing RHA are better characterized in the literature than other material properties, such as those that would affect durability. However, more work on assessing the effects of RSA and appropriate ash treatment methods to attain specified mechanical properties would elevate the literature.

5. Durability properties of concrete with rice ash

In addition to contributions to mechanical properties, the use of RHA in concrete is often reported as improving durability properties (Salas *et al.*, 2009; Soltani *et al.*, 2015) with improvements similar to those noted for conventional pozzolans (Ramezanianpour *et al.*, 2011). As with the literature on mechanical properties, the majority of the literature on the effects on durability using rice-based ashes focus on use of RHA and limited results are present for the effects of RSA.

5.1. Permeability

The use of RHA can lead to reduced permeability and porosity. If not fully reactive, the small particles of RHA can act as a filler, strengthening the packing between concrete constituents, reducing porosity and permeability (Antiohos *et al.*, 2014). Reactive RHA used as an SCM can further reduce pores a result of the calcium silicate hydrate gel formed through cement hydration and the secondary pozzolanic reaction of the RHA and the calcium hydroxide (He *et al.*, 2017). However, use of unground RHA can lead to unchanged or even increased porosity; an appropriate level of grinding to create small particles, without aggregation of particles, can contribute to reduced porosity relative to a solely portland cement paste (Venkatanarayanan and Rangaraju 2015; Xu *et al.*, 2015). Reduced porosity can lower water uptake (Venkatanarayanan and Rangaraju 2015) and trends of reduced water absorption, or sorptivity, have been reported as a function of increased RHA content (Sugita *et al.*, 1999; Sousa Coutinho 2003; Antiohos *et al.*, 2013; Zareei *et al.*, 2017). As with the mechanical properties, some have suggested there may be an optimal level of RHA replacement beyond which additional reduction in porosity may not be as readily achieved (Venkatanarayanan and Rangaraju 2015). Further, the use of additional treatments to RHA, such as the addition of NaOH, can contribute to beneficial reductions in porosity relative to control mixtures (Prasittisopin and Trejo 2017).

As with water uptake, it has been shown that the use of RHA can reduce air permeability (Sugita *et al.*, 1999; de Sensale 2006, 2010). The use of ash from controlled combustion has been shown to provide more notable improvements to air permeability than ash from uncontrolled combustion (de

Sensale 2010). Additionally, at high water-to-binder ratios, it has been reported that replacement with RHA results in reduced permeability (for ash from both controlled and uncontrolled combustion); at low water-to-binder ratios, the use of RHA from controlled combustion results in reduced permeability, but RHA from uncontrolled combustion leads to increased permeability (de Sensale 2006). Specifically, de Sensale (2006) found use of RHA from controlled combustion can lead to a 50 to 95% reduction in permeability coefficient, but RHA from uncontrolled combustion can result in a 95% reduction in permeability to a 150% increase in permeability coefficient depending on the water to binder ratio used.

5.2. Chloride ingress

Several authors have reported a reduced degree of chloride penetration as a result of the inclusion of RHA in concrete (Sugita *et al.*, 1999; Antiohos *et al.*, 2014; Gastaldini *et al.*, 2014). Venkatanarayanan and Rangaraju (2015) showed a reduction in rapid chloride ion permeability with the use of ground RHA relative to unground RHA. Additionally, at high water-to-binder ratios, the use of either RHA from controlled or uncontrolled combustion have been reported as reducing chloride penetration; however, at lower water-to-binder ratios, the use of RHA from uncontrolled combustion was reported as increasing chloride ion penetration and the use of RHA from controlled combustion was shown to be equivalent to that of the control concrete with portland cement (de Sensale 2010). Similar studies have shown at higher water to binder ratios, the use of untreated rice ash does not consistently reduce chloride ion penetration relative to concrete with portland cement as the sole binder (Antiohos *et al.*, 2013). Examples of the effects of RHA on concrete material durability, including chloride ingress, are displayed in Figure 7. From this plotted example by Mehta and Folliard (1995), the use of 15% RHA replacement of portland cement resulted in a 75 to 85% reduction in chloride permeability.

