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Increased Hot-Plate Ignition Probability for Nanoparticle-Laden Diesel Fuel

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ABSTRACT

The present study attempts to improve the ignition properties of diesel fuel by investigating the influence of adding aluminum and aluminum oxide nanoparticles to diesel. As part of this study, droplet ignition experiments were carried out atop a heated hot plate. Different types of fuel mixtures were used; both particle size (15 and 50 nm) as well as the volume fraction (0%, 0.1%, and 0.5%) of nanoparticles added to diesel were varied. For each type of fuel mixture, several droplets were dropped on the hot plate from a fixed height and under identical conditions, and the probability of ignition of that fuel was recorded based on the number of droplets that ignited. These experiments were repeated at several temperatures over the range of 688−768 °C. It was observed that the ignition probability for the fuel mixtures that contained nanoparticles was significantly higher than that of pure diesel.

Introduction. The addition of nanoparticles to solid fuels and propellants has been part of several recent studies.[1–5] Such studies have shown that there are multiple advantages of adding nanoparticles to propellants and solid fuels such as shortened ignition delay, increased energy density, and high burn rates.[1] Recent studies have also shown that the addition of nanoparticles to a fluid can enhance its physical properties such as thermal conductivity,[6–8] mass diffusivity,[9] and radiative heat transfer.[10,11] As a result, it is possible in principle to achieve the desired properties of a fluid by adding some specifically tailored nanoparticles. However, little work has been reported in the past on the effect of adding nanoparticles to *liquid* fuels.

Some research, however, has taken place to study the feasibility of adding additives and micrometer-sized metallic particles to solid and gel propellants to provide high-energy output.[12] However, among liquid fuels, the micrometer-sized particles tend to sediment quite quickly. Studies have also shown that because the nanoparticles are small enough to approach molecular dimensions, their properties can be significantly different from those of larger, micrometer-sized particles.[3] At such dimensions, the surface-area-to-volume ratio of the particle increases considerably and hence allows more fuel to be in contact with the oxidizer.[5] Furthermore, because of the small distances (interparticle as well as particle size) involved with nanoparticles, the time scales of the chemical reactions are very different compared with those associated with larger size particles, and as a result, the ignition delay time for nanosized particles would be much shorter than those of micrometer-sized particles.[4]

In particular, ignition delay and ignition temperature are critical parameters that characterize the performance of a diesel engine. Both efficiency as well as emission levels from a diesel engine can potentially be improved by optimizing the ignition delay and ignition temperature.[13] The objective of the present study is to quantify the influence of adding aluminum and aluminum oxide nanoparticles to diesel fuel on its hot plate ignition probability. To characterize this, hot plate ignition experiments were carried out in which individual droplets of the fuel were allowed to fall in a controlled environment on a heated metallic plate and the event of ignition or nonignition was noted. Such experiments were carried out using different concentrations of nanoparticles + diesel mixtures and at several temperatures in the range of 688−768 °C. It was observed that the hot plate ignition probability of the diesel fuel increases significantly by the addition of the nanoparticles. Moreover, the presence of some residue particles on the hot plate surface is also believed to contribute to an increase in the ignition probability of pure diesel.

Experimental Setup. The work presented here was carried out in a closed fume hood. The schematic of the experimental setup is shown in Figure 1. As can be observed from the

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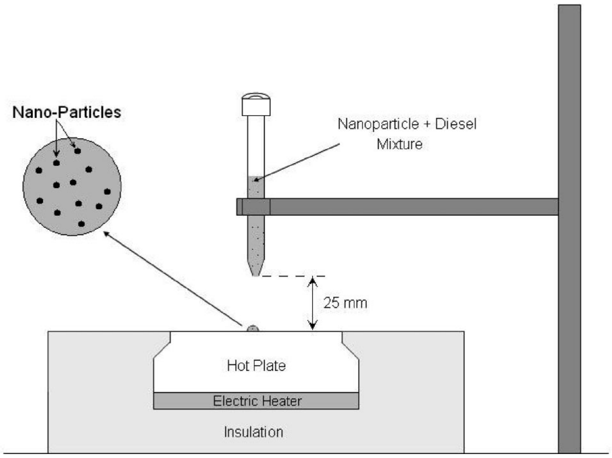


Figure 1. Schematic of the experimental setup. A single droplet of fuel was allowed to fall from the pipet on to the top of the hot plate.

