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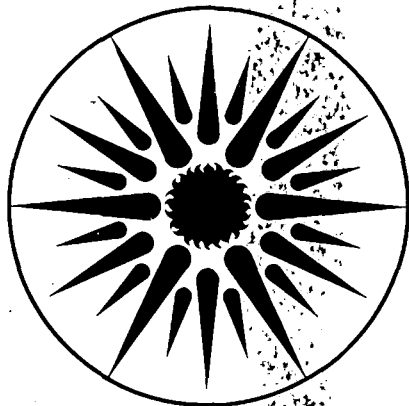
## APPLIED SCIENCE DIVISION

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# **NEAR- AND SUPER-CRITICAL DROPLET COMBUSTION**

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### **Abstract**

A review is presented of theoretical and experimental studies carried out over the last 25 years on the problem of droplet heating, vaporization, and combustion at near- and super-critical conditions. Following a detailed description of the classical quasi-steady theory, particular attention is given to major departures from it, that must be considered in a super-critical droplet combustion model. This includes unsteady heating and gas-phase diffusion, as well as the influence of ambient conditions, fuel-vapor accumulations, convective heat transfer and gas solubility.

## INTRODUCTION

Direct fuel injection into a combustion chamber is today a well accepted engineering practice, covering a wide range of applications such as industrial heaters, diesel engines, and gas turbines. In this type of a system the process of combustion is carried out by a diffusion flame. The characteristic feature of such a flame is that it is established at the stoichiometric contour where all the oxygen is consumed and the temperature reaches the maximum that the fuel is capable of producing. In approaching it in the absence of oxygen, the fuel is pyrolyzed forming particulates, whereas the high temperature at the flame front enhances the formation of nitric oxide and its relatively narrow width inhibits the attainment of a sufficiently long residence time of the substance in the zone of reaction to assure its completion. In essence then one has thus a process that automatically optimizes the formation of all the pollutants, including smoke.

This situation could be greatly improved if the fuel were mixed with air before it is ignited. Preheating it to near- or super-critical temperature may be quite beneficial in this respect. In principle, by cutting out the time for evaporation and the effect of surface tension, the high velocity "droplets" could be mixed with air into which they are injected sufficiently fast to overtake the chemical induction time for ignition.

The purpose of this report is to examine the scientific background available at this time, laying thus proper foundation for our studies of the means whereby a controlled combustion process could be actually

attained in a diesel engine. The advent of adiabatic engines is, of course, conducive towards this aim, although it is not absolutely necessary. Heating hydrocarbon fuels to supercritical temperatures of about 500 °C and pressures of about 20 atmospheres is technologically quite feasible in any diesel engine.

Provided here, accordingly, is an assessment of the theoretical and experimental work carried out over the last quarter of a century upon the problem of droplet heating, vaporization, and combustion at near- and super-critical conditions. Following the description of the classical quasi-steady theory, the major modifications one has to introduce in order to treat super-critical droplet combustion are presented. This includes unsteady droplet heating, gas-phase diffusion, influence of ambient conditions, fuel vapor accumulation, convective heat transfer and gas solubility.

## THE QUASI-STEADY THEORY

The classical theory of droplet vaporization and combustion considers a single-component, spherical fuel droplet in an infinitely large oxidizing gas which is otherwise undisturbed. By undisturbed it is meant that the large gas reservoir is at rest except for the radial motion induced by the mass transfer outward from the droplet. In the absence of relative velocity between the gas and the droplet, and in the absence of buoyancy forces, the flow field is spherically symmetric, which means that all variables of the problem are only functions of one spacial coordinate, the radial coordinate, and perhaps, functions of time. Because of the low-Mach-number flows normally encountered, the process is considered to be isobaric. The droplet temperature is assumed to be constant and uniform, meaning that there is no droplet heating and that liquid-phase heat transport processes are neglected. As the droplet vaporizes in the hot oxidizing flow, its mass and size are reduced at a rate dictated by the influx of heat at the droplet surface. Because of the large density difference between liquid and gas, the quasi-steady theory states that the rate of change of droplet radius is so slow that time-derivatives can be dropped out of the governing equations. Diffusion of heat in the gas phase is assumed to be dictated by Fourier's law while Fick's law of diffusion governs the molecular transport of species. Second-order diffusion, as represented by the Soret and Dufour effects, is neglected. In general, constant gas-phase transport properties are assumed. A single binary



