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Article

Assessment of Carbon Nanotubes as Ignition Booster Under Dual Fuel Combustion with Hydrogen-Derived Fuels

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Abstract

Dual-fuel combustion is often proposed for diesel engines as a means to partially replace conventional diesel with cleaner and/or more sustainable alternatives, such as those derived from green hydrogen. However, the low reactivity of these fuels (i.e. methane, hydrogen, and ammonia) often leads to prolonged ignition delay and combustion instability. This challenge could potentially be overcome using nanomaterials, which are additives that could improve reactivity and compensate for autoignition deficiencies. Thus, this study evaluates the effect of carbon nanotubes (CNTs) dispersed in diesel fuel on the autoignition process under dual-fuel operation. CNTs were dispersed at a concentration of 100 mg/L and stabilized with surfactant sodium dodecylbenzene sulfonate (SDBS). The resulting nanofuels were then tested in a constant volume combustion chamber (CVCC) using methane, hydrogen, and ammonia as secondary fuels across various energy substitution ratios and temperatures (535 °C, 590 °C and 650 °C). The results show that the impact of CNTs on ID is negligible and highly temperature dependent. At the lowest tested temperature (535 °C) and 40% methane substitution ratio, only slight reductions in ID were obtained. Nevertheless, this effect vanished at higher temperatures (590 °C and 650 °C). Regarding pressure gradient, the addition of CNTs and SDBS generally induced a decrease in pressure peak of up to 15%. This trend is attributed to the trapping of fuel droplets within the CNT structures, which creates a physical barrier that delays vaporization. These findings suggest that the practical benefits of CNTs-SDBS dispersions in diesel engine operating under dual mode with sustainable low reactivity fuels remains limited since the main engine-related phenomena which could be affected, which is autoignition, is not really enhanced.

Keywords: pristine carbon nanotubes; autoignition; dual fuel CI combustion; H₂ carriers

1. Introduction

Globally, there is growing concern about the environmental impact of fossil fuels in sectors such as energy generation, transportation, and industry [1,2]. This raises a need for the transition towards eco-friendly alternatives. Fossil fuel-powered vehicles, particularly those based on diesel fuel, are anticipated to remain prevalent in the transportation sector in the short and midterm, as electrification in heavy-duty applications, including marine and freight transport, faces significant challenges related to cost and limited autonomy [3]. Furthermore, stationary applications (gensets, CHP plants, etc.) are expected to continue being dominated by diesel engines.

To mitigate the environmental impact of diesel engines during this transitional period, advanced combustion technologies, such as dual-fuel operation, have been explored [4]. These technologies aim to substantially replace fossil fuels while preserving performance attributes, including high efficiency and low pollutant emissions. Consequently, gaseous fuels, such as natural gas [5], hydrogen [6,7], ammonia [8], are commonly employed in dual-fuel applications. In fact, dual-fuel engines running with natural gas and even light alcohols (methanol) are common, for example, in the marine sector [9,10]. However, the low autoignition tendency of these secondary fuels requires the use of a more reactive compound, such as a diesel-type fuel, to cause ignition [11].

Furthermore, the use of nanomaterials as fuel additives has been widely reported to present a potential strategy to enhance reactivity and improve autoignition, as well as to improve combustion efficiency in diesel engines [12]. Existing literature has suggested that these materials provide benefits such as increased fuel energy density and volatility [13,14], which would require further exploration since it appears to be not coherent with the nanomaterials concentration (usually below 200 mg/L) and their physical properties. On the contrary, nanomaterials can exhibit large reactive surface area, high thermal conductivity, and some catalytic behavior, these properties being more likely to contribute to faster combustion [15].

Most works dealing with nanomaterials and dual-fuel CI combustion are focused on engine performance and pollutant emissions. Manigandana et al. [16] investigated a dual-mode diesel engine using hydrogen (with a mass substitution percentage of 20%) with different nanomaterials, i.e. TiO_2 , Al_2O_3 , CuO , CeO_2 and CNTs dispersed in the diesel fuel. They found that these compounds increase the burning rate, whereas the use of CNTs reduced the autoignition time, thus lowering the weight of the premixed phase and, consequently, reducing NO_x emissions.

Javed et al. [17] investigated the effects of ZnO nanomaterials in dual-mode combustion in a diesel engine utilizing hydrogen at flows of 0.5 and 1.5 L/min. The presence of ZnO demonstrated a 28.9% reduction in NO_x emissions. However, this effect diminished as the hydrogen flow rate increased. For instance, at a flow rate of 1.5 L/min, NO_x emissions increased, and the ZnO effect was found to be negligible. Furthermore, hydrogen addition led to an average 35% and 30% increase in CO and HC emissions, respectively, primarily due to oxygen displacement. Conversely, the utilization of ZnO nanomaterials resulted in a 40-45% reduction in HC emissions. In another study, Javed et al. [18] observed that a compression ignition engine fueled with biodiesel blended with nanomaterials and hydrogen exhibited superior performance and reduced pollutant emissions and noise levels.