figure, the hot plate is placed directly on top of an electrical heater. Insulating material (glass wool) was placed around them in order to minimize the heat losses. The hot plate was constructed of stainless steel and had a diameter of 75 mm at the top. It had a small concave curvature at the top in order to ensure that the droplet did not move off the top surface. A plastic pipet was connected to a movable arm such that it could be positioned directly above the hot plate when desired. To minimize the movement of the droplet, the hot plate was carefully leveled and positioned such that the droplet fell as close as possible to the center of the plate. To begin with, the hot plate was carefully polished and cleaned in order to minimize the influence of the surface or any effects of hot spots. This was done by rotating the hot plate in a lathe and then polishing it using a very fine grit sand paper until a mirror-like reflectivity was obtained. Moreover, whenever a new set of fuel mixtures that contained a different type of nanoparticles was measured, a freshly polished plate was used to start those experiments. The same pipet was used throughout this study in order to prevent any changes in the droplet size. Moreover, the height from which the droplets were allowed to fall (25 mm) was chosen by minimizing the chance of the flame igniting the fuel in the pipet, while keeping it as close as possible to the hot plate to eliminate the chances of drop breakup, and was kept the same throughout these experiments. The temperature of the hot plate was continuously monitored using three thermocouples. The thermocouples were k-type and had an uncertainty of ±2 °C. The instantaneous temperature data were recorded on a desktop computer using a data acquisition system. A LabView program was used in order to check and record the temperatures. A simple procedure was followed during these experiments. At any given time, a single droplet of the fuel was allowed to fall on top of the hot plate. Either of the two possible events (droplet ignition or no droplet ignition) was recorded. This process was repeated 50 times for each sample in order to obtain a statistically sound data set. Such measurements were then repeated at different hot plate temperatures and for different nanoparticle/fuel mixtures.

To characterize the size of the droplets used in these experiments, a size distribution of the droplets was carried out by weighing a large set of droplets (50 droplets) in a mass balance. The analysis showed a mean droplet size of 25.3 mg with a standard deviation of 1.5 mg. It was observed that the distribution remained the same irrespective of the type of fuel mixture used. In addition to the size of the droplet, the time required for a droplet to ignite once it landed on the hot plate was also measured using a high speed camera. This measurement was carried out on a small sample of droplets, and it was found that the droplets typically required 1.1 s to ignite and a little less than 4 s to completely burn up. Also, it was recorded that the droplets were ignited in sequence with about a 10 s gap on average between any two droplets.

Several types of nanoparticle + diesel mixtures were examined in these experiments. The detailed description of all the mixtures and the various parameters that were varied are described in this paragraph. There were four main parameters that were varied in this study: nanoparticle material, nanoparticle size, volume fraction of nanoparticles in the fuel mixture, and the hot plate temperature. First, the nanoparticles used in this study consisted of two types: aluminum nanoparticles and aluminum oxide nanoparticles. Second, two sizes of nanoparticles were chosen: the aluminum nanoparticles were of only one size specified by the manufacturer (50 nm), while the aluminum oxide nanoparticles were of two sizes, 15 and 50 nm, again as specified by the manufacturer. Third, the volume fraction of the nanoparticles present in the fuel mixture was varied: 0%, 0.1%, and 0.5%. And finally, the hot plate temperature was varied from 668 to 768 °C in increments of 20 °C during the course of these experiments. At each condition, two sets of experiments were carried out, with each set consisting of 50 droplets. To discuss the results obtained in this study, the following nomenclature is used: For example, when experiments were conducted on the mixture that had 0.1% volume fraction of 50 nm aluminum particles, it is referred to as "0.1% Al (50nm) + diesel mixture" from this point forward in this paper. Similarly, when experiments were conducted on the mixture that had 0.5% volume fraction of 15 nm aluminum oxide particles, it is referred to as "0.5% Al_2O_3 (15 nm) + diesel mixture" from this point onward in the text as well as in the figures.

In this study, aluminum nanoparticles were chosen based on their prior history of use in the literature in various combustion applications, especially in propellants, as mixtures in solid and gel fuels.[1–3,5] The aluminum nanoparticles used in this study were purchased from Nanotechnologies Inc. The mean diameter of the particles, according to the manufacturer's specifications, was 50 nm. Because these experiments were performed in an open enclosure, for safety concerns, the particles were passivated in advance, i.e., they contained a thin oxide layer (1.5 nm). On average, the resulting particles consisted of 71% aluminum and 29% aluminum oxide by weight. In addition to aluminum nanoparticles, aluminum oxide particles (which consisted of more than 99% aluminum oxide by weight) were also examined

