

Article

Study on the Combustion Characteristics of Ethanol Nanofuel

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Abstract: This study investigates the thermophysical and combustion characteristics of ethanol-based nanofuels incorporating aluminum (Al) and nickel-coated aluminum (Ni-Al). The nanofuels are prepared with varying concentrations of Al and Ni-Al nanoparticles. The results reveal that, despite the non-uniform deposition of nickel on Al particles, a sintering reaction occurs between the two materials. Nanofuels containing Al exhibit unburned Al residues after combustion, while nanofuels containing Ni-Al show intense AlO radical emission during combustion termination, indicating enhanced combustion. However, nanofuels containing Ni-Al demonstrate a lower burning rate compared to Al nanofuels, attributed to the lower thermal conductivity of nickel. Overall, the findings suggest that nanofuels containing Ni-Al possess higher energy potential but extended combustion duration.

Keywords: nanofuel; droplet; combustion; aluminum particles; nickel coating; burning rate

1. Introduction

Ever since the inception of the first internal combustion engine, research has been dedicated to exploring diverse propulsion systems in vehicles, aircraft, and ships that rely on hydrocarbon-based fuels. Consequently, the aviation sector has witnessed a remarkable enhancement in the efficiency of propulsion systems over time. Conversely, investigations into hydrocarbon-based fuels have remained relatively unexplored. However, recent technological strides in nanomaterial fabrication and measurement have invigorated the pursuit of studies involving high density energy, high specific impulse fuels that harness these nano materials.

The definition of nanofuel is a fuel to which nanomaterials are added, suspended in the form of a colloid within the base fuel. This addition of nanomaterials enhances thermal conductivity and heating value in comparison to conventional fuels [1]. When nanofuel is employed within a heat engine in the form of a spray, it possesses the potential to augment overall efficiency and performance by amplifying the heat release from the fuel. As a result, numerous researchers have devoted their efforts to the study of nanofuel droplet combustion. Their focus revolves around several factors, including the base fuel type, the variety of nanoparticles integrated, and their concentration for optimizing combustion performance [2–5].

Tyagi et al. [1] aimed to augment the ignition properties of diesel fuel by investigating the impact of introducing aluminum (Al) and aluminum oxide (Al₂O₃) nanoparticles. They executed droplet ignition experiments on a heated hot plate, employing diverse fuel mixtures characterized by different particle sizes and volume fractions of nanoparticles. Droplets were deposited onto the hot plate, and the probability of ignition was quantified based on the number of ignited droplets. The outcomes underscored that fuel mixtures containing nanoparticles exhibited markedly higher ignition probabilities in contrast to pure diesel.



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Gan et al. [6] delved into the combustion characteristics of fuel droplets containing both nano and micron-sized Al particles. Various parameters like particle size, surfactant concentration, and base fluid type were systematically altered. This study revealed that micron suspensions displayed delayed but more intense micro-explosion behavior, subsequently leading to extensive particle combustion. Conversely, nanosuspensions encountered delayed and incomplete combustion due to the formation of oxide shells on larger agglomerates. This disparity originated from distinct particle agglomerate structures that manifested during early stages, influenced by Brownian motion for nanosuspensions and fluid motion for micron suspensions.

Wang et al. [2] explored the influence of adding cerium oxide (CeO_2) nanoparticles to diesel fuel on the evaporation and micro-explosion characteristics of droplets under high temperature and pressure conditions. Leveraging high-speed imaging, the study unveiled those nanoparticles initiated distinct micro-explosion processes, resulting in an initial increase in evaporation rates followed by a subsequent decrease due to shell formation. Micro-explosion delays exhibited a significant reduction with rising nanoparticle concentrations. The study introduced two novel parameters, expansion and micro-explosion intensity, to quantitatively measure these effects. The intensity notably amplified with escalating nanoparticle content.

Nevertheless, several researchers have reported that many of the Al nano particles in the nanofuel droplets were unburnt and remained as a residue even after the combustion completed. This phenomenon emerges from the inhibitive effect of the oxide layer present on Al particles with a high melting point, which impedes ignition characteristics. The failure of ignition during aluminum combustion results in unburnt residue after droplet combustion [7]. In addition, this leads to a decrease in the heat released from the droplet combustion which worsens the utilization of aluminum nanofuel. To surmount this challenge, studies have been carried out to enhance the reactivity of Al particles in the nanofuel.

Chen et al. [8] coated Al nanoparticles with ammonium perchlorate (AP) at varying levels, resulting in uniform coatings. These coated nanoparticles were integrated with JP-10 to produce a nanofuel. The testing affirmed that the AP coating substantially enhanced ignition combustion by boosting Al nanoparticle oxidation and reducing incomplete JP-10 combustion. Nevertheless, the study noted that while combustion intensity and temperature initially ascended with greater AP content, they subsequently declined. This observation indicates that an excessive amount of AP, with its lower calorific value, is counterproductive in achieving optimal combustion enhancement.