diffusion coefficient is used for all pairs of species. The specific heat and thermal conductivity are assumed to be constants, while the Lewis number is assumed to be unity. These constraints, however, are relaxed in some revised droplet models. Since chemical reaction rates are much faster than gas-phase diffusion rates, the flame-sheet combustion model is employed: the flame has an infinitesimal thickness and is treated as a sink for the reactants and a source of heat and products.

The quasi-steady theory also assumes that all the fuel gasified at the droplet surface is instantaneously consumed at the flame, therefore neglecting any fuel accumulation and depletion between the surface and the flame. The main result of the quasi-steady theory is the so-called  $d^2$ -Law, which states that the square of the droplet diameter decreases linearly with time, or equivalently, that the rate of change of the square of the droplet diameter is a constant, frequently called the evaporation or burning rate constant. Because of its simplicity, the  $d^2$ -Law has been extensively used in modelling droplet and spray combustion. In particular, the qualitative behavior of this law has been found to be largely correct, while the burning rate constant can generally be predicted to within a factor of two. Discrepancies do exist, particularly those related to the initial conditions when some of the heat must be used to internally heat the droplet and fuel accumulation occurs near the surface. In addition to the  $d^2$ -result, the quasi-steady theory also yields estimates for the flame temperature and the flame stand-off distance. The prediction of the flame temperature is generally somewhat higher than the experimentally observed values. Similarly, the predicted flame

stand-off distance typically exceeds the observed values by a factor of about three. Moreover, the flame standoff ratio is not constant but varies with time, mainly as the result of fuel accumulation and depletion.

In the following sections, we present a discussion of important deviations from the quasi-steady theory that must be incorporated in a study of near-critical and supercritical droplet combustion.

### **DROPLET HEATING**

In most practical applications, the droplets are initially at a temperature substantially below their boiling temperature. In some cases, the droplet temperature will also be relatively low when ignition occurs. Before the droplet reaches a state of temperature equilibrium, it must undergo a heating process during which the droplet temperature varies with both time and location inside the droplet (Law, 1982). In the absence of any external convective fields, there will be no internal circulation and the droplet temperature will be a function of the radial coordinate and time. During the initial heating period, the heat arriving at the droplet surface is used in part to gasify some liquid at the droplet surface, while the rest of the heat flux is conducted into the droplet interior for droplet heating. This heating period is followed by a process of vigorous gasification with almost no droplet heating and nearly constant droplet surface temperature. Since the droplet has to be moderately hot before ignition is possible, droplet heating after ignition should be quite limited.

As a first approach to the transient heating problem, it is assumed that the droplet temperature is spatially uniform but time-varying. This is equivalent to stating that the thermal conductivity of the liquid is infinitely large or that very rapid mixing occurs inside the droplet so that the temperature is instantaneously uniformized. This model is referred to as the rapid-mixing or distillation limit. It has been recognized, however, that it is more reasonable to consider a finite thermal conductivity in the liquid, with the droplet temperature varying with both space and time. This is the so-called diffusion limit and it presents further complications because of the continuously regressing surface and the continuously changing surface temperature. The distillation limit is obviously an artificial one in the sense that in order to increase the heating rate beyond the one established by the diffusion limit, some sort of internal mixing must occur which is incompatible with the assumption of spherical symmetry. The infinite diffusivity model underpredicts the surface temperature and the vaporization rate during the initial period and overpredicts them later in the lifetime. It should be noted that the bulk gas-phase combustion characteristics, such as the instantaneous burning rate, are insensitive to the detailed heat transfer processes within the droplet (Law, 1982). On the other hand, the droplet temperature distribution may remain non-uniform and temporarily varying throughout the lifetime. Consequently any combustion characteristics depending on this temperature should be evaluated using the more complete diffusion-limit model. Finally, it should also be noted that at near-critical and supercritical pressures, droplet heating can comprise a