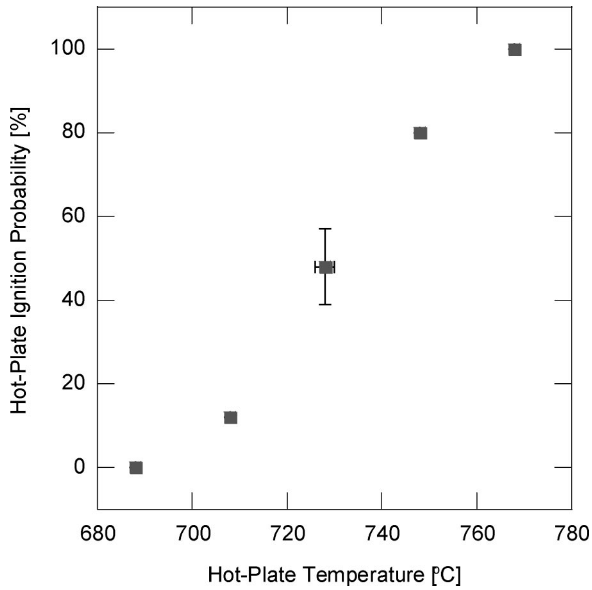


Figure 2. Ignition probability of pure diesel.

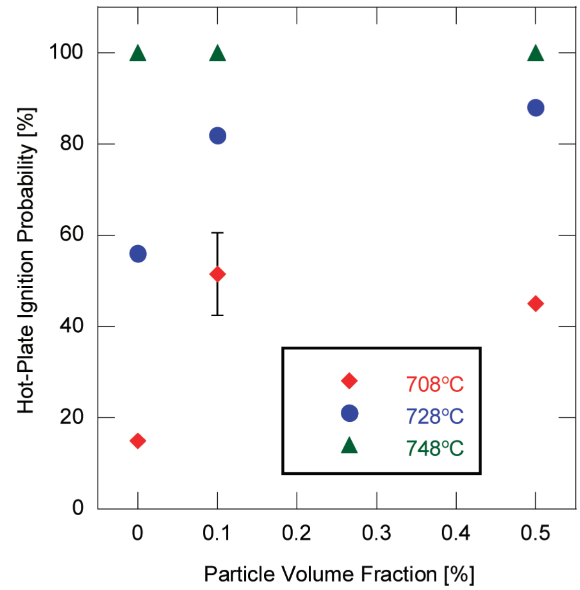


Figure 3. Ignition probability of Al (50 nm) + diesel mixtures at hot plate temperatures of 708, 728, and 748 °C.

in this study. The motivation behind this was to choose a material that would not oxidize/combust so as to compare its results with those of aluminum nanoparticles, which could potentially oxidize/combust under certain conditions. Two sizes of aluminum oxide nanoparticles were chosen in order to investigate any link between the particle size and the ignition characteristics of the fuel mixtures. The two samples of aluminum oxide particles used in this study were also purchased from Nanotechnologies Inc. and had mean diameters, again according to the manufacturer's specifications, of 15 and 50 nm, respectively.

The nanoparticle and fuel mixtures were created by careful measurement of the individual masses and then combining the components at the desired concentration. To ensure proper mixing of the particles as well as to separate any particles that might have clustered together, each of the mixtures was freshly prepared just before the experiments by sonicating them for at least 30 min in a Hielscher GmbH UP200S ultrasonic processor. The mixtures were sonicated at a frequency of 24 kHz and at a power rating of 200 W. After sonication, the mixtures were allowed to cool down to room temperature, or approximately 60 min, before starting the experiments. No surfactants were added to the samples.

Results and Discussion. Figure 2 shows the hot plate ignition probability results obtained for pure diesel. Such results were obtained by conducting experiments starting at the lowest temperature of 688 °C and then repeating the experiments by increasing the plate temperature at increments of 20 °C. Hence this way experiments were conducted at temperatures of 688, 708, 728, 748, and finally at 768 °C. At each temperature, the experiments were repeated twice and the mean value of the ignition probability was calculated by averaging the results over the two runs. As shown in this figure, the ignition probability was found to be 0% at the lowest temperature (688 °C) and then gradually increased to 12%, 48%, and 80% at temperatures of 708, 728, and 748 °C, respectively, and eventually reached 100% at 768 °C. On the basis of the two sets of results, the uncertainty in the ignition probability was calculated. The uncertainty in ignition probability was found to be ±9.1% (absolute), and the uncertainty in hot plate temperature was found to be ±2 °C. These values are represented in Figure 2 using error bars on a single point. The uncertainty in ignition probability was calculated using the Student-*t* method[14] and assuming a 95% confidence level.