A study led by Ao et al. [9] investigated a novel nanofluid fuel formulated by blending kerosene with nano-aluminum (n-Al) particles coated with polydopamine (PDA). The research probed the influence of varying PDA coating durations on combustion stability. The ignition behavior of n-Al/kerosene droplets exhibited limited sensitivity to the PDA layer. The n-Al@PDA (2 h)/kerosene demonstrated superior ignition and combustion attributes when contrasted with uncoated n-Al/kerosene and other PDA-coated nanofluid fuels.

In the solid propellants research, diverse studies were conducted on the ignition behavior of micro-sized Al particles coated with various organic [10,11] or inorganic materials, such as copper [12], carbon [13], iron [14–16], lithium [17], and nickel [14,18]. Among these, nickel coating has been recognized for significantly reducing ignition delay time and ignition temperature through nickel-aluminum intermetallic reactions [18,19].

In this study, n-Al particles underwent nickel coating followed by an examination of the image and composition of the nickel-coated aluminum (Ni-Al) particles using scanning electron microscope/energy dispersive spectrometer (SEM/EDS) analysis. Additionally, the sintering reaction between nickel and Al was scrutinized through thermogravimetric analysis/differential scanning calorimetry (TGA/DSC) analysis. Two variants of nanofuel were created: one containing Al and the other containing Ni-Al. Experimental research on combustion characteristics was carried out using photo multiplier tube (PMT) and

high-speed imaging measurements. Furthermore, SEM/EDS composition analysis of the residue post-combustion provided insights into the combustion status of the nanofuel.

2. Experimental Setup

The experimental setup employed in this study is depicted in Figure 1 and comprises three primary components: the test bed for droplet combustion, the optical measurement system for quantifying droplet combustion characteristics, and the control unit responsible for ignition sequencing and synchronized data collection.

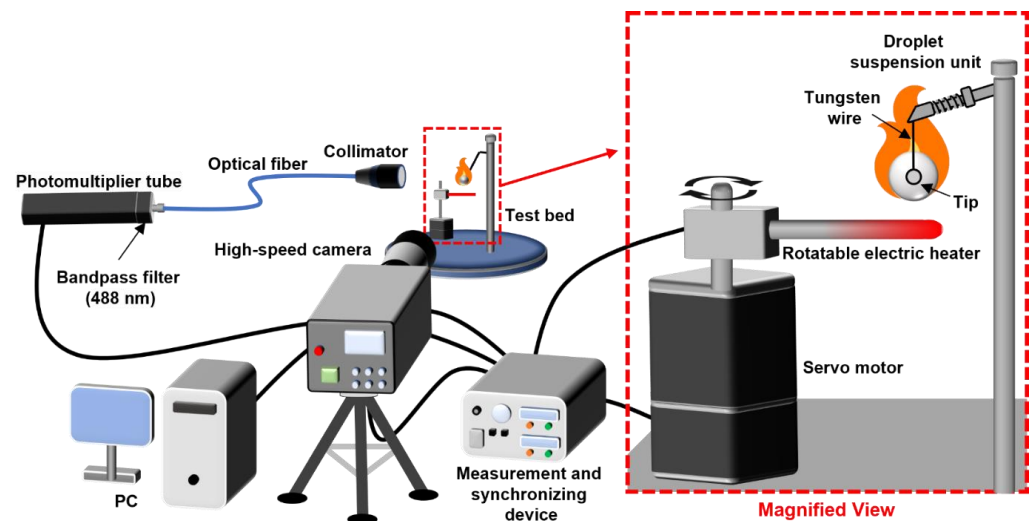


Figure 1. Illustration of experimental apparatus.

2.1. Test Bed

The bed consists of an airborne droplet suspension unit and an electric heater for initiating droplet ignition. The droplet is securely placed at the tip of a tungsten wire, which has an approximate diameter of 100 μm . Tungsten was chosen due to its high melting point, low thermal expansion rate, and strong resistance to corrosion, making it the ideal material for immobilizing burning droplets. To effectively secure the droplet, a ceramic glue bead is affixed to the end of the wire.

Prior to the ignition of the nanofuel droplet, the electric heater must reach a temperature suitable for ignition. However, this process entails a certain time delay that often results in evaporation before the droplet can ignite. In order to minimize droplet evaporation, the electric heater is positioned away from the droplet. Once the heater reaches the required temperature, a servo motor is engaged to pivot the electric heater toward the droplet's ignition point.

2.2. Optical Measurement System

The measurement apparatus consists of a PMT (H10722-20, Hamamatsu Photonics, Japan), along with a high-speed camera (Fastcam Mini UX100, Photron, Japan). The use of the high-speed camera is identical to the study conducted by authors [20]. MATLAB post-processing code is utilized for image analysis to determine droplet diameter, enabling combustion rate calculation. PMT used for detecting emission of AIO radicals, exhibits robust intensity in the $\text{B}^2\Sigma^+ - \text{X}^2\Sigma^+$ band within the visible spectrum. Previous studies have extensively measured the most intense emission at 488 nm in relation to Al combustion [21,22]. In this investigation, the PMT is employed to measure the emission of AIO radicals from the combustion flame of nanofuel droplets, which is filtered through a bandpass filter. Further elaboration on this process is found in Section 3.3.