significant portion of the droplet lifetime. Wieber (1963) calculated the temperature histories of vaporizing droplets at pressures to 2000 psia. Liquid heptane and liquid oxygen droplets were studied using the distillation limit of uniform droplet temperature and assuming that transport processes in the gas phase were quasi-steady. Some vaporization histories showed that the droplet temperature did not attain equilibrium at high pressure but instead continued rising to the critical temperature isotherm. For example, a 100- $\mu$ -radius heptane droplet vaporizing in a combustor whose chamber pressure is 1000 psia, shows a behavior characteristic of low pressure cases up to about 1 msec after injection. When the critical point is reached at about 1.2 msec after injection, the fraction of the initial drop weight still remaining is about 0.8. The droplet radius increases rapidly near the critical point as a result of the rapidly decreasing liquid density and despite the high vaporization rate. The history calculations of Wieber indicate that droplets evaporating at high chamber pressures will reach the critical point before an appreciable amount of mass is evaporated. Once the liquid reaches its critical point, the liquid interface vanishes and diffusion of the fuel vapor into the gaseous environment becomes the rate controlling step of the combustion process.

Experimental temperature measurements of suspended droplets ranging from 1 to 2 mm in diameter, in near-critical and slightly supercritical gaseous environments were performed by Kadota and Hiroyasu (1981) using a 25.4- $\mu$ m-diameter Pt-PtRh thermocouple. At subcritical pressures, following an initial rise after ignition, the droplet

temperature almost leveled off (for an n-heptane droplet at a reduced pressure of 0.22, the temperature leveled off at 150K after 0.5 sec). As the pressure increased, the rate of temperature rise of the liquid droplet increased and the leveling-off tendency became less evident. At reduced pressures as high as 1.5, there was only an inflection of the temperature trace. Similar behavior of the temperature histories of n-decane droplets with initial diameters in the range 600-1000 $\mu$  burning in air was observed by Faeth et al (1969). At pressures below the critical pressure of the droplet, the measured wet bulb temperature at which the droplet temperature would level out, was close to the boiling temperature. At reduced pressures above 2.5, there was little evidence of a significant amount of evaporation prior to the time when the temperature exceeded the critical temperature of the fuel. These experiments were conducted in a free-falling chamber under zero-gravity conditions, and the results confirmed the theoretically predicted supercritical burning time proportional to the cubic root of the pressure.

### **THE UNSTEADY GAS PHASE**

The validity and utility of the quasi-steady gas-phase formulas are doubtful at the higher pressures encountered in many combustion systems. Two principal reasons for this failure are: first, as the pressure approaches the critical pressure of the injected fuel, the latent heat of vaporization decreases to zero and the quasi-steady formulas predict a vaporization rate which increases to infinity; and second, at

near-critical and supercritical pressures and temperatures, the characteristic times for transient behaviors in the gas-phase are comparable to the droplet lifetime. The upper limit of pressure at which the quasi-steady is valid may be arbitrarily defined as one-tenth of the fuel critical pressure. At this pressure, the density of saturated vapor is of the order of one percent of the density of saturated liquid (Brzustowski, 1965). The rate-limiting process at pressures above the arbitrarily defined limit (but still below the fuel critical pressure) is unsteady heat and mass transfer in the gas film surrounding the decreasing droplet. When a liquid fuel is injected into an oxidizing gas, and the pressure of the gas exceeds the critical pressure of the fuel, transient heat transfer and diffusion are the significant physical phenomena. Following Brzustowski, consider the events after the injection of a compressed liquid fuel droplet into its gas. Let the liquid be at a temperature  $T_1 < T_{crit}$  and the surrounding fuel gas at  $T_2 > T_{crit}$ . This system liquid-gas is not at equilibrium but the states 1 of the liquid and 2 of the gas can be shown on the P-v diagram of this one-component system (See Fig.1, taken from Brzustowski (1965)). The droplet is heated at constant pressure and a transient temperature wave moves into the droplet. The point on the temperature wave where  $T = T_{crit}$  indicates the instantaneous phase boundary between compressed liquid and dense gas. When the point  $T = T_{crit}$  reaches the center of what was the droplet, all of the compressed liquid has become dense gas and the subsequent heating merely increases its temperature to  $T_2$ . Finally, the equilibrium state 2