After completing the experiments with pure diesel, next a similar procedure was followed, but this time it was carried out on diesel mixtures that contained aluminum nanoparticles. Three types of fuel mixtures were examined: pure diesel, 0.1% Al (50 nm) + diesel, and 0.5% Al (50 nm) + diesel. The reader is referred to one of the previous paragraphs where these notations are described in detail. These experiments were performed in a particular sequence, starting at the lowest temperature (708 °C) first with pure diesel and then with 0.1% Al (50 nm) + diesel mixture and then with 0.5% Al (50 nm) + diesel. Next, the temperature was raised to 728 °C and the measurements were repeated starting once again with pure diesel and then with 0.1% Al (50 nm) + diesel and then with 0.5% Al (50 nm) + diesel. The same process was again repeated after increasing the hot plate temperature to 748 °C. At each of these conditions, two sets of experiments were carried out, with each set consisting of 50 droplets. The results obtained from these two sets were averaged, and the mean values are shown in Figure 3. The individual uncertainties in ignition probability were evaluated, and a representative value is shown in Figure 3 as error bars. Note that the results in Figure 3 for the pure diesel are not from the same experimental runs as the results shown in Figure 2. As can be clearly seen in Figure 3, the ignition probability for the 0.1% Al (50 nm) + diesel mixture is much higher than that of the pure diesel sample at a hot plate temperature of 708 °C. At that temperature, the mean ignition probability for pure diesel is about 15% and that for 0.1% Al (50 nm) + diesel is about 51%. The uncertainties of

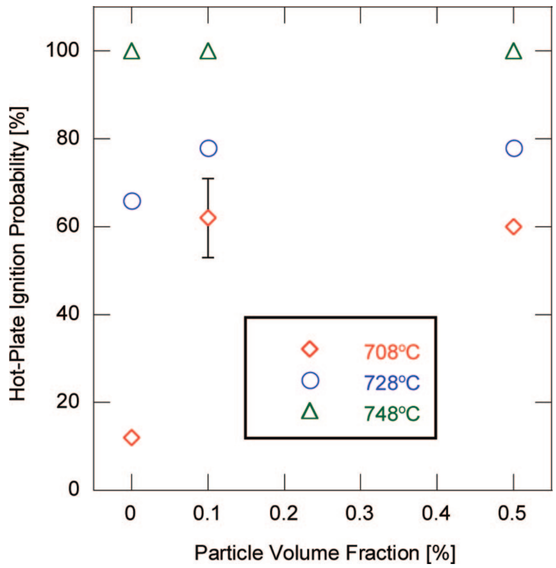


Figure 4. Ignition probability of Al_2O_3 (50 nm) + diesel mixtures at hot plate temperatures of 708, 728, and 748 °C.

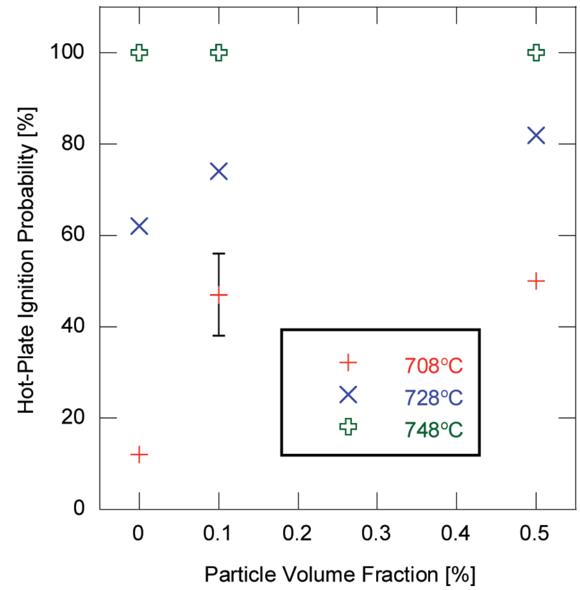


Figure 5. Ignition probability of Al_2O_3 (15 nm) + diesel mixtures at hot plate temperatures of 708, 728, and 748 °C.