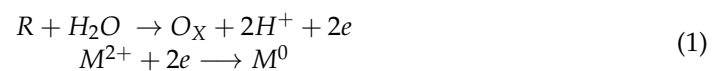
2.3. Nanofuel Preparation

Ethanol serves as the base for the nanofuel. Its minimal emission of visible light during combustion facilitates accurate droplet size measurement and has a negligible impact on the measurement of AlO radicals. Two types of additives were prepared: commercially available Al powder with a nominal diameter of 100 nm and Ni-Al powder synthesized through an electroless plating process [23]. Nanoparticles were added to ethanol and ultrasonically dispersed. The composition of the nanofuel used in this study is detailed in Table 1.

Table 1. Composition of the nanofuels used in this study.

Base Fuel	Metal Powder	Concentration (wt.%)
Ethanol	-	-
Ethanol	Al	2.5%
Ethanol	Ni-Al	2.5%
Ethanol	Al	5%
Ethanol	Ni-Al	5%

The electroless plating method is a coating technique that employs chemical reduction, eliminating the requirement for an electrical power source to deposit metals. This approach is known for its simplicity, capacity for thickness regulation, capability to achieve uniform plating, and extensive application across various industries. Its adaptability to objects of varying sizes and shapes makes it particularly well-suited for coating Al particles. The chemical process responsible for nickel coating on Al particles can be depicted using the following equation.



Initially, when the reducing agent R comes into contact with water, it undergoes oxidation, leading to the formation of oxide O_x , while simultaneously releasing hydrogen and electrons. Within this process, the released electrons combine with metal ions M^{2+} and effectively reduce them to their elemental state M^0 . Specifically, in the context of this study, the metal being reduced to its elemental state is nickel and reducing agents such as sodium hypophosphite, borohydride compounds, and hydrazine can be employed.

For this study, sodium hypophosphite (NaH_2PO_2) served as the designated reducing agent, while nickel sulfate ($NiSO_4$) was employed as the source of nickel ions. Sodium citrate ($Na_3C_6H_5O_7$) was selected to act as the complexing agent. Additionally, maleic acid was introduced to fulfill the role of stabilizer, ensuring consistent conditions. The subsequent coating process spanned a duration of 2 h and was executed while maintaining the solution's temperature at approximately 338 K. Throughout this meticulous procedure, NH_4OH was judiciously incorporated as a pH controller, which is crucial for maintaining the pH level at around 9.0.

3. Results and Discussion

Prior to conducting the droplet combustion experiment, the quality of the coating was assessed through SEM analysis. Additionally, the thermophysical characteristics of the Ni-Al powder were determined using TGA/DSC analysis. In the droplet experiment, the combustion of the nanofuel, and the impact of nickel coating on the combustion of the aluminum nanofuel, were quantitatively measured. To facilitate comparison, two types of nanofuels were prepared: pure Al nanofuel and Ni-Al nanofuel containing 2.5 and 5 wt.%. The initial droplet size was set to 1 ± 0.1 mm. The experiment was replicated at least five times under identical conditions to ensure result consistency. All experiments were conducted in an open atmosphere, with a temperature of approximately 293 K.

It is important to note that the outcomes of droplet combustion exhibited variability across each case due to the inherent heterogeneity of the nanofuel. The results presented capture the representative and overall experimental behavior.

3.1. Thermophysical Characteristics of Ni-Al Powder

The SEM images depicted in Figure 2 display a comparison between Al powder and Ni-Al powder. Notably, an examination of Figure 2b reveals particles that are relatively brighter and more opaque. EDS analysis outcomes, as presented in Table 2, corroborate these observations by confirming the presence of deposited nickel particles. Unlike the conventional approach in micro-sized aluminum coating, where nickel particles uniformly cover the aluminum surface [24], this study demonstrates a distinct pattern: nickel particles sparsely adhere to the aluminum particle surfaces, lacking complete encapsulation.

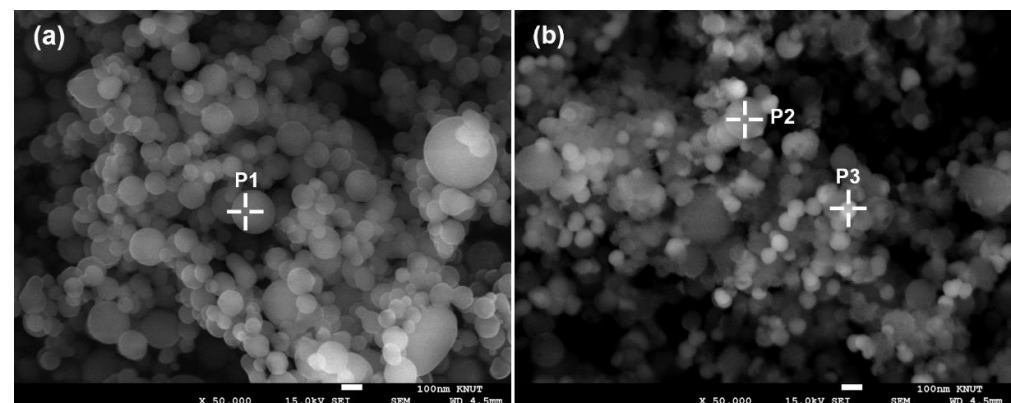


Figure 2. SEM images of pure (a) and nickel-coated (b) aluminum powder.