is reached. As a first estimate of the characteristic times involved in the above mechanism, the process can be divided in two parts: First, the transient heating of the droplet and the accompanying phase change, and second, the subsequent diffusion of the resulting pocket of dense fuel gas. Using well known heat transfer results, Brzustowski estimates the heating time (defined as the time up to the moment when the surface reaches the critical temperature) for a 200- $\mu$ -diameter droplet with  $T_{crit} = 600K$  injected at 400K into a gas at 2300K to be approximately 1.5  $\mu$ sec if evaporation is entirely neglected during the heating period. The phase change is completed after 7.5 msec but this is an overprediction due to the fact that the droplet regression is neglected in the calculation. Finally, the time necessary for the gas pocket to diffuse away and react is estimated to be as long as 0.1 sec.

Spalding (1959) and Rosner (1967) presented two simplified theories of unsteady droplet combustion at supercritical pressures. The first author used an already existing heat transfer result in his treatment of the problem as a diffusing point source. The mathematical model yields closed-form solutions for the flame radius and combustion times in the case of a spherically symmetric process. An illuminating result is the dependence of the supercritical burning time on the cubic root of the pressure, while the quasi-steady theory, valid at low pressures, predicts a burning time independent of pressure. Spalding's solution, however, is valid only at values of time larger than  $r_0^2/D$ , where  $r_0$  is the initial pocket radius and  $D$  is the diffusion coefficient. Rosner (1967) goes one

step further by representing the initial gas pocket as a distributed source. His results are qualitatively similar to those of Spalding. Sanchez-Tarifa et al (1972) were able to obtain closed-form expressions for the burning rates, combustion times, flame radius, and temperature of the droplet center, using an asymptotic analysis valid when the initial droplet temperature is small as compared with the temperature of the surrounding atmosphere ( $T_0/T_\infty \ll 1$ ). In the vaporizing-droplet case, it is found that an apparent droplet exists throughout most of the process. This droplet practically keeps its initial temperature and composition and its surface is characterized by an abrupt change in the temperature and mass fraction profiles. This result is physically explained by the strong dependency of the transport coefficients on temperature. Analytical results were obtained for the combustion case. The droplet lifetime which is independent of pressure at sufficiently low subcritical conditions, decreases markedly with pressure as the critical state is approached, reaches a minimum at critical conditions, and then increases steadily in proportion to the cubic root of the pressure at supercritical conditions. The abrupt decrease of the droplet lifetime as the critical state is approached is explained by the continuous reduction of the latent heat of vaporization of the fuel. At critical conditions the droplet interface vanishes and the heat of vaporization is zero. The droplet then becomes a gas pocket and gasification is no longer the rate controlling step. Instead, gas-phase diffusion becomes the rate controlling process, and the droplet lifetime decreases with increasing pressure due to the lower diffusion rates at



higher pressures. Fig.2, taken from Sanchez-Tarifa et al (1972), indicates the described trends for an n-decane droplet of constant initial mass equivalent to a liquid droplet of 875  $\mu$  in diameter.

### **INFLUENCE OF AMBIENT GAS COMPOSITION**

The experimental work of Faeth et al (1969) shows the influence of different ambient oxygen concentrations on the droplet temperature history in the region of supercritical combustion. The liquid temperature always increases more rapidly at higher oxygen concentrations for a given droplet size. This is because at higher oxygen concentrations the combustion zone is closer to the droplet and therefore, there are larger temperature gradients in the gas phase and more heat transfer to the droplet.

