VAPORIZATION CHARACTERISTICS OF ETHANOL DROPLETS: INFLUENCE OF THE ENVIRONMENT HUMIDITY

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ABSTRACT

The study of the gasification of a droplet via vaporization, which involves heat, mass and momentum transfer processes in gas and liquid phases, and their coupling at the droplet interface, is necessary for better understanding and modeling of complex spray and mixture formation issues.

A detailed description of the vaporization of an isolated droplet has been realized in this experimental study aimed at investigating the impact of the water vapor contained in the surrounding gas on the evaporation of an ethanol droplet. The experimental set-up consists of a heated chamber with a cross quartz fibers configuration as droplet support. An ethanol droplet is located at the intersection of the cross with a controlled initial diameter (300 - 550µm). Ambient temperature is varied from 350 to 850 K.

The real impact of the water concentration on the vaporization rate of an ethanol droplet in a large range of temperature is examined, showing that the vaporization of an ethanol droplet is accompanied by the simultaneous condensation of water vapour on the droplet surface and thus the temporal evolution of the droplet squared diameter exhibits an unsteady behaviour. The histories of the instantaneous vaporisation rates calculated from the $d^2(t)$ curves confirms this non-stationary aspect of the phenomenon.

NOMENCLATURE

K: vaporization rate

Kinst: instantaneous vaporization rate

- K_i: initial vaporization rate
- K_f: final vaporization rate
- d₀: initial droplet diameter
- d: droplet diameter

t_{vap}: total vaporization time

T: temperature

T_s: droplet surface temperature

INTRODUCTION

Recent growing concern on depletion of conventional petroleum based fuels particularly in global automotive industry has raised significant interest in bio fuels research. Bio fuels such as pure vegetable oils, biodiesel based on cross transesterification of vegetable oils or animal fats and ethanol based on sugar or starch crops are currently readily available. These bio fuels issued from the biomass which are known to be renewable, biodegradable, oxygenated and produce less pollutants [1] when compared with conventional fuels require further and more extensive investigations on these fields. This paper only focuses on an alcohol, ethanol which is already known as to hold potential as viable alternate fuel. As pointed by Agarwal [1] and Jeuland et al. [2], ethanol poses some important assets as a feasible alternate fuel. As mentioned, ethanol has a very high octane number, similar density to gasoline. The existence of oxygen in the formula of ethanol will provide a more homogeneous fuel/air mixing and will therefore produce fewer pollutants, such as unburned hydrocarbons and carbon monoxide. Ethanol also has a very high latent heat of vaporization which enables a 'cooling effect' of air. Ethanol is largely used in SI engines as pure fuel or flex fuel until 85 % in gasoline in Europe. With this high percent, ethanol vaporization which is a fundamental process in combustion phenomenon must be correctly characterized. When compared to gasoline, with a higher latent heat of vaporization, ethanol can involve problems of start in cold conditions. With a lower heat of combustion, it is necessary to

ignite in rich mixture, that is unfavourable for pollutant emissions. Many studies were carried out on ethanol only concentrating on the pollutant emissions and engine performance [1, 3-11]. Only few studies on fundamental issues were carried out on gasification of ethanol droplet and its vaporization behaviour [12-19].

Droplet vaporization behaviour is one of the main characteristics that have been continuously studied for years especially on n-alkanes and vegetable oil methyl esters [16, 20-23] due to its importance in processes as combustion systems in diesel and propulsion engines. In these combustion systems, the liquid fuel is always atomized as a cloud of droplets in the chamber, which then vaporize, and mix with the oxidant and burn to release heat. Therefore, to provide a better knowledge in modelling complex spray flows, the study of the droplet vaporization which involves mass, heat and momentum transfer processes is really needed.