ignition probability for these two points were found to be ±9.1% and ±9.5%, respectively. Hence, on the basis of these data, we can clearly conclude that adding aluminum nanoparticles to pure diesel significantly increases the ignition probability. A similar trend is also observed if we compare the results for pure diesel and 0.5% Al (50nm) + diesel mixtures at 708 °C. The mean value of ignition probability for 0.5% Al (50 nm) + diesel mixture at 708 °C is 45%, with an uncertainty of about ±9.9%. Although the results at 708 °C are roughly equivalent for the 0.1% Al (50 nm) + diesel and 0.5% Al (50 nm) + diesel mixtures, it is quite clear that both of them exhibit significantly higher hot plate ignition probabilities than pure diesel. A similar trend is also observed for the data points that were performed at 728 °C, where it can be observed that the ignition probabilities for both 0.1% Al (50 nm) + diesel mixture and 0.5% Al (50 nm) + diesel mixture are much higher than that for pure diesel. Finally, when the hot plate temperature was increased to about 748 °C, the ignition probabilities for all the three mixtures, including pure diesel, were found to increase and reach a value of 100%.

Figure 4 shows the results that were obtained using the various Al_2O_3 (50 nm) + diesel mixtures. Before beginning these experiments, a freshly polished plate was prepared in order to eliminate any influence of prior residue deposition on the hot plate. Once again, the exact same procedure and the same sequence of experiments was followed as was mentioned in the previous paragraph, corresponding to the Al (50 nm) + diesel mixtures, with the only difference being that this time the samples consisted of Al_2O_3 (50 nm) + diesel mixtures. From Figure 4, it can be clearly observed that the mixtures containing nanoparticles show significantly higher ignition probability compared to pure diesel (at temperatures of 708 and 728 °C), while all the mixtures burn 100% of the time at a temperature of 748 °C. Similarly, Figure 5 shows the results that were obtained when the experiments were repeated (starting with a freshly polished hot plate) using Al_2O_3 (15 nm) + diesel mixtures. Here again the same observations can be drawn, that adding nanoparticles tends to significantly increase the ignition probability of the fuel.

On the basis of the results obtained in this study, several conclusions can be made about the effect of parameters such as particle material and particle size on the ignition probabilities of the fuel mixtures by comparing the plots shown in Figures 3–5. To observe the effect of particle material on ignition probability, Figures 3 and 4 can be compared because the same sizes of nanoparticles were used in both. Figure 3 shows the comprehensive results of ignition probability that were obtained using the various Al (50 nm) + diesel mixtures and, similarly, Figure 4 shows the complete set of results that were obtained by using Al_2O_3 (50 nm) + diesel mixtures. Taking into account the uncertainties involved in these experiments (approximately 10%), it seems that the mixtures containing these two types of nanoparticles behave quite similarly and show quite analogous results. In both cases, the addition of nanoparticles increases the ignition probability quite significantly (at hot plate temperatures of 728 and 748 °C), and in both cases, there is very little difference in the ignition probabilities between the 0.1% volume fraction mixtures and the 0.5% volume fraction mixtures. However, there was one difference that was observed between the mixtures that contained aluminum particles with those that contained aluminum oxide particles. It was observed that in almost all cases where the mixtures contained aluminum nanoparticles, once the droplet combustion took place, it was followed by a visible "glowing" of the aluminum particles left on the hot plate. In contrast, when the aluminum-oxide + diesel samples were ignited in a similar fashion, no such "glowing" was ever observed.

It was interesting to compare the ignition behavior of the droplets at the beginning of the experiments with those toward the middle and at end of the experiments. Figure 6 shows the ignition probability of individual subsets of 10 droplets in each of 2 sets of 50 droplet experiments that were

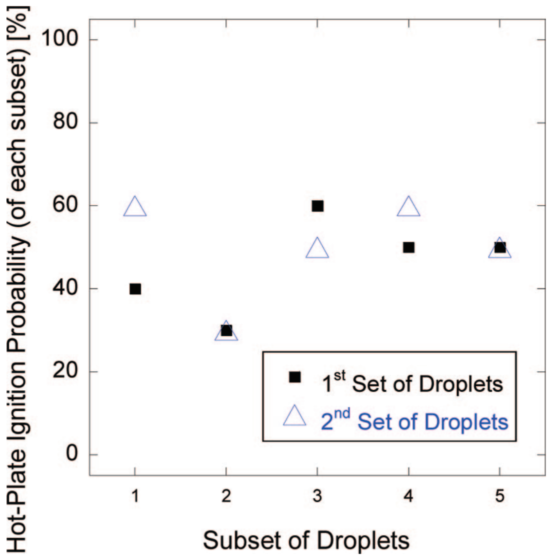


Figure 6. Ignition probability of individual subsets (10 droplets each) of 0.5% Al (50 nm) + diesel mixture at hot plate temperature of 708 °C.