### **FUEL-VAPOR ACCUMULATIONS**

Droplet heating and unsteady gas-phase diffusion are not the only transient processes occurring in droplet combustion. Fuel vapor accumulation and depletion plays a significant role as will be appreciated in the following discussion. At the moment of ignition, the droplet is still undergoing transient heating with only a fraction of the incoming heat flux being used to vaporize the liquid. As a consequence, a small amount of vapor fuel is surrounding the droplet at the instant of ignition, and this results in a flame that lies close to the droplet surface (Law, 1982). The

presence of this flame greatly increases the heat flux onto the droplet surface and therefore further increases the droplet surface temperature. With this increased gasification, the fuel vapor present between the surface and the flame increases, and the flame is pushed outwards (the constant flame-to-droplet radial ratio predicted by the quasi-steady theory has been observed to be incorrect even at low ambient pressures). Therefore some of the fuel vaporized during this period is accumulated inside the flame envelope as the flame expands. Then during the latter part of the droplet lifetime this amount may be consumed as the flame collapses toward the droplet. Theretical calculations performed by Law (1982) support the following conclusions: The flame stand of ratio increases linearly after ignition and, in a rich oxidizing environment, approaches a constant value different from the quasi-steady result. After ignition the fuel consumption rate is less than the gasification rate and in the latter part of the life time the fuel consumption rate is greater than the gasification rate, implying that the initially accumulated fuel is subsequently depleted.

## **THE EFFECTS OF A CONVECTIVE ENVIRONMENT**

The influence of natural convection was qualitatively observed by Faeth et al (1969), and it was seen to cause substantial reductions in combustion times over the convection-free results. The presence of free convection in droplet combustion destroys the spherical symmetry of combustion and distorts the flame from its spherical shape. At subcritical

conditions, natural convection effects on the burning constant amount to less than 10% for droplets of diameter of 1mm or less. The experimental results of Kadota and Hiroyasu (1981) with suspended droplets throw some light into the effects of free convection on the burning characteristics of droplets. These authors found that at very high subcritical and near critical pressures ( $0.3 < P_r < 1$ ), increasing pressure causes the combustion lifetime to decrease abruptly. This observation agrees well with the theoretical expectation, as the heat required to vaporize the fuel reduces to zero. In supercritical conditions, however, the combustion lifetime of the buoyancy-affected droplet decreases gradually with increasing pressure as opposed to the zero-gravity expectation that the combustion lifetime increases in proportion to the cubic root of the pressure. While in zero-gravity conditions, an increase in pressure produces a decrease in the gas-phase diffusion rates and consequently an increase in the lifetime, the presence of natural convection results in a buoyancy-induced enhancement of gas-phase motion and mixing and a reduction of the lifetime.

Forced convection is the result of relative motion between the droplet and the surrounding oxidizing gas. It is apparent that in any real situation the droplets are injected in the combustion chamber with some initial velocity and consequently the effects of a forced-convective flow must be carefully considered in a theoretical analysis. Among these effects, the enhancement of vaporization and diffusional rates, brought about by the presence of a boundary layer surrounding the droplet, are of primary interest. One way of dealing with this phenomenon is through the use of

empirically obtained corrective factors to be used in the prediction of gasification rates. Such factors generally incorporate a Reynolds number that contains the effect of the convective flow, and one or more dimensionless parameters such as the Prandtl number, to account for the effect of physical properties. The main disadvantage of using empirical corrective factors is that their application should be limited to situations that resemble as much as possible the experimental conditions under which the factors were obtained. In the search of general mathematical models, it is convenient to consider the solution of the governing equations including the effects of the convective conditions, i.e.: buoyancy terms, relative velocities, shear stresses, etc. It should be expected that the complexity of such models requires the use of numerical techniques for their solution.

## **GAS SOLUBILITY IN THE LIQUID PHASE**

According to the experimental observations and theoretical predictions of Lazar and Faeth (1971), the solubility of the ambient gas in the fuel becomes quite large at high pressures. For pressures greater than the critical pressure of the pure fuel, the concentration of nitrogen in the liquid phase increases at higher liquid temperatures for a fixed total pressure. Thus, as the droplet temperature increases, gas goes into solution causing additional complications in the analysis of droplet heat-up at high pressures. The authors suggest that corrections should be

made in the use of Rosner's supercritical combustion model (Rosner, 1967) to include solubility effects. Rosner and Chang (1973) did not deal with the most general case in which gas dissolution in the droplet proceeds simultaneously with droplet evaporation, but did consider the important limiting case of vaporization of a droplet which is already gas-saturated at  $t=0$ . Their model considers a droplet evaporating into a binary mixture of two gases **A** and **B**, the latter being soluble in liquid **A**. An approximate expression is obtained for the ratio of the droplet lifetimes including and neglecting solubility effects but no quantitative examination of this expression is presented.