Earlier studies around 1950s by Godsave [24] and Spalding [25] on the droplet vaporisation have introduced the development of the quasi-steady theory also which leads to the d^2 -law. This model is based on several assumptions. The assumptions include gas and liquid phases' steady behaviour, constant thermodynamic and transport diffusion properties. This theory leads to the prediction of linear evolution of the droplet surface area with time and therefore permits an estimation of the average vaporisation rate, K as:

$$d^2 = -Kt + d_0^2$$
 (1)

Ethanol has a relatively volatile and very miscible behaviour with water. Therefore it is important to investigate the impact of different initial water concentration and environment humidity on the vaporization rate of ethanol. Previous works such as Law et al. [14] who have investigated experimentally and theoretically on the humidity of air effect on suspended methanol droplet and also as Mukhopadhyay et al. [26] on the simulation of pure vaporization of methanol droplet, have found a significant deviation from the d²-law behaviour. The complete droplet gasification lifetime is also further prolonged in cases of humid environment on methanol vaporization. Their agreement is based on the argument that the existence of water concentration from the droplet and humid environment sensibly interfere with the subsequent vaporization of methanol droplet. Therefore the K, average vaporisation rate ceases to be a constant during all the droplet lifetime.

In this work, experimental results concerning vaporization of ethanol in nitrogen at atmospheric pressure are presented. The average and instantaneous vaporization rates have been determined for two types of ethanol forms: ethanol (95% vol. ethanol + 5% vol. water) and anhydrous ethanol for the temperature range from 350 to 850K. The impact on the droplet vaporization rate of both initial water concentration and ambient moisture is discussed here. A new and wide range of temperatures is covered in this paper.

EXPERIMENTAL

The experimental set-up is already described elsewhere in Ref. [27] and schematically represented in Fig. 1. The experiments were performed by using the 'cross fibre' technique. With this technique, the droplet shape can be reasonably assumed as spherical (cf. Figure 2). The droplet is created at the intersection of two 14 µm diameter quartz wires fixed perpendicularly on a frame (therefore the effect of heat transfer from the fibre to the droplet through conduction could be significantly minimized [27]). In this case, a piezo-electric injector generates the droplet, by supplying a monodispersed liquid jet impacting the support. Nitrogen (99.95% purity) fills the medium of the furnace to allow pure vaporization and to avoid any oxidation or ignition to occur.



Figure 1: Schematic representation of the experimental set-up

The droplet injection on the frame is carried out in a region of the vaporization chamber at ambient temperature, called cold zone in order to avoid pre-vaporization before the start of the experiment. Once the droplet of controlled initial diameter (300 - 600μ m) is formed on the intersection of the quartz fibres, it is then introduced into the furnace by a motorized displacement system. The total transfer time is of 700 ms. When the droplet is exposed to the hot environment in the furnace, the temporal regression is recorded using a high-speed video camera with various frame rates from 20 to 400 fps (depending on the temperature). For each experiment set, a minimum of 700 images are recorded to allow sufficient temporal resolution. To ensure and verify the repeatability of the data, at least six experiments are performed for each test condition.



Figure 2: Cross-fiber supported droplet technique

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Two forms of ethanol, anhydrous ethanol with high purity GC grade and standard ethanol, called later "Ethanol 95%" (95% ethanol with 5% water) are used for these experiments. Ethanol (CH₃CH₂OH) is an alcohol; its molecular structure shows a polar fraction due to the hydroxyl radical and a non polar fraction in its carbon chain. Due to its short carbon chain, the properties of ethanol polar fraction overcome the non polar properties. That explains the hygroscopic nature of ethanol.

The physical and chemical properties of ethanol are presented in table 1.

Density, ρ (kg/m3 @ 298K)	790
Dynamic viscosity, µ (mPa.s @ 298K)	1.074
Surface tension, σ (10 ⁻³ N.m @ 293K)	22.75
Latent heat of vaporization,	42.32
$\Delta H (kJ/mol @ 298K)$	
Solubility (g/100ml water)	∞
Boiling temperature for ethanol, T _b (K)	351.32
Molecular weight M (g/mol)	46.07

Table 1: Physical and chemical properties of Ethanol (CH₃CH₂OH)

In all experiments, the pressure in the furnace is kept at atmospheric while the temperature is varied from 350K to 850K. The ambient gas is nitrogen. The homogeneity of the temperature is controlled thanks to three thermocouples placed inside the furnace.

COMPUTATIONS

The images captured by the high speed video camera are transferred to a computer and are analyzed by post-processing to deduce the droplet instantaneous surface area and hence its diameter temporal variation. Note that the calculation error in determining the droplet diameter is of the order of about 3%. A resolution of 10 μ m/pixel is obtained thanks to the macro lens coupled to the high speed video camera.