conducted using 0.5% Al (50 nm) + diesel mixtures. The ignition probability of the first set as a whole was 46% and that for the second set was 50%. However, the ignition probability of each individual subset was quite different from one another and exhibited a scattered behavior. As can be seen in Figure 6, the ignition probabilities of individual subsets may vary by as much as 30% (absolute), however, the overall ignition probabilities of the two sets still remained quite close (46% and 50%). According to these results, it seems that the ignition probability does not seem to follow any trend (either increasing or decreasing) when progressing from the start toward the end of a set of 50 droplets.

Finally, to observe the effect of particle size on ignition probability, Figures 4 and 5 can be compared. Figure 4 shows the comprehensive results of ignition probability that were obtained using the various Al_2O_3 (50 nm) + diesel mixtures, and Figure 5 shows the complete set of results that were obtained by using Al_2O_3 (15 nm) + diesel mixtures. Comparing these figures suggest that the overall trend of the ignition probability remains the same even when different sizes of nanoparticles (of the same material) were used. Although, because in this study only two sizes of particles (15 and 50 nm) were used, it is difficult to forecast that size would never have any effect on the ignition probability. Moreover, there seems to a small difference between the two; the results for Al_2O_3 (15 nm) show a slightly lower ignition probability compared to the Al_2O_3 (50 nm) samples, however, only at the hot plate temperature of 708 °C.

Interestingly, when the corresponding results of pure diesel (shown in Figure 2) are compared with the results of pure diesel (shown in Figures 3, 4, and 5), one significant difference can be observed. The ignition probability of pure diesel at a hot plate temperature of 748 °C is 80% according to the data shown in Figure 2, however, the value of ignition probability of pure diesel is 100% in each of Figures 3, 4, and 5. A similar trend is also visible if the ignition probability for pure diesel at 728 °C (which according to Figure 2 is

(a)
(b)

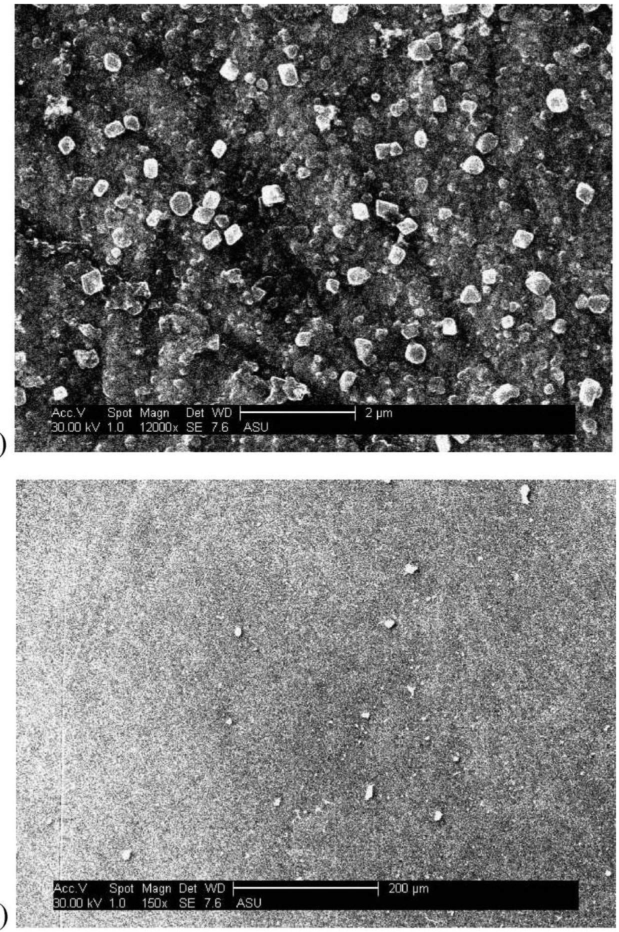


Figure 7. Photograph of the top surface of disk 1, taken by scanning electron microscope (SEM) at a magnification of (a) 12000× and (b) 150×. This disk was heated to 750 °C and then several 0.1% Al (50nm) + diesel droplets were ignited on top of it.