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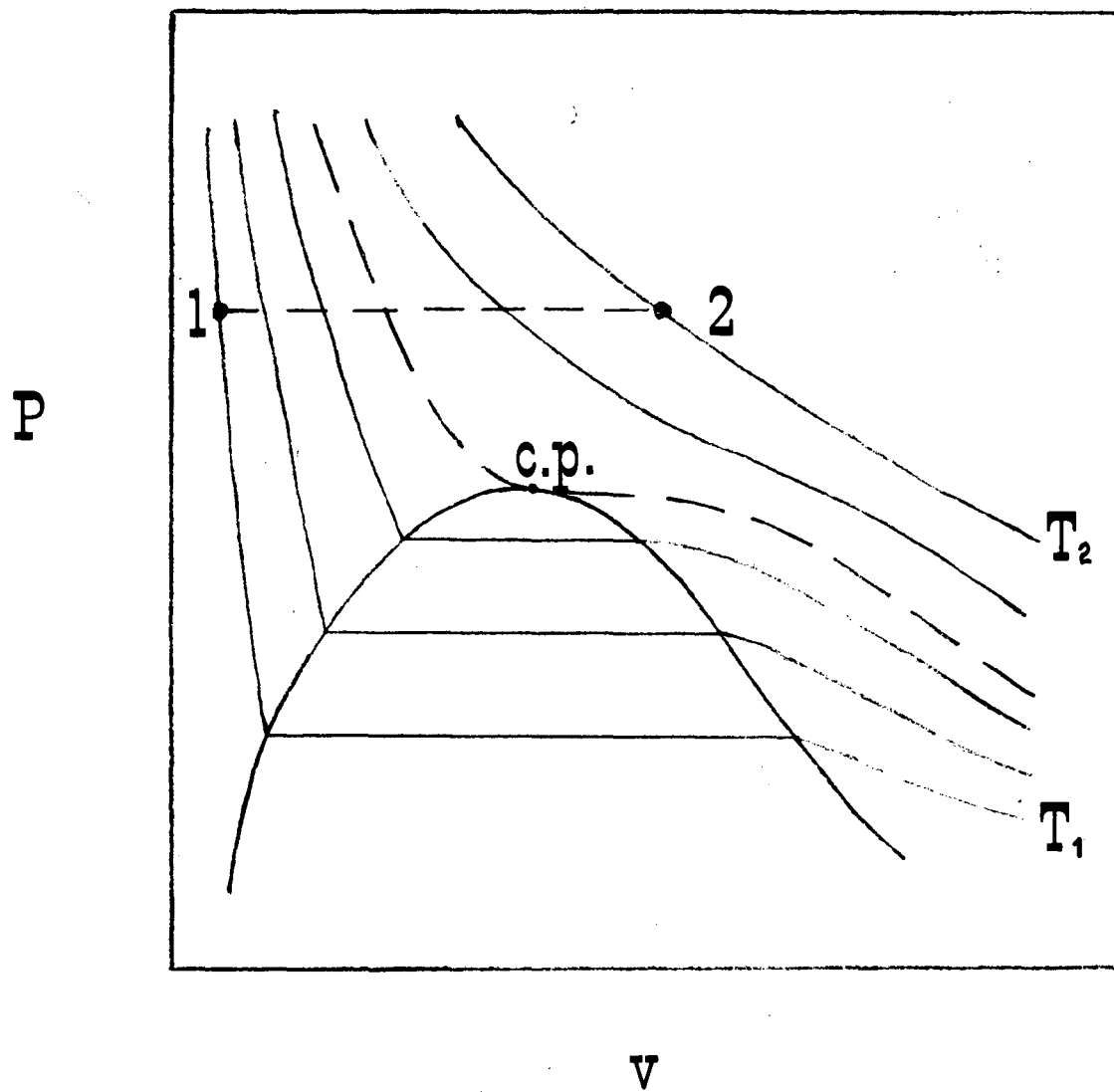