5.3. Alkali-silica reaction

Past literature has shown that biomass ash can contribute to reduced alkali-silica reaction in mortars (Esteves *et al.*, 2012) and most of the literature suggests the use of RHA in mortars and concrete appear to follow the same beneficial trend. Significant reduction in expansion as a result of alkali-silica reactions have been noted from several studies as a function of incorporating RHA in mixtures (Mehta 1977; Mehta and Folliard 1995; Hasparyk *et al.*, 2000; de Sensale 2010; de Sensale and Rodríguez Viacava 2018). Mehta (1977) showed that the use of RHA could be more effective than conventional pozzolans. In Mehta's study, 10% replacement of cement by weight with RHA resulted in similar reductions to expansion from 25% replacement by weight of calcined shale. An example of reductions in alkali-silica expansion is depicted in Figure 7. The use of RHA from controlled combustion has been shown to better contribute to reduced expansion from alkali-silica reactions than ash from uncontrolled combustion (de Sensale 2010). Grinding of RHA has also been shown to better contribute to reduction in expansions (Zerbino *et al.*, 2012). Yet depending on the composition and morphology of the RHA, there is a potential for RHA to contribute to increased alkali-silica reaction (Zerbino *et al.*, 2012). However, based on the literature, use of factors such as proper combustion and grinding could limit this issue.

5.4. Deterioration under varying pH levels

Sulfate attack causes the pH of the concrete pore water to fall, reversing the chemical reactions that give concrete its strength. Unlike portland cement concrete, which looses strength, the use of RHA can lead to an increase in strength under alkaline curing conditions (Ambedkar *et al.*, 2017). Even with use of a high crystalline content RHA, the ability for the ash to act as a filler can lead to improved chemical resistance in concrete mixtures tested in aggressive salt solutions (Khan *et al.*, 2012). RHA has been shown to reduce the expansion of concrete and mortar due to sulfate attack (Mehta and Folliard 1995; de Sensale 2010). This reduction was noted as being approximately 45% for mortar containing 10% RHA replacement of cement at 4 weeks and a reduction of over 80% was noted for the 10% RHA mortar at 16 weeks in a study by Mehta and Folliard (1995) (see Figure 7).

The literature suggests the use of RHA can assist in improving concrete resistance to acid exposures. Ambedkar *et al.* (2017) showed concrete mixtures containing RHA exhibited a lower loss in strength than control concrete when cured in an acid solution. Further, it has been shown that mortar or concrete containing RHA exhibited a large reduction in mass loss after exposure to a hydrochloric-acid solution (Mehta and Folliard 1995; Sugita *et al.,* 1999; de Sensale 2010).

5.5. Shrinkage

The reported contributions of RHA to concrete shrinkage vary. Gastaldini *et al.* (2014) showed a reduction in total shrinkage through use of RHA, but greater levels of cement replacement with RHA led to higher shrinkage than the lower levels of use. de Sensale *et al.* (2008) and Nguyen *et al.* (2010) found a reduction in autogenous shrinkage through use of RHA. Both Gastaldini *et al.* (2014) and Mahmud *et al.* (2005) identified similar behavior of RHA concrete to silica fume concrete, but Mahmud *et al.* (2005) reported increased shrinkage relative to the control concrete. Habeeb and Fayyadh (2009) also reported increased shrinkage with use of RHA, but attributed this shrinkage to the microfine particle size of the RHA used.

5.6. Other durability parameters

In addition to the contributions of RHA to reducing susceptibility of concrete to acid solutions, alkali-silica reaction, and chloride ingress as well as to lowering permeability, the use of RHA has been reported to aid in other concrete durability parameters. The pozzolanic reaction from RHA can increase the electrical resistance of concrete, a non-destructive testing method for assessing susceptibility for corrosion of rebar to take place in concrete (Chao-Lung *et al.,* 2011). The use of RHA in concrete can lead to improved frost resistance in non-air-entrained concrete mixtures, with higher contribution to frost resistance than similar concrete mixtures containing silica fume (Mehta and Folliard 1995). Some results have indicated that RHA can contribute to a reduced rate of carbonation (Sugita *et al.,* 1999); yet others have suggested that compared to a mixture containing portland cement as the sole binder, that use of RHA increases susceptibility to carbonation (Antiohos *et al.,* 2013). Use of RHA in concrete can also reduce the creep strain of concrete (He *et al.,* 2017). While these durability parameters are not as thoroughly discussed in the literature, current reports suggest there is a strong ability for RHA to improve many durability properties of concrete.