Table 2. EDS composition analysis of pure and nickel-coated aluminum powder.

Point	O (wt.%)	Al (wt.%)	Ni (wt.%)
P1	13.49	86.51	-
P2	15.39	69.37	15.24
P3	15.22	53.23	31.55

This departure from uniformity in the deposition of nickel arises due to multiple influencing factors within the electroless plating process, including the concentration of the reductant, complexing agent, stabilizer, pH, and temperature. Consequently, the uneven distribution of nickel coating underscores the inapplicability of parameter values employed for micro-sized aluminum coating to the coating of nano-sized aluminum. This accentuates the imperative nature of determining optimal parameter values for the latter.

The thermophysical characteristic analysis through TGA/DSC analysis, as depicted in Figure 3, confirms the occurrence of the sintering reaction in the case of nano-sized Ni-Al powder. When investigating the Al sample, the overall thermophysical characteristics of conventional aluminum powder, encompassing oxide layer growth, phase transitions, melting, heat flow, and weight increase, become distinctly evident as the temperature rises [25]. In contrast, Ni-Al exhibits a distinct behavior compared to Al and micro-sized Ni-Al. Within the range of amorphous oxide layer transition to the γ -phase, the DSC curve displays a rightward slope, accompanied by points of inflection in the weight curve. These phenomena emerge when the sample's temperature temporarily exceeds the furnace-applied temperature. Based on these results and the strong heat generation reaction that could not be observed in the DSC results of Al, it seems that sintering reactions occurred in the Ni-Al powder.

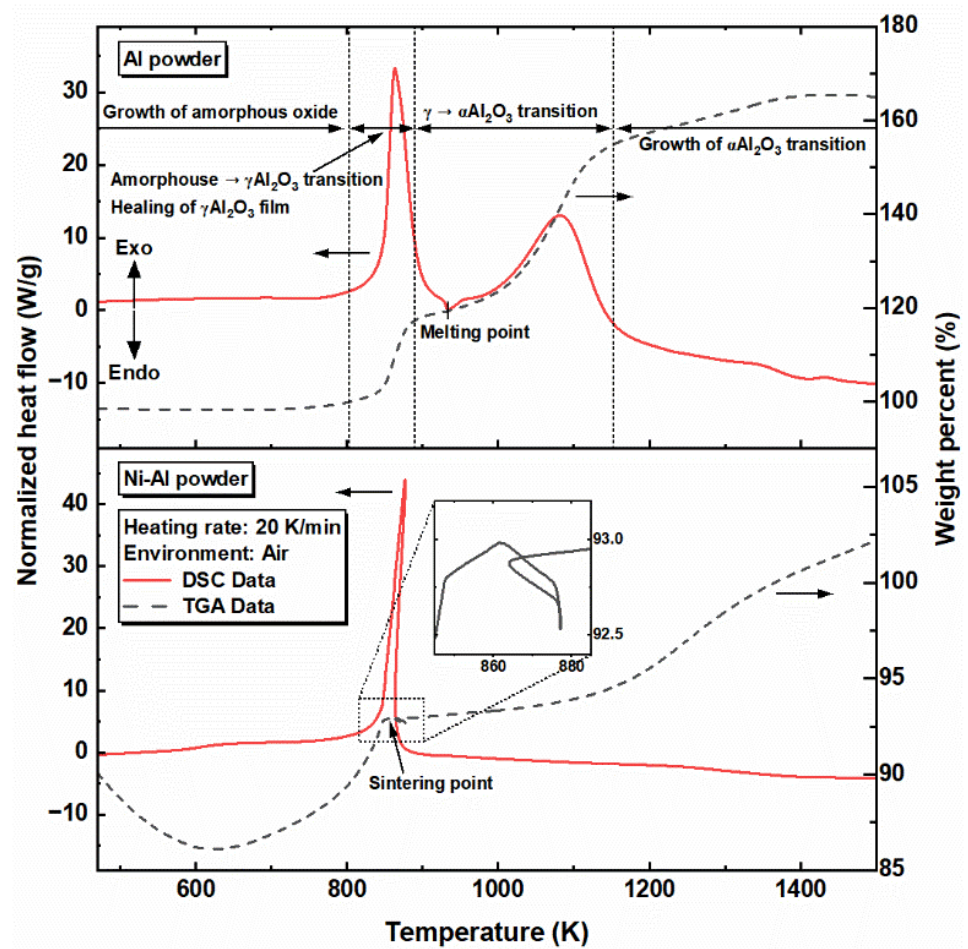


Figure 3. TGA/DSC analysis of nickel-coated aluminum (bottom) powder compared with pure (top) aluminum powder.