Finally, the temporal evolution of the squared diameter or the squared diameter normalized by the initial droplet diameter can be plotted for each explored case. The normalized evolutions allow explicit comparison of temporal evolutions independently of the initial diameters.

RESULTS AND DISCUSSION

Average vaporization rate

Average vaporization rate from the experiments is calculated by a linear least-square fit in the quasi steady zone of the d^2 curves (Figure 3). In these experiments, the so-called 'quasi-steady' period for both ethanol (95%) and anhydrous ethanol occurs two times throughout the droplet lifetime. Average vaporization rates are deduced from the d^2 -curves presenting two parts: the first linear part allows to determine a first average vaporization rate called thereafter "initial vaporization rate K_i" and the second linear part a second average vaporization rate called "final vaporization rate K_f". Figure 4 and Figure 5 show the normalized evolution of the squared diameter of the droplet against normalized time. Both Figure 4 and Figure 5 show few common features, the d^2/d_0^2 versus t/d_0^2 curves for all temperatures are deviated from the d^2 -law. The initial average vaporization rate, K_i ceases to be constant at a certain point through the droplet life. For ethanol (95%) the deviation from the linear part occurs at mid stage of the vaporization of the droplet (and $d^2/d_0^2 < 0.4$) whereas for anhydrous ethanol, the deviation only starts towards the end of the droplet life (and $d^2/d_0^2 < 0.2$). When the droplets are formed in a closed chamber filled with nitrogen gas, one prevents the combustion of the droplets and excludes the effect of ambient moisture on the evaporation process. Even so, there is still some humidity in the chamber (leak, wall adsorption...). This could explain why even anhydrous ethanol produces a nonlinear d²-law.



Figure 3: Calculation of initial average vaporization rate, K_i and final average vaporization rate, K_f from the d²(t) curve for ethanol (95%) at T= 473 K and P = 0.1 MPa. K_i and K_f are calculated from the red and blue part respectively



Figure 4: d² (t) curves for ethanol (95%) at different temperatures; P = 0.1 MPa



Figure 5: d² (t) curves for anhydrous ethanol at different temperatures; P = 0.1 MPa

The different values in initial water concentration in both ethanol forms are also affecting the droplet lifetime. With greater initial water concentration, the droplet lifetime is significantly prolonged (+30%) as shown in Figure 6.

As shown in Figure 7, the average vaporization rates, K_i calculated from the first linear part of the d² curves are similar for both ethanol forms. Therefore it shall be noted that the first linear part of d² curves for both ethanol (95%) and anhydrous ethanol may entirely and totally consists only of ethanol vaporization. Ethanol is the major component evaporating at this first stage as it has lower boiling temperature than water.



Figure 6: d^2 (t) curves for ethanol (95%) and anhydrous ethanol at T= 623 K and P = 0.1 MPa



Figure 7: Average vaporization rates for ethanol (95%) and anhydrous ethanol at different temperatures. K_i is calculated from the first linear part of the d²-curves

For the calculation of the final average vaporization rate, K_f , a comparison has been made with an existent fit of pure water vaporization rate [28]. As observed in Figure 8, at lower temperature (T < 600 K), K_f of both ethanol (95%) and anhydrous ethanol behave as the pure water. However, as the temperature increases, the values of K_f start to deviate further from the fit significantly for anhydrous ethanol. It may be due to the fact that at these higher temperatures, one exceeds the boiling temperatures of both ethanol and water. Therefore, the ethanol and water components simultaneously evaporate resulting in higher K_f .



Figure 8: Average vaporization rates calculated during second linear part of the d² curves for ethanol (95%), anhydrous ethanol (this work), with pure water vaporization rate fit at different temperatures [28].

Instantaneous vaporization rates

The instantaneous vaporization rate, K_{inst} , are calculated from the d²-curves by determining the derivative of these curves. According to [14], the derivative of the $d^2(t)$ will give the instantaneous vaporization rate,

$$K_{inst}(t) = -d \left(\frac{d^2(t)}{dt} \right)$$
(2)

In order to avoid errors in the calculation of the derivative a smoothing is carried out on the $d^2(t)$ curves by using a FFT low-pass filter. This method removes only the high frequency components with a parabolic window (Origin[®] function). Then the derivative is calculated on this smoothed curve.