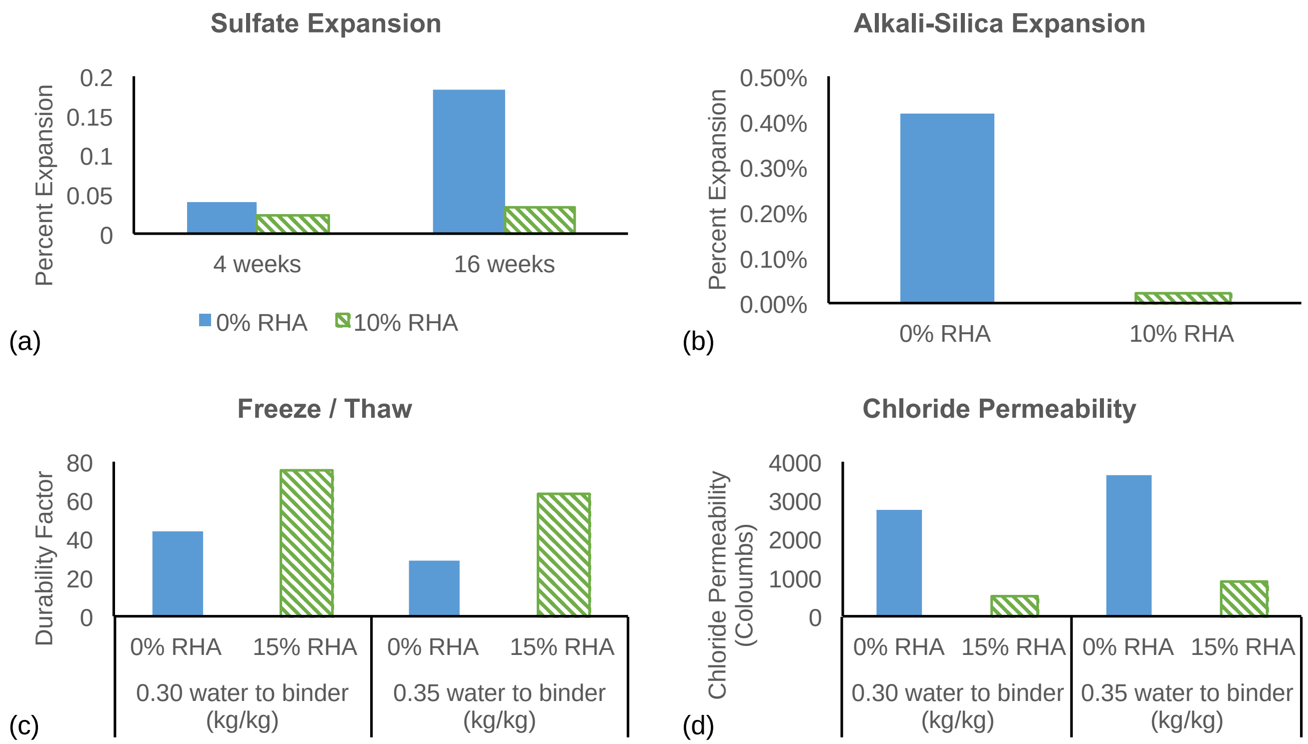


Figure 7. The effects of use of rice hull ash (RHA) on (a) the sulfate expansion and (b) alkali-silica expansion in mortars as well as the effects of RHA on (c) freeze-thaw durability and (d) chloride permeability of concrete. (Data from (Mehta and Folliard 1995))

As can be seen in Figure 8, RHA contributes many similar properties to conventional SCMs in concrete. RHA is often reported as leading to increased long-term strength as well as reducing concrete permeability and chloride ingress. Use of RHA is shown to aid in reductions in alkali-silica reaction induced expansion and it has been reported to improve sulfate resistance. Reports suggest that, unlike other SCMs, RHA may provide a beneficial contribution to freeze/thaw behavior. There have been varied results for concrete shrinkage using RHA, whereas for conventional SCMs, these parameters have been reported as not changing. Yet, further analysis into parameters that influence the durability properties of concrete for which the literature is less conclusive is still needed.

SCM Type	Class F fly ash	Class C fly ash	Ground slag	Silica fume	Calcined shale	Calcined clay	Metakaolin	Rice Hull Ash
Early Strength	↓	—	↓	↑	↓	↓	↑	↕
Long-term strength	↑	↑	↑	↑	↑	↑	↑	↑ *
Permeability	↓	↓	↓	↓	↓	↓	↓	↓ **
Chloride ingress	↓	↓	↓	↓	↓	↓	↓	↓ ***
Alkali Silica Reaction	↓	↕	↓	↓	↓	↓	↓	↓ ****
Sulfate resistance	↑	↕	↑	↑	↑	↑	↑	↑
Freezing and thawing resistance	—	—	—	—	—	—	—	↑
Drying shrinkage	—	—	—	—	—	—	—	↕