48%) is compared to the values shown in Figures 3, 4, and 5 (which are 58%, 62% and 60%, respectively). Clearly, there are significant differences in the ignition probabilities of pure diesel depending on whether a particular set of experiments was performed on an *uncontaminated* surface (which is the case in Figure 2) or if it was performed on a surface on which prior experiments had been carried out using any nanoparticle + diesel mixture (which is the case in each of Figures 3, 4, and 5). Also, it can be clearly observed that these increases in ignition probabilities are comparatively larger than the uncertainties associated with measurement of ignition probability in the current experimental setup. It is suspected that when experiments using fuel mixtures that contain nanoparticles are performed, some residue is deposited on the hot plate surface. The presence of any residue on the hot plate might influence the ignition probabilities of the subsequent batches of pure diesel droplets, which in normal circumstances would have encountered an *uncontaminated* surface with no deposited residue. To confirm this possibility, it was decided to analyze the top surface of the hot plate using an scanning electron microscope (SEM). However, the hot plate dimensions were too large to fit inside the SEM machine, hence an indirect method was employed to analyze the surface. For this purpose, three small steel disks (diameter

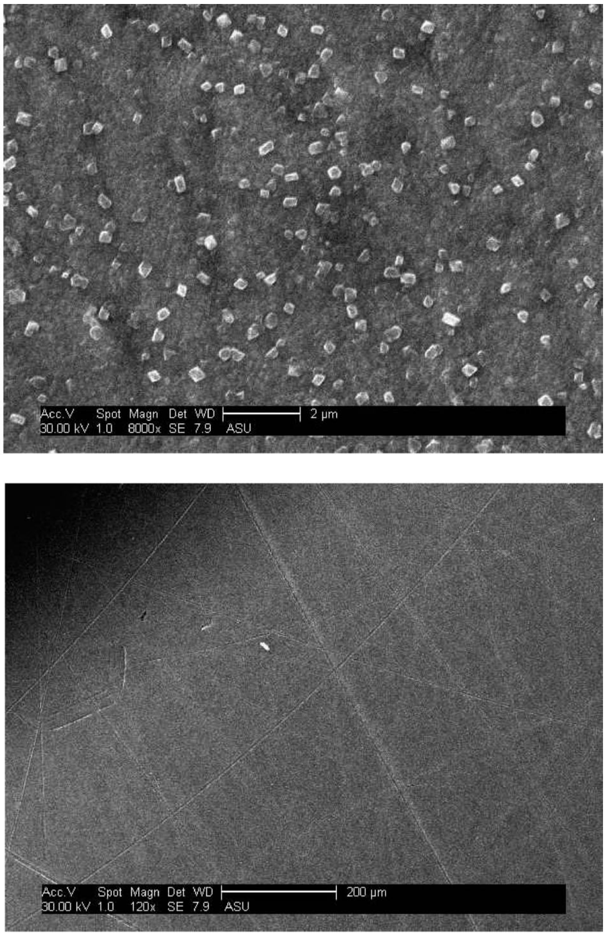
(a)
(b)


Figure 8. Photograph of the top surface of disk 2, taken by scanning electron microscope (SEM) at a magnification of (a) 8000× and (b) 120×. This disk was heated to 750 °C and then several pure diesel droplets were ignited on top of it.

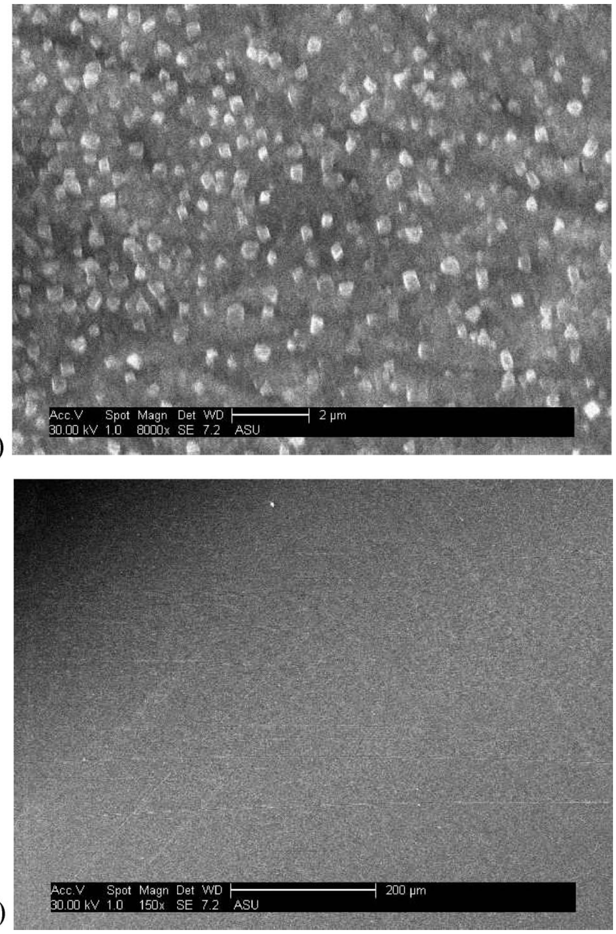
(a)
(b)


Figure 9. Photograph of the top surface of disk 3, taken by scanning electron microscope (SEM) at a magnification of (a) 8000× and (b) 150×. This disk was just heated to 750 °C and then cooled down (no droplets were ignited on top of it).