Moreover, the more pronounced exothermic reaction in comparison to Al provides further support for these findings. Analyzing the temporal evolution of TGA/DSC results in Figure 4 reveals an instantaneous temperature surge at the onset of the exothermic reaction [26,27]. However, unlike combustion, there is minimal weight increase.

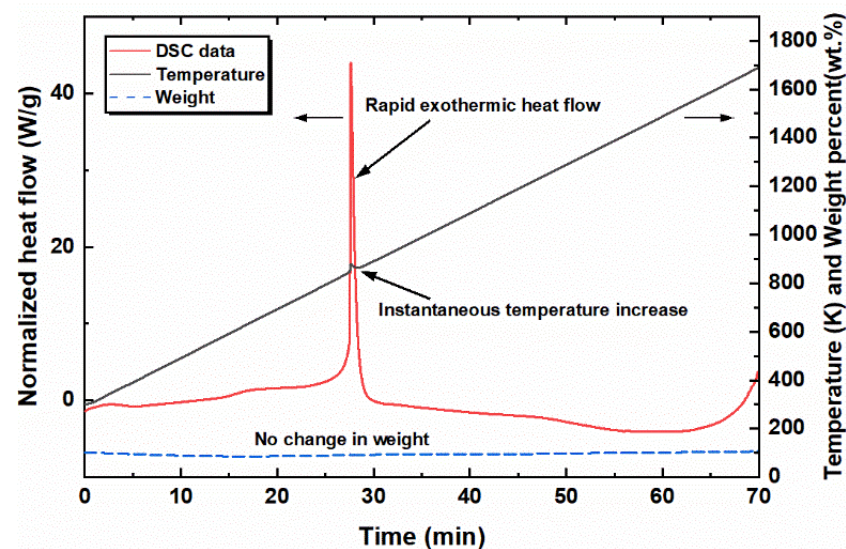


Figure 4. TGA/DSC analysis of nickel-coated aluminum powder depends on time.

3.2. Combustion Characteristics of Nanofuel

The high-speed camera captures the combustion process of nanofuel droplets, as illustrated in Figures 5 and 6. Across all experimental scenarios, the emission of particles resulting from micro-explosions during combustion was observed. Although differences in nanofuel concentration throughout the combustion process were hard to discern, a distinct contrast arose during the extinction phase, particularly influenced by the presence of nickel coating. In the instance of nanofuel containing Al, the combustion of partial residual Al during extinction was noticeable, propelling it away from the droplet. Conversely, in nanofuel containing Ni-Al, most of the residual Al combusted intensely and luminously during extinction.

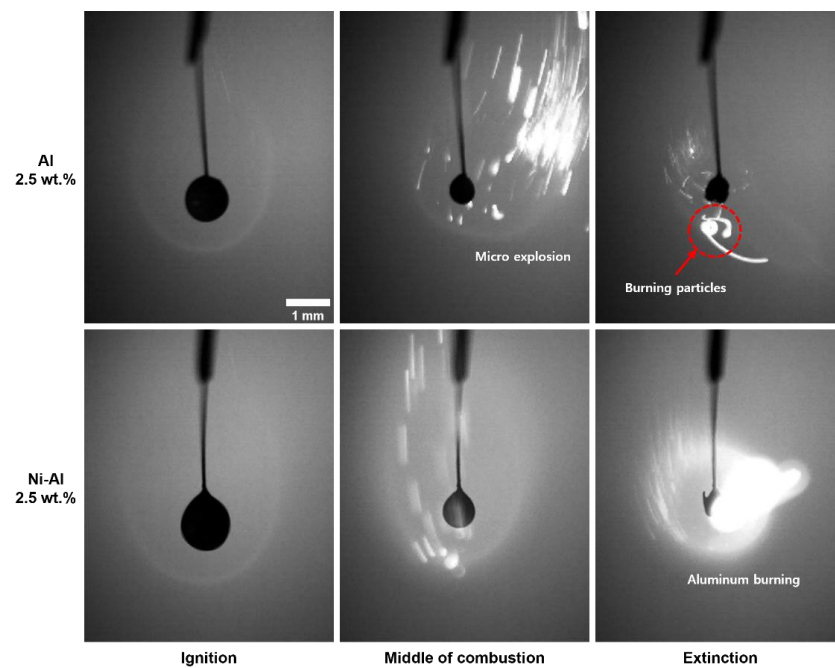


Figure 5. Combustion images of 2.5 wt.% droplet.