↑ Increased
↓ Reduced
↕ Change varies
— No significant change

Notes:
* Alex 2016 showed variation, Chao-Lung 2011 showed no change, Salas 2009 showed no change for the uncontrolled combustion RHA, Bie 2015 showed variation for the RHA combusted at 700C, Zerbino 2012 showed reduction
** de Sensale 2006 showed variation for the uncontrolled combustion RHA
*** Antiohos 2013 showed similar or slightly decreased resistance to penetration
**** Zerbino 2012 showed increase

Figure 8. Changes to hardened and durability properties of concrete with conventional mineral admixtures and with rice hull ash (figure based on (Taylor *et al.*, 2007); trends for RHA based on data and descriptions of individual findings in Appendix B)

6. Specialized concrete mixtures with rice hull ash

6.1. High- and ultra-high-performance concrete

As noted, RHA has similar chemical composition and high surface area to silica fume, a common constituent in high performance and ultra-high performance concrete. Reports indicate that RHA can increase the degree of cement hydration in ultra-high performance concrete at later ages (Nguyen *et al.*, 2011). Nguyen *et al.* (2011) show that the use of RHA, while not leading to high early strength, resulted in higher strengths at 7-days and beyond, when compared to the control concrete, as well as resulted in higher ultimate strengths than mixtures containing silica fume. The delayed curing was in part attributed to internal curing from RHA releasing water (Nguyen *et al.*, 2011). It is suggested that RHA is suitable for use in ultra-high performance concrete and could benefit regions where RHA is available and silica fume is too expensive to obtain (Nguyen *et al.*, 2011; Le *et al.*, 2015). Further, the use of RHA in reactive powder concrete, a type of ultra-high-performance concrete, has been reported to lead to increased 28-day strength, reduced porosity, and reduced chloride ingress (Vigneshwari *et al.*, 2018). Similar to RHA concrete, RHA-ultra-high-performance concrete can exhibit an optimal level of cement replacement (Vigneshwari *et al.*, 2018). Additionally, the combination of RHA with silica fume has been shown to enhance compressive strength and impermeability of ultra-high performance concrete (Huang *et al.*, 2017). Ternary blends with Metakaolin have also been found to provide desirable properties for RHA concrete (Shatat 2016). Additionally, it was reported that curing methods, namely steam curing, can further improve mechanical and some durability properties of RHA ultra-high-performance concrete (Vigneshwari *et al.*, 2018).

6.2. Self-compacting concrete

Use of high amorphous silica-content RHA has been reported to be a suitable pozzolanic material in the formation of self-compacting concrete (Sandhu and Siddique 2017). The use of RHA can facilitate desired long-term strength, increased splitting tensile strength, increased flexural strength, and reduced porosity (Sandhu and Siddique 2017). These beneficial properties are attributed to an improved microstructure and pore structure in concrete (Sandhu and Siddique 2017). Use of RHA in self-compacting concrete has also been found to lead to reduced workability (Sandhu and Siddique 2017), a parameter noted previously in the production of RHA concrete. The use of RHA predominantly has been reported to lead to increased yield stress and viscosity of mortar compared to a mixture containing only portland cement as the binder, factors that seemed to increase with increased RHA content and particle size (Le *et al.*, 2015). The incorporation of either silica fume or RHA was found to decrease the shear thickening over time, with greater reductions noted with use of silica fume (Le *et al.*, 2015). Additionally, rheology of the mixtures containing RHA as a partial replacement of cement with quarry dust as a partial replacement of fine aggregate was not significantly different from a code-specified self-compacting mortar mixture when superplasticizer and viscosity modifying agents were used (Gowda *et al.*, 2011).

6.3. Alkali-activated materials

The use of alkali-activated materials (AAMs) has been proposed for decades as a material alternative to cements with the potential to lead to lower environmental impacts, specifically GHG emissions, by removing the need for portland cement (Miller *et al.,* 2018b; Provis 2018). However, research on the use of RHA in AAMs is limited. Reports show that RHA can provide adequate silica for the production of AAMs; however, it, like many other biomass ashes, lacks the necessary aluminous compounds and needs to be blended with another solid precursor, such as ground granulated blast furnace slag, to form an AAM (Villaquirán-Caicedo *et al.,* 2015; Matalkah *et al.,* 2016; Patel and Shah 2018). Due to limited studies, the desirable composition of RHA, aluminous compound source, and alkali-activator for improved properties has not been specified in the literature. However, Villaquirán-Caicedo *et al.* (2015) showed that the use of RHA with Metakaolin and a potassium hydroxide activator in an AAM was able to contribute to lower bulk density and lower thermal conductivity than the other AAMs investigated in their study, which could lead to other benefits from their use. Additionally, Patel and Shah (2018) attempted use of RHA with ground granulated blast furnace slag and an alkali-activator of sodium hydroxide and sodium silicate to produce an AAM. Similar to some findings from the production of RHA concrete, the authors noted a decrease in workability and an optimum level of RHA beyond which hardened properties did not improve.