3 mm, and height 1 mm) were fabricated and were used as a proxy for the actual hot plate for performing the SEM analysis. These disks were placed on top of the hot plate and allowed to reach a temperature of 750 °C. The following notation and procedure was followed: Disk-1: this disk was heated to 750 °C and then several 0.1% Al (50 nm) + diesel droplets were ignited on top of it. Disk-2: this disk was heated to 750 °C and then several pure diesel droplets were ignited on top of it. Disk-3: this disk was just heated to 750 °C and then cooled down (no droplets were ignited on top of it). Hence by comparing the SEM images of these three disks, some conclusions can be drawn about the presence of residue on the disks. These SEM images for disk-1, disk-2, and disk-3 are shown in Figures 7, 8, and 9, respectively. Figures 7a, 8a, and 9a show the images that were taken at a higher magnification, while Figures 7b, 8b, and 9b show the images taken at a lower magnification. The actual magnification values are mentioned in the captions underneath Figures 7–9. As can be observed in Figures 7a, 8a, and 9a, there appear to be some nanostructures present on all the three disks. However, using the energy dispersive spectroscopy (EDS) technique, it was found that those nanostructures were not made of aluminum but instead consisted predominantly of iron. Because the three disks were made of stainless steel, it seems that those structures were just part of the disks themselves. However, when Figures 7b, 8b, and 9b are compared, there are some clear distinctions between the three disks: there are clearly some residue deposits visible on top of disk-1, on which 0.1% Al (50 nm) + diesel droplets were ignited. Moreover, these residue particles, seen on top of disk-1 in Figure 7b, are quite large in size (~10 μm) and are clearly distinguishable from the much smaller nanostructures (~100 nm) that are present on all three disks and are seen in Figures 7a, 8a, and 9a. Using the EDS technique, it was confirmed that the residue particles (~10 μm) seen on disk-1 are predominantly made of aluminum. On disk-2 and disk-3, where no nanoparticle + diesel mixtures were ignited, there are no such residue particles present. Hence, on the basis of all the images obtained by SEM in this study, it can be inferred that some residual deposition occurs on the hot plate surface when any of the nanoparticle + diesel mixtures are used. And they could be responsible for the increase in ignition probability of pure diesel droplets, which were subsequently ignited on a contaminated surface, as was suspected earlier.

Moreover, it was observed that the amount (and size) of the residue remaining on the hot plate depended primarily on the volume fraction of nanoparticles present in the fuel mixture and did not depend on either the particle size (15 or 50 nm) or the particle type (Al or Al_2O_3). Also, the large size of residue particles remaining on the hot plate (~10 μm) as seen in the SEM images are most likely due to the fact that, as the droplets evaporated and ignited, the nanoparticles inside it agglomerated and melted to form larger size residue particles.

Conclusion. Hot plate ignition probability measurements were carried out on various volume fractions of aluminum + diesel mixtures and aluminum-oxide + diesel mixtures. In addition, two sizes (15 and 50 nm) of nanoparticles were used in these experiments. These measurements were conducted at several temperatures within the range 688 °C up to 768 °C. The ignition probability of the nanoparticles + diesel mixtures in all cases was observed to be much higher than that of pure diesel. It was observed that neither the change in nanoparticle material nor the nanoparticle size influenced the ignition probability of the nanoparticle + diesel mixtures. It is possible that adding nanoparticles to the fuel caused significant improvements in its radiative and heat/mass transfer properties and hence the droplets ignited at a much lower temperature and also more often as compared to pure diesel. Such an increase in heat and mass transfer properties of the fuel has the potential of reducing the evaporation (and ignition) time of droplets within a diesel engine and hence should favorably influence its ignition delay. Moreover, SEM analyses of the hot plate surface indicate that the presence of residue particles on the hot plate surface may have also participated in the enhancement of the ignition probability of pure diesel.

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