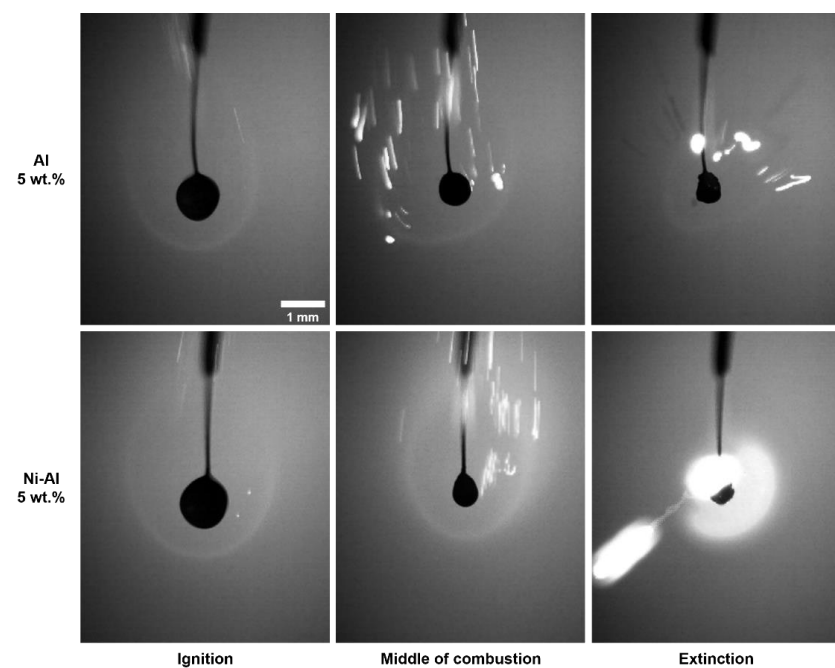


Figure 6. Combustion images of 5 wt.% droplet.

Turning to the depiction in Figure 7, acquired through PMT measurements, a clearer understanding of the nanofuel combustion process emerges. While the high-speed camera images verified the ejection of illuminated Al particles due to micro-explosions, these occurrences were infrequently captured by the PMT results. This implies that the Al particles emitted by micro-explosions were either dispersed while heated without undergoing full combustion, or the quantity of combusting Al particles was too small to be effectively measured by the PMT. Furthermore, mirroring Figures 5 and 6, the termination of combustion highlights that nanofuel containing Ni-Al emits AIO radicals more intensely compared to its Al counterpart, as indicated.

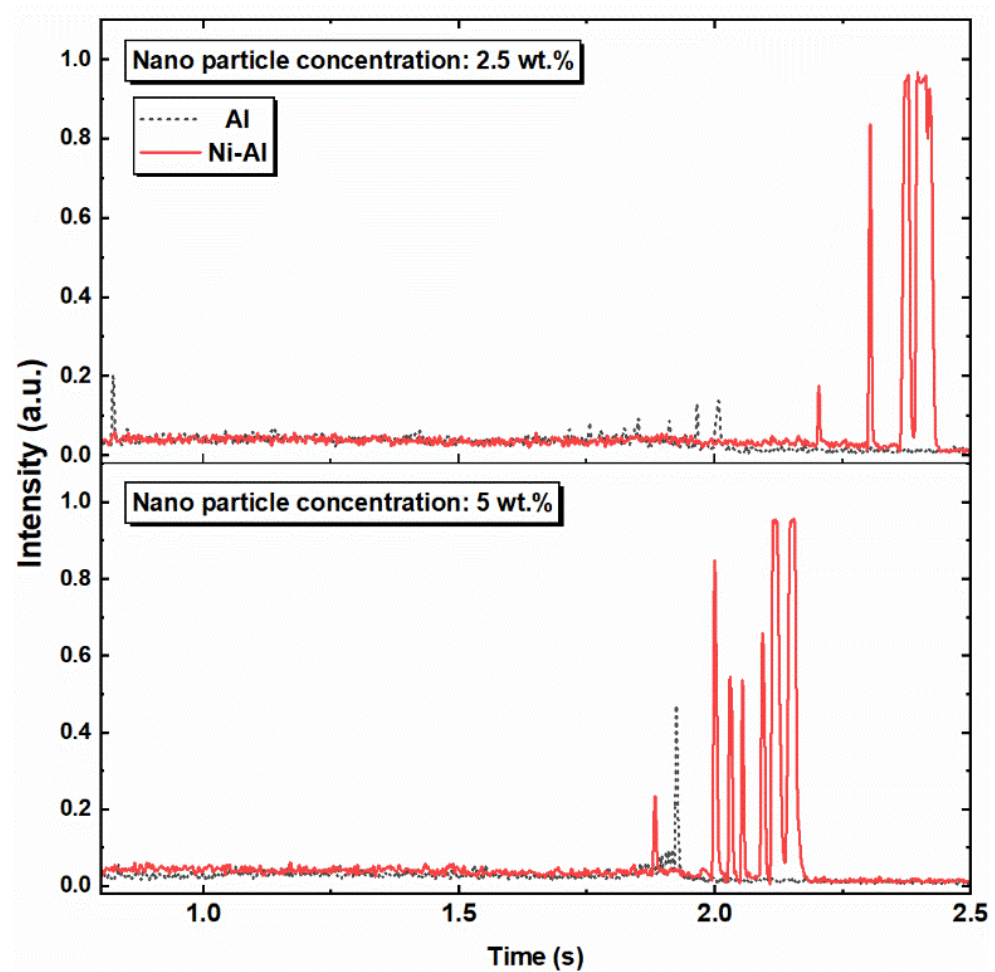


Figure 7. Measurement result of AIO radical emission intensity during combustion of nanofuel droplets.