Feroskhan et al. [19] tested a diesel engine utilizing nanofuels based on cerium oxide (CeO_2) under dual-mode with biogas at energy ratios ranging from 10% to 75%. They concluded that the results were primarily influenced by the biogas effect, while nanomaterials exhibiting a marginal effect. Pugazhendhi et al. [20] investigated the effects of nanofuels dispersed in diesel fuel-castor oil at concentrations of 50 and 100 mg/L and, under dual-mode with ammonia at flow rates of 10 L/min. Their findings revealed that while ammonia reduced combustion efficiency, the inclusion of nanomaterials improved the combustion process and led to a decrease in pollutant emissions such as CO, HC, smoke opacity and NO_x . Similarly, Arslan et al. [21] reported in an experimental work involving CNTs in dual-mode combustion with natural gas that CNTs counteracted the increase in CO and hydrocarbons caused by natural gas slipping.

As mentioned above, the previously commented works deal with engine tests, the combustion process being inherently very complex, and highly affected by the piston movement, turbulence level and other phenomena such as wall heat transfer and jets interactions [22]. Furthermore, the inherent dependence of diesel engines performance and pollutant emissions on operational parameters including injection strategy, exhaust gas recirculation (EGR) rate, intake pressure and temperature could potentially mask the true impact of fuel additives, such as nanomaterials. For this reason, phenomenological investigations utilizing specific combustion devices are necessary to isolate nanomaterial-derived phenomena and elucidate the underlying physics and chemistry involved. For dual-fuel combustion, researchers have employed, among others, constant volume combustion

chambers (CVCC). The CVCC, although commonly used to determine the derived cetane number (DCN) of diesel-type fuels and thus quantify autoignition quality [23,24], facilitates the analysis of autoignition through the determination of parameters such as multi-stage ignition, heat release rate, and pressure evolution. These measurements are typically obtained using appropriate combustion diagnostics models [11,25].

Conversely, Mei et al. [16] investigated the effects of CNTs and cerium oxide (CeO₂) on the evaporation of diesel fuel. They utilized a constant-volume and quasi-steady evaporation system, conducting experiments at controlled temperatures of 673 and 973 K. The evaporation process was examined using a high-speed camera optical system. The results indicated that nanomaterials acted as heterogeneous nucleation sites, enhancing fuel droplet expansion and increasing the frequency of microexplosions, thereby facilitating overall fuel droplet evaporation. On the contrary, Zhang et al. [26] reported that nanomaterials can agglomerate during the evaporation stage. While this phenomenon facilitates radiation absorption and promotes microexplosions, it inhibits the diffusion of superheated vapor.

In a similar study, Jiang, et al. [27] studied the effect of CeO₂ nanomaterials on the evaporation process of Jatropha methyl ester-diesel fuel blends at concentrations of 0.05, 1.0 and 2.0 wt.% and temperatures of 873 and 973 K. Their findings revealed that concentrations of 0.05 wt.% promoted heat absorption within the droplet, consequently increasing the evaporation rate. On the contrary, a higher concentration (2.0 wt%.) of nanomaterials exhibited an inhibitory effect on droplet evaporation, leading to an increase in the evaporation time of the droplet, which is possibly caused by the reduced heat transfer rate resulting from large agglomerates behaving as thermal resistance. Similarly, Wang et al. [28] observed that the use of nanomaterials can lead to an increase in the frequency of microexplosions during the evaporation of the fuel droplet. However, they also noted that nanomaterials can accumulate on the surface of the droplet, eventually forming a porous and spherical shell. This shell can act as a barrier, hindering the evaporation of the fuel droplet.