7. Environmental impacts of rice-based ash and its use in concrete

There are high demands for the production of cement and cementitious materials for use in concrete, with annual hydraulic cement production exceeding 4 billion metric tons annually (van Oss 2018). The GHG emissions from cement production are of global concern (IEA 2018). One of the primary proposed means for reducing GHG emissions, from portland cement production has been the use of alternative materials that can partially replace the cement needed (Scrivener *et al.,* 2017). However, the availability of the conventionally used SCMs are becoming limited, which is encouraging research on the use of alternative SCMs and alternative cements (Miller *et al.,* 2018b); such alternative materials include biomass resources. With any alternative material, environmental impact assessments that quantify potential benefits or drawbacks to use are necessary.

Environmental impact assessments of rice combustion and ash products are limited (Gursel *et al.,* 2016; Prasara-A and Gheewala 2017). More thorough assessment of potential benefits and drawbacks from an environmental, social, and economic impacts viewpoint for RHA and RSA is needed (Prasara-A and Gheewala 2017). Some work in the area has examined the environmental impacts of conversion of rice hull to energy sources, such as electricity, syngas, or bio-ethanol (Prasara-A and Grant 2011; Shafie *et al.,* 2012; Silalertruksa *et al.,* 2013). It has been shown that the use of RHA in products such as lightweight concrete blocks (Prasara-A and Grant 2011) and in mortars (Moraes *et al.,* 2010) could lead to reduced environmental impacts. As was noted by Prasara-A and Grant (2011) the benefits noted in use of RHA in concrete blocks, such as reduced GHG emissions, could be attributed to offsetting the high impacts associated with the production of portland cement. Also, noteworthy in the study by Prasara-A and Grant (2011) the use of rice as a source of energy seemed to reduce most environmental impact categories explored, with the exception of particulate matter, when compared to the conventional electricity grid as well as to the use of petrol in vehicles. These findings suggest co-benefits in reduced environmental impacts may be achievable through influencing two markets: energy and cement.

In a study by Gursel *et al.* (2016), a robust assessment of the environmental GHG emissions and criteria air pollutants associated with preparing (i.e., grinding) RHA and transporting it was incorporated into an assessment of “green” concrete alternatives. The authors found that the energy for processing needed for RHA was approximately 5% the energy needed for the production of portland cement and as a result, the replacement of portland cement with RHA resulted in reductions in the impact categories investigated. The authors’ findings did show that the grinding required for the production of reactive RHA led to it having higher energy demands, and associated environmental impacts, than fly ash or limestone. Although, for all mineral admixtures assessed, GHG emissions were lower than that of portland cement or transportation. Noting that for both RHA and fly ash, the impacts associated with acquisition, combustion, and additional product of electricity were considered outside the scope of the assessment. Also, consistent with other studies, it was noted that the impacts from transportation of RHA to the location of desired application in some cases could outweigh those from the collection and refinement of RHA (Shafie *et al.*, 2012; Gursel *et al.*, 2016), leading to recommendations to utilize regionally available SCMs (Gursel *et al.*, 2016).

More prevalent in the literature are studies on the environmental impacts associated with rice cultivation and energy recovery from rice biomass. For example, a detailed study by Brodt *et al.* (2014) examined the GHG emissions from the cultivation of rice. Their work showed that methane from the fields could lead to high GHG emissions values depending on the time horizon considered for assessment. Silalertruska *et al.* (2013) studied the GHG emissions associated with cultivation of rice through the use of rice straw to produce syngas that could offset demand for liquefied petroleum gas. Similar to Brodt *et al.* (2014), Silalertruska *et al.* (2013) found that methane from the fields for rice cultivation could lead to significant contributions to the GHG emissions from this energy alternative. However, even with these methane emissions, the authors found that the production of syngas was still favorable relative to use of fossil fuels. Parvez *et al.* (2016) also examined the environmental impacts associated with use of rice biomass in the production of syngas. Through use of a carbon dioxide enhanced gasification process, these authors found that rice hulls could be successfully combusted at 800-900°C to obtain high-energy gas and could act as a contributor to carbon capture and utilization. Despite this work, there is a critical need for research examining the environmental impacts associated with different energy production methods and the reactivity of associated ash produced, while allocating impacts from cultivation. Work in this area would facilitate understanding of the value added by producing a reactive cementitious material replacement and energy production. Further, such assessment would allow for evaluating the options of producing either higher reactivity ash or higher energy output if optimal production methods cannot be determined for both concurrently.