Based on these observations, it is evident that droplets containing Al powder did not undergo aluminum combustion upon the completion of the combustion process. However, in contrast, droplets containing Ni-Al powder exhibited combustion of residual particles during the final phase of combustion. To substantiate this, the composition of combustion residues adhering to the tungsten wire was scrutinized utilizing SEM/EDS, as illustrated in Figure 8 and elaborated in Table 3. The data presented in Table 3 reveals a notable increase in the weight fraction of oxygen atoms for droplets containing Ni-Al. This enhancement signifies that the particles within the droplets experienced oxidation as a result of the combustion process.

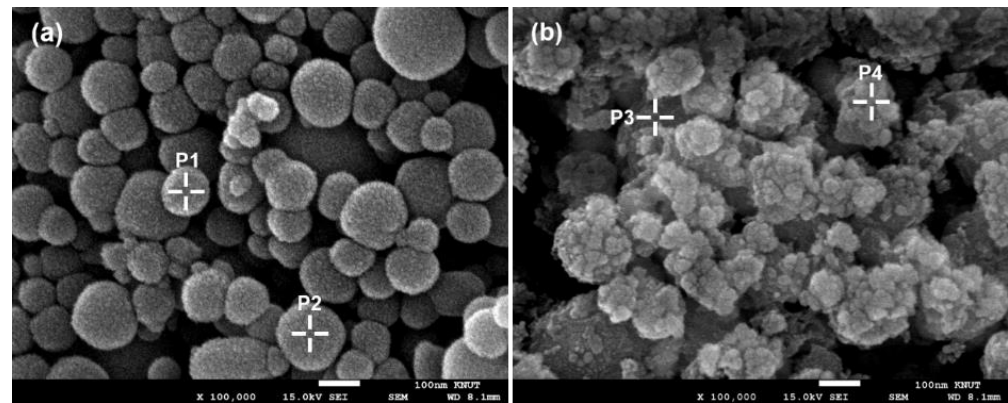


Figure 8. SEM images of combustion residue of pure (a) and nickel-coated (b) aluminum.

Table 3. EDS composition analysis of combustion residue.

Point	O (wt.%)	Al (wt.%)	Ni (wt.%)
P1	11.72	88.28	-
P2	13.60	86.40	-
P3	39.91	45.27	14.82
P4	46.69	43.49	9.82

3.3. Burning Rate of Droplet

Utilizing a high-speed camera, sequential combustion images of the nanofuel droplet were captured to facilitate the analysis of temporal changes in droplet diameter. Subsequent post-processing of these images allowed for the determination of ignition and extinction timings, in addition to the computation of the burning rate. The combustion rate signifies the quantity of liquid burned within a consistent time frame.

The concepts of droplet ignition, extinction timings, and burning rate are visually depicted in Figure 9. Temporal progression is represented along the horizontal axis, while the squared droplet diameter, suitably normalized by the initial squared droplet diameter, is depicted along the vertical axis. This normalization procedure ensures consistent comparison across all experimental scenarios. The equation governing the calculation of the effective burning rate, $K_{b,eff}$, is explicated as follows, with comprehensive elaboration available in the prior work of Kim et al. [28].

$$K_{b,eff} = \frac{d_i^2 - d_e^2}{t_b} \text{ (mm}^2/\text{s)} \quad (2)$$

where $K_{b,eff}$ represents the burning rate of the droplet, d_i indicates the diameter at ignition, d_e represents the diameter at extinction, and t_b denotes the time from ignition to extinction.

The obtained burning rates are graphically depicted in Figure 10. Initially, a direct correlation is observed between the concentration within the droplets and the burning rate, aligning with precedent research findings [1,29]. However, diverging from the earlier outcomes related to thermophysical and combustion characteristics, the burning rate of Ni-Al nanofuel is found to be comparatively slightly lower than that of nanofuel containing Al. It is because the burning rate of droplets is notably influenced by thermal conductivity. Given aluminum's thermal conductivity of approximately 220 W/m·K at 800 K, and nickel's thermal conductivity of around 67.4 W/m·K at 800 K, it can be logically deduced that the thermal conductivity of Ni-Al is inferior to that of Al. However, the variation in burning rates was found to be less than 10%. This allows us to conclude that nickel-coated nanofuel is a promising candidate for addressing the challenge of low ignition in aluminum-based nanofuels.

