



# Autoignition and combustion characteristics of heptane droplets with the addition of aluminium nanoparticles at elevated temperatures



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## ABSTRACT

We investigate the effects of high ambient temperatures and various concentrations of nanoparticles (NPs) on the autoignition and combustion characteristics of heptane-based nanofluid droplets. A single, heptane ( $n\text{-C}_7\text{H}_{16}$ ) droplet containing 0.5%, 2.5%, or 5.0% by mass of aluminium (Al) NPs mounted on a silicon carbide fibre was exposed to a rapid increase in temperature (from room temperature to temperatures in the range 600–850 °C) at atmospheric pressure and under normal gravity, and the autoignition and combustion characteristics were observed. The ignition delay, burn rate, and combustion characteristics of pure and stabilised heptane droplets were also examined for comparison. The results show that, as with the pure heptane droplets, the ignition delay of the NP-laden heptane droplets ( $n\text{-Al/heptane}$ ) followed an Arrhenius temperature dependence. The reduction in the ignition delay time with increasing temperature depended on the loading of NPs in the droplet. The overall activation energy obtained for dilute NP concentrations (0.5% by mass) was smaller, and that for dense (2.5% and 5.0%) concentrations of NPs was larger, than that of the pure heptane droplets. Consequently, the addition of 0.5% Al NPs to heptane resulted in ignition at 600 °C, which is not observed with stabilised heptane or 2.5% and 5.0%  $n\text{-Al/heptane}$  droplets. At higher temperatures, the ignition delay of the  $n\text{-Al/heptane}$  droplets was comparable to that of pure heptane droplets. The combustion of the  $n\text{-Al/heptane}$  droplets did not follow the classical  $d^2$ -law, in contrast to the combustion of the pure and stabilised heptane droplets. Regardless of the NP concentration, the  $n\text{-Al/heptane}$  droplets exhibited disruptive burning behaviour, which was characterised by multiple expansions and ruptures or ‘microexplosions’. During these microexplosions, the NPs were ejected from the droplets, and the intensity of the microexplosions increased with increasing temperature; consequently, the burning time and total combustion time of the droplet was reduced. Due to these intense and frequent microexplosions, almost no residue from the Al NPs remained on the fibre following combustion, and a separate Al flame was not observed. The average gasification (burn) rate of the  $n\text{-Al/heptane}$  droplets remained equal to that of pure heptane droplets at relatively low temperatures (600–700 °C); however, at higher temperatures (750–850 °C), it was significantly faster than that of the pure heptane droplets.

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## 1. Introduction

Energetic materials including aluminium (Al) and boron (B) have long been of interest to the propulsion and combustion community due to their large combustion energies. Recent advances in nanotechnology have enabled the large-scale production of nanoparticles (NPs), which have shown significant

advantages over micron-sized particles for combustion applications. When the size of metallic particles is reduced from the micron- to nanometer-scale, the thermophysical properties often change substantially. Numerous studies have reported a decrease in the melting point and heat of fusion of nanometer-scale metal particles [1–3]. There have been a number of reports of the effects of addition of NPs to solid fuels and propellants [4–8], citing advantages over micron-sized particles, including improved energy density, a shorter ignition delay, and an increased burn rate [4].

The use of metallic nanopowders as fuel additives has received much recent attention. Here, we describe the addition of NPs to liquid fuels, forming so-called nanofluid fuels, which have received

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considerable interest for applications in combustion and propulsion. Nanofluid fuels are a new class of nanofluids, and consist of stable suspensions of energetic NPs in conventional liquid fuels. Nanopowders of energetic materials, including Al, B, carbon (C), iron (Fe), and cerium oxide ( $\text{CeO}_2$ ), have been used as additives suspended in conventional liquid fuels in such a way as to improve the ignition and combustion characteristics. Previous studies have shown significant advantages of such nanofluid fuels. Van Deventer and Anderson [9] found enhanced catalytic effects for  $\text{CeO}_2$ /JP-10 nanofluid fuel, and reported a significant reduction in the ignition temperature of JP-10. Tyagi et al. [10] observed an increased hot-plate ignition probability of diesel fuel laden with Al and  $\text{Al}_2\text{O}_3$  NPs. Sabourin et al. [11] obtained enhanced burn rates and reduced ignition temperatures by adding functionalised graphene sheets to monopropellant nitromethane. Allen et al. [12] found a 32% reduction in the ignition delay of Al/ethanol and a 50% reduction in the ignition delay of Al/JP-8 nanofluid fuels compared with their base fuels. Jones et al. [13] obtained an increased heat of combustion with high loadings of Al NPs in ethanol.

As with the evaporation [14] of NP-laden fuel droplets, the combustion of nanofluid fuel droplets is more complex than that of pure liquid fuel droplets due to the multi-component, multi-phase, and multi-scale nature. There have been a number of studies of the combustion of nanofluid fuel droplets. Gan and Qiao [15] observed disruptive combustion of droplets with micron- and nanometer-scale Al particles in suspension with ethanol and *n*-decane. The disruptive behaviour of the micron-scale suspensions occurred later and with a greater intensity compared with nanometer-scale suspensions with the same surfactant and particle concentrations. Gan et al. [16] observed simultaneous combustion of both the droplet and the NPs with dilute suspensions of B and Fe NPs in *n*-decane and ethanol. However, in dense suspensions, most particles were combusted as large agglomerates following the consumption of the liquid fuel.