While the limited environmental impact literature on the use of rice ashes in concrete focus on RHA, due to identical crop cultivation, the only differences in the use of RSA would arise from differences in combustion processes (if deemed necessary vary from RHA).

8. Market potential for rice ash in concrete and cement-based materials

There are many factors that would influence the market value of RHA for concrete. If it provides similar properties to conventional concrete through partial replacement, one could argue the market should be able to bear a similar or slightly higher cost per ton to cement, as has been the case in California with fly ash and slag, which provide greenhouse gas reductions that provide an economic value. RHA is not necessarily cheaper than fly ash counterparts (Arel and Aydin 2018). However, due to RHA's pozzolanic properties, there is market potential. As the literature suggests, the RHA formed with controlled combustion and proper processing can provide properties similar to silica fume, facilitating the production of higher performance concretes. In this case, an increased price could conceivably be warranted for the RHA.

Further, the cost of carbon could act as an incentive to drive use of RHA as a partial replacement for cement. The global cost of carbon is on average $7 / metric ton (IEA 2015a). Global emissions caps required to limit global temperature rise to 2°C is 33-73g CO_2 / $ gross domestic product (GDP) (Rozenberg *et al.*, 2015). In 2014, the United States CO2-eq emissions were 6,870 million metric tons CO_2-eq (USEPA 2017) with a country GDP of $1.7312 E+13 (BEA 2018). In California, the 2015 CO_2-eq emissions were 440.4 million metric tons CO_2-eq (CARB 2017) and in 2017, the State's GDP was $2.5102E+12 (BEA 2018). Given these values, the US is at approximately 397 g CO_2 / $GDP and California is at approximately 175 g CO_2 / $GDP. The approximate emissions to cost ratio of cement production in 2012 (based on US data) was 8.95 kg CO_2 / $ cost (Miller *et al.*, 2016), making it a target for policy-driven mitigation strategies that improve this ratio and drive down the emissions per GDP for the state. The application of alternative materials that can reduce the dependency on cement are a prime area for this reduction.

9. Areas for Future Exploration

The literature indicates that RHA could be a valuable addition to concrete mixtures, especially for use in high strength concrete and repair mortars, and in regions with limited access to high cost materials, such as silica fume (Antiohos *et al.*, 2014). While a great deal of literature exists on the use of RHA as an SCM, there has been comparatively very little work done on the assessment of RSA in concrete. Further, there are several areas of analysis for the incorporation of these ashes that would benefit from further exploration; the key areas for further research and development are highlighted in Figure 9.

In order for RHA and RSA to become competitive in the market, there are several assessments of the properties of these materials in concrete that need to be carried out. In order to consider both as alternatives to current materials, more research into the changes in mechanical and durability properties associated with the use of RSA must be conducted. These studies should incorporate consideration of the roles pre-combustion treatments, post-combustion treatments, and combustion methods have on the amorphous silica content, alkali-metal content, and surface area of the RSA, as these parameters have been shown to affect the reactivity of RHA. Further assessment of certain durability-related properties (e.g., carbonation, coefficient of thermal expansion, creep, abrasion) of concrete containing RHA or RSA should be conducted to clarify the parameters that influence the effects of these ashes on concrete durability. Also needed is a robust assessment of constructability of concretes using RHA or RSA, beyond the laboratory assessments of workability of RHA in concrete.