Fig. 1



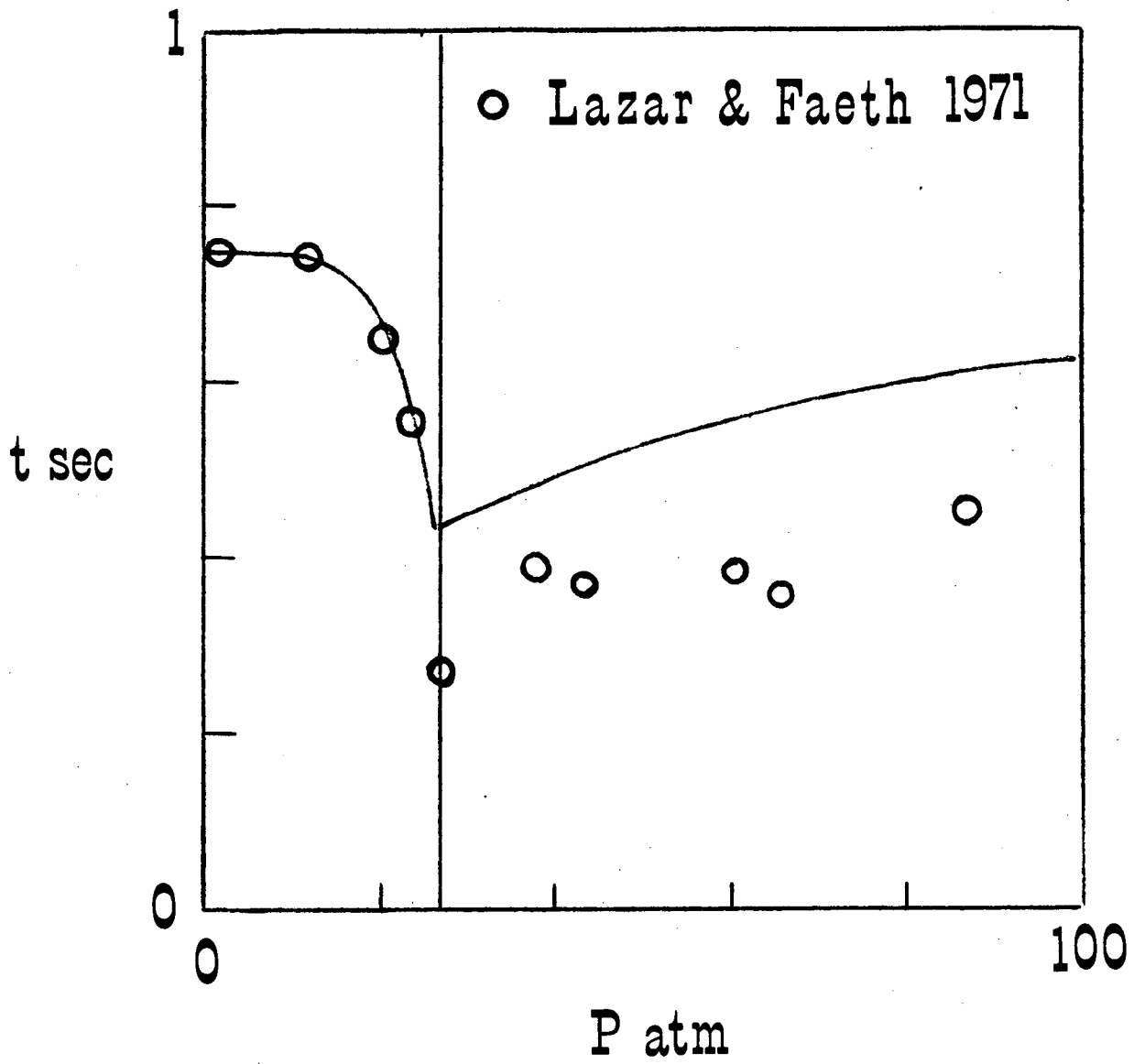


Fig. 2

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