These reports of the combustion of nanofluid fuels [15,16] describe the simple qualitative behaviour of the combustion of the nanofluid droplets, where the droplets are ignited by a heated wire. Therefore, the effects of NPs on the autoignition delay times and the droplet combustion behaviour at various elevated temperatures are not accurately described. In addition, they are not able to explain the effects of the NPs on combustion rate constants. To the best of our knowledge, there have been no studies reporting the effects of the addition of various concentrations of NPs on the autoignition and combustion characteristics of nanofluid fuel droplets at elevated temperatures.

Here, we experimentally investigate the autoignition and combustion behaviour of heptane-based nanofluid droplets. The effects of the NP loading and ambient temperature on the droplet autoignition and combustion characteristics were examined. We used *n*-heptane ( $n\text{-C}_7\text{H}_{16}$ ) as the base fuel because it is a simple, highly pure, single-component liquid hydrocarbon with an abundance of available experimental data regarding the evaporation, autoignition, combustion, and reaction characteristics. It is therefore useful to investigate the effects of the addition of NPs to this liquid hydrocarbon fuel. Al NPs were used as energetic additives, and the ambient temperature was varied in the range 600–850 °C. These temperatures are higher than the autoignition temperature of heptane (220 °C), and also both below and above the melting point of Al NPs (660 °C). The ambient pressure was maintained at atmospheric pressure, i.e., 0.1 MPa.

## 2. Experimental method

The materials and instruments used in this work were the same as those used in the experimental investigation of the evaporation

characteristics of heptane-based nanofluid droplets described in Ref. [17]. The size distribution of Al NPs is in the range of 30–250 nm with average diameter of most probable particles 70 nm [17]. All materials were used as received and without further treatment, except for the Al NPs, the surfaces of which were coated with oleic acid (OA). A planetary ball mill (Retsch PM100, GmbH, Germany) was used to modify the surface of the Al NPs.

### 2.1. Preparation of stable nanofluid fuels

The heptane-based nanofluid fuels were prepared using a two-step method. In this method, the NPs were uniformly suspended in a base fluid by applying various physical and chemical homogenisation techniques. Because the metallic NPs were available at an industrial scale, it is economical to produce a number of nanofluids with various concentrations of NPs, either in the laboratory or at a larger scale. Surface modification was applied to the Al NPs in order to improve the dispersion stability in heptane. The surfaces of the NPs were coated with OA using a ball mill, and oxide-free, ligand-protected and fuel-soluble Al NPs were obtained [18]. The NP-to-OA ratio was optimised as 1:1 in order to obtain stable suspensions of NPs in heptane. The resulting NP/OA paste was dispersed in heptane via vigorous agitation. In this manner, a homogeneous suspension was obtained, which remained stable for 2 h with no obvious sedimentation of the NPs. Further details of this procedure can be found in Refs. [14,19].

### 2.2. Experimental apparatus and procedure

The experimental setup used to investigate the autoignition and combustion characteristics of a single fibre-suspended nanofluid fuel droplet at elevated temperatures was the same as that described in Refs. [20–23]. This experimental arrangement has been used to study the evaporation behaviour of heptane- and kerosene-based nanofluid fuel droplets [14,17,19]. The experimental procedure, data reduction, and analysis are described in detail in Refs. [14,17,19]. Experiments were carried out with a single nanofluid fuel droplet suspended by a 100- $\mu\text{m}$ -diameter SiC fibre. The initial average diameter of the droplets was  $1.0 \pm 0.10$  mm. The ambient temperature was varied in the range 600–850 °C at intervals of 50 °C, and the ambient pressure was maintained at 0.1 MPa. A high-temperature ambience was provided using a free-falling electric furnace, and the oxidizing environment was dry air. The combustion process was recorded using a high-speed charge-coupled device (CCD) camera (X-Stream XS-3 with a maximum speed of 1390 fps at a resolution of  $1280 \times 1024$ ). Due to low internal memory (4 GB), the images were recorded with a speed of 300 fps at a resolution of  $512 \times 512$ . A flexible image-processing method was developed using Matlab to obtain the diameter of the droplets from the images. The procedure for calculating the diameter of droplet from the captured images was described in our previous study [17]. Briefly, in this method, a threshold value for pixel gray level was carefully set to count the pixels in the droplet zone. Then area of a circle having the same number of pixel which droplet had is calculated, which in turns give the diameter of the droplet using the law of proportions with the reference diameter of SiC fibre (100  $\mu\text{m}$ ). In some cases, the droplet is being distorted during the combustion process. However, by using this code, the diameter of this irregular-shaped droplet was calculated within a reasonable accuracy. This method was executed iteratively for each image, allowing us to obtain the temporal variation of the droplet diameter  $d$  during combustion. If the combustion of the droplet follows a  $d^2$ -law, the combustion rate constant may be expressed as the time derivative of the droplet diameter squared, i.e.,  $K_c = -d(d^2)/dt$ . In this manner, the droplet combustion rate con-

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