Figure 9 and Figure 10 show the original $d^{2}(t)$ curve and their corresponding instantaneous vaporization rate, Kinst versus time for both ethanol (95%) and anhydrous ethanol. It is clearly observed, that in both cases the instantaneous vaporization rate, K_{inst} is significantly unsteady for the initial part corresponding to the ethanol vaporisation period, K_i. The second period, K_f, attributed to the water vaporization is almost quasi steady. In order to illustrate that, the equivalent average value of Kinst is plotted in Figures 9 & 10, corresponding to the vaporizing rate K_i and K_f respectively. That shows that although it is easy to determine a linear trend on the $d^{2}(t)$ curve, the unsteadiness of the phenomenon is clearly revealed by the evolution of K_{inst} according to time. This evolution is certainly due to the interference of water concentration on the ethanol droplet vaporization and also to the water condensation from the ambient moisture, due to the temperature decrease at droplet surface.



Figure 9: Evolution of the instantaneous vaporization rate, $K_{inst}(t)$ and the squared droplet diameter for ethanol (95%) droplet (d₀ = 413 µm) at T= 473 K.



Figure 10: Evolution of the instantaneous vaporization rate, $K_{inst}(t)$ and the squared droplet diameter for anhydrous ethanol droplet (d₀ = 575 µm) at T= 473 K.

In order to compare these evolutions for different temperatures, a normalization of these curves has been conducted. The time has been normalized by the droplet total vaporization time t_{vap} .

Figure 11 shows the variation of normalized instantaneous vaporization rate, K_{inst} for both ethanol (95%) and anhydrous ethanol against the time normalized by t_{vap} . It is observed that by considering normalized time by t_{vap} , the instantaneous vaporization rate, K_{inst} presents the two domains, previously described, ethanol vaporization first and then the water vaporization. In this figure one can observe that the first part of the vaporization process occurs mainly at 1/3 of the total vaporization time for the ethanol (95%), even though this occurs around at 70% of the total vaporization time for the anhydrous ethanol.



Figure 11: Comparison of the instantaneous vaporization rate, $K_{inst}(t)$ of anhydrous ethanol and ethanol(95%) droplets at different temperatures.

Figure 7 has revealed that the vaporization rate of the initial part, K_i , has almost similar evolution for the two ethanol forms for the global range of temperature examined here. Nevertheless, one can observe in Figure 11, that the instantaneous vaporization rate for the anhydrous ethanol is always higher than the ethanol (95%) for the initial part of the total vaporization time, corresponding to the K_i . This can be explained by the fact that for the calculation of a mean vaporization rate the variation of instant vaporization rates is divided by a time period. For the anhydrous form the time period considered is longer thus compensating the systematic higher values observed for instant vaporization rates and therefore making mean values equivalent to (95%) form.

An interesting observation could be made from Figure 11 is the behaviour of anhydrous ethanol at temperature 673 K where the K_{inst} is always almost constant, 'quasi-steady' and equal to K_i throughout droplet lifetime. The disappearance of ethanol component only occurs at the end of the lifetime, as underlined also by Marchese and Dryer [22]. It seems that at this higher temperature anhydrous ethanol behaves as a single component without or with slight water concentration interference.

Zang and Williams [29] studied the combustion of spherical alcohol droplets under microgravity conditions by theoretical analyses. They explained the water dissolution phenomena. The same observation arises from Marchese and Dryer [22] on methanol droplet combustion where the d^2 curve deviated significantly from the d^2 -law predictions. This behavior is a result of the absorption of combustion intermediates and products. Water is one of the main combustion products and it produces a non-linear d^2 -law behavior. During the alcohol droplet combustion, water first diffuses back to the droplet, and it is then absorbed during the first half of the burning history. Then, the water gradually