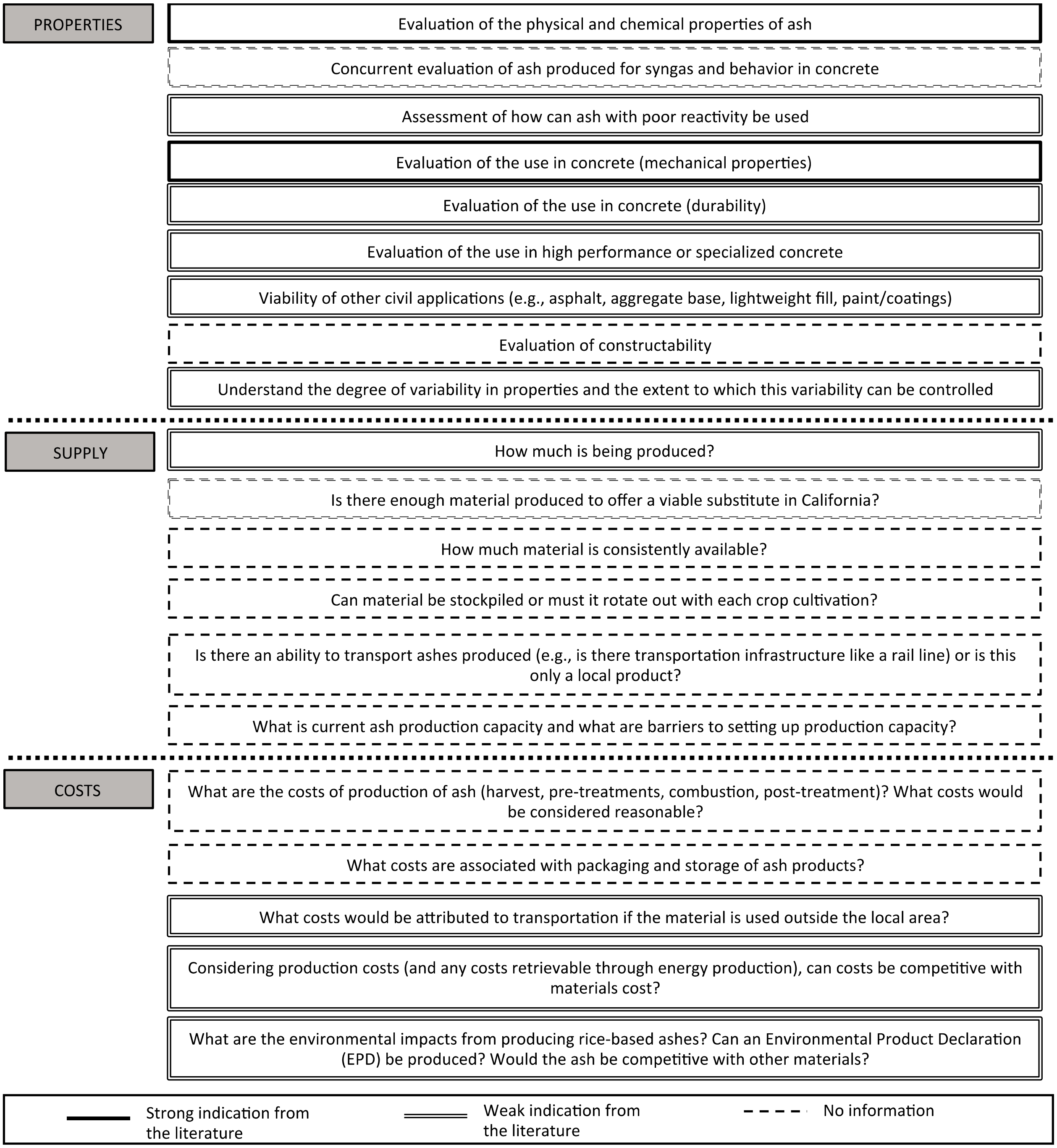


Figure 9. Roadmap for future work in the assessment of rice hull ash (RHA) for use in cement and concrete broken down into areas for further study in material properties, supply, and costs

One of the largest potential hurdles in implementing rice-based ashes as SCMs in concrete is acceptance from the cement and concrete industries. These industries are often hesitant to adopt new materials if there is a perceived limited understanding of the performance of said material, particularly with respect to fresh properties and durability properties. Further, due to the high levels of material consumption for these industries, having sufficient quantities of consistently available, high quality ash with little variation in properties would be needed. To lower uncertainties in costs, identification of codes and measures implemented in other regions that use rice-based ashes more readily in civil engineering materials can be of benefit; for example, they are used as a mineral admixture in parts of Asia (Ferraro *et al.*, 2017).