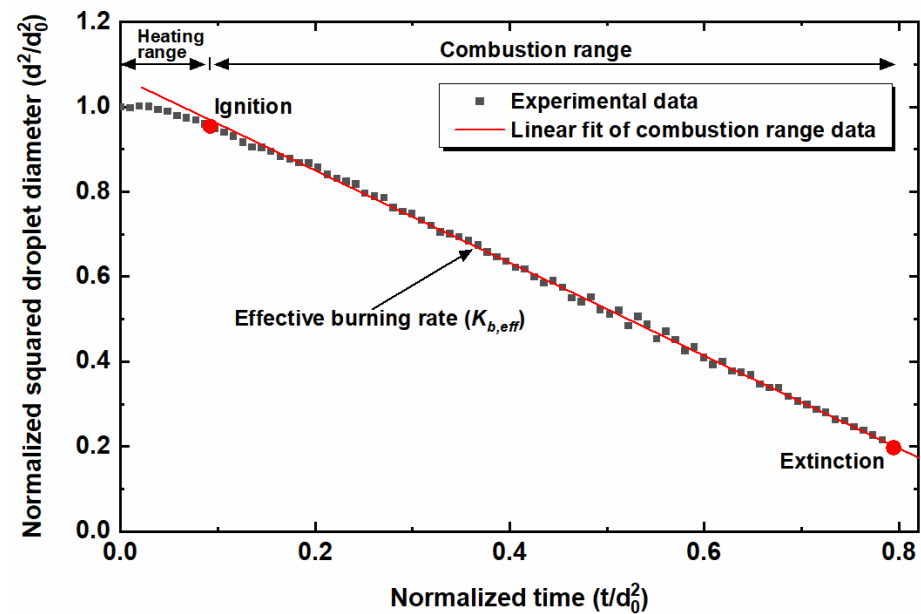


Figure 9. Definition of combustion range and effective burning rate.

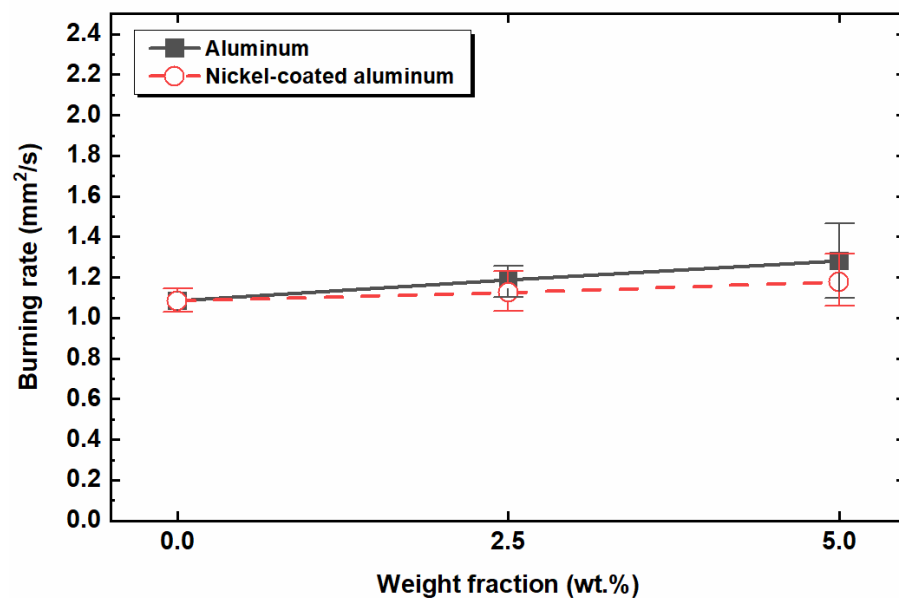


Figure 10. Burning rate calculation in relation to nickel coating presence and concentration.

By deepening our comprehension of energy density and reactivity within nanofuels, this study has the potential to advance the field. Moving forward, it is imperative for forthcoming inquiries to tackle critical aspects such as the optimization of nickel coating uniformity and the evaluation of combustion characteristics across various coating durations. This development could contribute to the advancement of nanofuel commercialization in the field of aerospace propulsion systems, such as rocket and supersonic propulsion.

4. Conclusions

This study presents an investigation into the thermophysical and combustion characteristics, alongside the burning rate, of ethanol-based nanofuels containing Al and Ni-Al components. The findings are summarized as follows:

1. The investigation into nickel coating on aluminum powder unveiled a non-uniform surface deposition of nickel, failing to fully encapsulate aluminum particles. However, SEM/EDS analysis confirmed the occurrence of a sintering reaction between aluminum and nickel;
2. For nanofuels containing Al, unburned aluminum residues persisted post-combustion. Conversely, the introduction of Ni-Al nanofuel exhibited a pronounced AlO signal during the combustion termination phase, with residue analysis corroborating substantial aluminum combustion;
3. Comparatively, nanofuel containing Ni-Al demonstrated a diminished burning rate in contrast to its Al counterpart. This phenomenon can be ascribed to the lower thermal conductivity of nickel in comparison to aluminum, resulting in reduced thermal conductivity of the Ni-Al nanofuel;
4. Considering the overall results, the outcomes suggest that nanofuels containing Ni-Al possess elevated energy potential relative to those with Al, albeit with an expected increase in total combustion duration.

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