builds up inside the liquid and during the second half of the combustion history, vaporizes along with alcohol. Lee and Law [15] reported the vaporization and combustion of freely-falling methanol and ethanol droplets in dry and humid environments. They demonstrated that water vapor, either from the ambience or generated at the flame, can freely condense at the droplet surface and subsequently dissolve into the droplet interior. Cho and al. [18] did the same observations earlier. As mentioned and fully described by Law et al. [14] we can assume that the same phenomena can occur for the vaporization phenomenon alone. During the initial fuel vaporization, the surrounding water vapor condenses at the droplet surface. Then the condensed water further diffuses into the droplet interior because of its miscibility with ethanol. Since the present vaporization rate is based on the rate of change of the droplet diameter, the condensed water tends to artificiality increase the droplet size, slowing down the instantaneous vaporization rate as that can be observed in Figures 9 & 10 during the K_i period. However, this continuous water condensation will decrease because of the reduction in the water vapor pressure difference between the ambience and the droplet surface. Law and collaborators [14] have reported the evolution of the droplet temperature for methanol droplets vaporizing in humid air, and demonstrated that T_s decreases to a minimum and then increases again. These authors have explained this increase by the condensation heat release as well as the favorable wet-bulb temperature of water. In our work, this temperature increase at the end of the ethanol vaporizing period could explain the increase of vaporization rate of water, K_f, observed in Figure 8, comparatively to the distilled water curve. The longer condensation period for the anhydrous ethanol, could explain the higher level of vaporization rate, K_f, especially at high temperatures.

To further substantiate the effect of initial water concentration and environment humidity on ethanol droplet vaporization under different temperatures, calculations of estimated water inside the droplet has been carried out. One of the possible approaches is to estimate the initial diameter of the droplet from the second linear part of the vaporization called afterwards "the water" droplet, from the existing d^2/d_0^2 vs t/d_0^2 curve, as shown in Figure 12. By identifying the inception point where the constant K_f is attained, a horizontal extrapolation will give the value of d^2/d_0^2 . Therefore, as the value of d₀ is known, the squared diameter of the water droplet d^2 is determined. In order to compare both fuels, the initial volume of water contained in the ethanol (95%) droplet is subtracted of the total water volume found at the inception point previously described. The ratio of volume of water droplet to volume of initial droplet is then calculated to estimate the percentage of condensed water existence during the vaporization process (see Figure 13). If the volume percentage is greater than the initial percentage of the composition of hydrous ethanol (95% ethanol + 5% water) this means that the difference comes from the condensation.



Figure 12: Example of determination of water diameter in the droplet from d^2 (t) curves for ethanol (95%) at 473K.

Figure 13 shows the volume percentage of condensed water for both ethanol forms. It is seen that the volume percentage of condensed water is almost constant at all temperature for anhydrous ethanol (approximately 4-6 %), which confirms that the water vaporization observed for anhydrous ethanol is due only to ambient constant humidity. However this is not the case for ethanol (95%). This compound seems to show potential to absorb more condensed water than anhydrous ethanol. The vaporization/absorption process of binary water-ethanol mixture and the pure ethanol is different. It may be noted here in Figure 13, an abnormal behavior for hydrous ethanol at a temperature of 673 K, the values are very low and comparable to anhydrous one, but with high variability. No coherent explanation was helpful up to now to clarify this strange behavior.



Figure 13: Volume percentage of condensed water at different temperatures for both ethanol forms.

CONCLUSION

The influence of water on ethanol isolated droplets vaporization has been explored in nitrogen at atmospheric pressure and for a wide range of temperatures. The experiments were conducted in a heated chamber with a system of cross quartz fibers as droplet support. Two types of ethanol were studied: anhydrous ethanol and a mixture of 95 % ethanol - 5 % water. From the d² curves, average and instantaneous vaporization rates for the two ethanol forms are presented and discussed. For temperatures from 350K to 850K, K_i is varied from 0.018 to 0.100 mm²/s and K_f is varied from 0.004 to 0.050 mm²/s for both ethanol forms. The presence of water initially dissolved in ethanol and water gradually condensing on droplet surface changes the way the droplet vaporization process by modifying the diffusion transport at the droplet surface. As the temperature and concentration at droplet surface change with time, the expected constant vaporization rate is replaced by a complex unsteady process for which the theoretical description must be completed. The need for further measurements and theory completion is strong as alcohol blends are more and more envisioned as alternative fuels. Engines running with such fuels require extensive testing for calibration of numerous functioning parameters. Efforts to reduce calibration costs and length by modeling are widely deployed. As almost every combustion model assumes a constant vaporization rate for droplets, errors arising from an idealized vaporization model are to be expected. Moreover, the strong differences in the vaporization behavior of pure and water blended forms of ethanol that have been put in evidence in this work, call for cautious specification of ethanol based fuels for experiments and engine use regarding initial water content.

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