There are many factors that would influence the market value of rice-based ashes for concrete. If they provide similar properties to conventional concrete through partial replacement of cement with lower environmental impacts, one could expect the market to be able to bear a similar price to other SCMs. In this consideration, more robust environmental impact assessments, including factors such as co-products, allocation methods, and assessment of any alternative processes in ash production must be quantified. Allowable thresholds in behavior, supply, and cost for the rice-based ashes to be competitive must be assessed. Similarly, any uncertainties associated with these phases must be examined. These avenues of research would lead to a better understanding of what benefits are possible in the concrete materials sectors.

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Appendix A:

Summary of individual paper findings on fresh concrete properties containing RHA

Source (Author, date)	SCM Type	Water requirements	Workability	Air content	Heat of hydration	Setting time
Mehta (1995)	RHA	-	-	reduced	-	-
Harish (2010)	RHA (unground)	increased	reduced	-	-	increased
Harish (2010)	RHA (ground)	increased	varies	-	-	increased
Chao-Lung (2011)	RHA (unground)	increased	-	-	-	-
Chao-Lung (2011)	RHA (ground)	increased	-	-	-	-
Salas (2009)	RHA (chemically pretreated)	increased	reduced	reduced	-	-
Salas (2009)	RHA (untreated)	increased	reduced	reduced	-	-
Mehta and Pirtz (1978)	RHA	-	-	-	reduced	-
Antihos (2014)	RHA	increased	-	-	-	-
Venkatanarayanan (2015)	RHA	increased	reduced	-	-	increased

Data from: (Mehta and Pirtz 1978; Mehta and Folliard 1995; Salas *et al.*, 2009; Harish *et al.*, 2010; Chao-Lung *et al.*, 2011; Antiohos *et al.*, 2014; Venkatanarayanan and Rangaraju 2015; Sandhu and Siddique 2017)

Appendix B:

Summary of individual paper findings on hardened concrete properties containing RHA

Source (Author, date)	SCM Type	Early Strength	Long-term strength	Permeability	Chloride ingress	ASR	Sulfate resistance	Freezing and thawing resistance	Drying shrinkage
Alex 2016	RHA (uncontrolled)	effect varies	effect varies	-	-	-	-	-	-
Mehta 1995	RHA	-	increased	-	reduced	reduced	increased	increased	-
Harish (2010)	RHA (unground)	-	increased	-	reduced	-	-	-	-
Harish (2010)	RHA (ground)	-	increased	-	-	-	-	-	-
Chao-Lung (2011)	RHA (ground)	reduced	no significant change	-	-	-	-	-	-
Salas (2009)	RHA (chemically pretreated)	increased	increased	reduced	reduced	-	-	-	-
Salas (2009)	RHA (untreated)	no significant change	no significant change	reduced	reduced	-	-	-	-
He (2017)	RHA	-	increased	-	-	-	-	-	reduced
de Sensale (2010)	RHA (amorphous)	-	-	reduced	reduced	reduced	-	-	-
de Sensale (2010)	RHA (some crystallinity)	-	-	reduced	effect varies	reduced	-	-	-
Sugita (1999)	RHA	-	increased	reduced	reduced	-	-	-	-
Mehta and Pirtz (1978)	RHA	increased	increased	-	-	-	-	-	-
Antihos (2014)	RHA	increased	increased	-	reduced	-	-	-	-
Bie (2015)	RHA (700C)	-	effect varies	-	-	-	-	-	-
Bie (2015)	RHA (600C)	-	increased	-	-	-	-	-	-
Gastaldini (2014)	RHA	-	increased	-	reduced	-	-	-	reduced
Venkatanarayanan (2015)	RHA	increased	increased	-	reduced	-	-	-	-
Zerbino (2012)	RHA	-	reduced	-	-	increased	-	-	-
Rego (2015)	RHA (controlled)	increased	increased	-	-	-	-	-	-
Rego (2015)	RHA (uncontrolled)	reduced	increased	-	-	-	-	-	-
Antiohos (2013)	RHA	reduced	reduced	reduced	similar or increased	-	-	-	-

Data from: (Mehta and Pirtz 1978; Mehta and Folliard 1995; Sugita *et al.*, 1999; Mahmud *et al.*, 2005; de Sensale 2006; Habeeb and Fayyadh 2009; Salas *et al.*, 2009; de Sensale 2010; Harish *et al.*, 2010; Chao-Lung *et al.*, 2011; Zerbino *et al.*, 2012; Antiohos *et al.*, 2013; Antiohos *et al.*, 2014; Gastaldini *et al.*, 2014; Bie *et al.*, 2015; Rêgo *et al.*, 2015; Venkatanarayanan and Rangaraju 2015; Alex *et al.*, 2016; He *et al.*, 2017)