Table 1 summarizes phenomenological studies exploring the effects of nanomaterials as additives to diesel fuel, demonstrating their ability to enhance both evaporation and combustion under controlled operation conditions such as droplet combustion. The nanomaterials are supposed to serve as nucleation sites, promoting the vaporization of adjacent diesel molecules and improving overall evaporation rates. These enhancements may eventually lead to more efficient combustion and lower emissions in real applications. Nevertheless, some reports indicate negative effects from nanomaterials use, particularly at high relative concentrations, such as limitations in evaporation rate, radiation absorption, and vapor diffusion. Anyway, defining high and low concentrations is always arbitrary, as these parameters depend on the kind of nanomaterial, base fuel, and stabilization method, among other factors. Consequently, there is currently no consensus on preparation methods for nanofuels, and well-stablished procedures have yet to emerge. Regarding the benefits of nanomaterials on engine emissions and performance, there is still no consensus. Although the previously mentioned literature review provided in general some benefits, recent research by the authors [29] demonstrated that the use of carbon nanotubes (CNTs) dispersed in diesel fuel did not improve pollutant emissions in a diesel engine operating under real driving conditions. In fact, a technical challenge was identified, specifically, the retention of the nanomaterials in the fuel filter and the release of unburned CNTs in the exhaust gas

Table 1. Effect of nanomaterials on the combustion characteristics of diesel fuel.

Base fuel	Nanoparticle	Concentration	Temperature	Positive effect reported	Negative effect reported-	References
Diesel/ biodiesel	CNTs CeO ₂	At different blends	673 K 873 K	Nanomaterials Accelerate droplet evaporation	CNTs aggregate more easily, and the evaporation process is prolonged	[30]

Diesel	CeO ₂	5 wt %.	673 K 873 K	CeO ₂ accelerates the droplet evaporation and promotes microexplosions	Not reported	[31]
Ethanol	Graphene oxide (GO)	0.1 wt %.	Room temperature	GO enhances the burning rate and promotes microexplosions	GO increases the ignition delay	[32]
Diesel/ n-butanol	CQDs	95 mg/L	Room temperature	CQDs enhance fuel evaporation.	CQDs lead to an increase in the ignition delay	[33]
Crude vegetable oils	Nanocarbon	5 mg/L		Nanocarbon increases fuel reactivity, droplet temperature, and shortens ignition delay.		[34]
2-methylfuran/ diesel	CNTs	25 mg/L, 50 mg/L, and 100 mg/L		CNTs decreases fuel ignition delay and increases combustion rate	CNTs increases micro-explosions intensity	[35]
Diesel	Al ₂ O ₃ and carbon nanomaterials	0.05 – 3.0 wt%	Room temperature	Nanomaterials Al ₂ O ₃ improves microexplosions	The aggregation of particles limits the diffusion of vapor within the droplet.	[26]
n-decane and ethanol	Boron and Iron	0.5 wt% and 5 wt%	Room temperature	The nanomaterials facilitated ignition and increased the burning rate of base fuels	At high particle loading rates, a large agglomerate will form. The large agglomerate may not be ignited, hindering the combustion process of the base fuel	[36]

In this regard, the existing literature reveals a limited research landscape. On one hand, phenomenological studies using well-controlled combustion devices have explored the effects of nanomaterials, but these investigations have been limited to diesel and alternative liquid fuels under conventional diesel operation. On the other hand, research into nanomaterials for dual-fuel operation has been conducted only under engine conditions. However, in such systems the fundamental chemical effects of the additives are often complex and masked by physical phenomena, such as turbulence, injection dynamics, and heat transfer.

The lack of knowledge about the potential of nanomaterials for engine applications is still significant. This gap is especially relevant in a context in which the importance of H₂ and H₂-carriers (such as ammonia and synthetic natural gas), commonly known as e-fuels, continues to grow [37,38]. Therefore, the current research is oriented towards evaluating whether CNTs can induce a positive effect on autoignition under dual-fuel combustion. Furthermore, it seeks to determine whether this effect is consistent when employing low-reactivity gaseous fuels such as hydrogen, methane, and ammonia. Specifically, autoignition within a CVCC is evaluated, varying the energy substitution percentage of the secondary fuels and the combustion chamber temperature. This fundamental approach will contribute to evaluating the technical feasibility of CNTs in dual-mode combustion as a means to overcome the challenging ignition phenomena in dual-fuel compression-ignition engines fueled with promising hydrogen-derived compound.

2. Methodology

2.1. Nanofuels Preparation

The methodology for nanofuels preparation followed the procedures outlined in previous research conducted by Gallego, et al. [39]. To avoid redundancy, the characterization and stability analysis of the nanofluids are not included in this work. This study utilized pristine carbon nanotubes (CNTs) with a purity of 95% and diameters ranging from approximately 17 to 75 nm, with an average 40 nm diameter obtained from SkySpring Nanomaterials, Inc. The CNTs were dispersed at a concentration of 100 mg/L in commercial diesel fuel supplied by Repsol, Spain. For nanofluid preparation, sodium dodecylbenzene sulfonate (SDBS), an anionic surfactant obtained from Sigma-Aldrich Pty. Ltd., was employed at a constant 200 mg/L concentration.

The two-step method was employed for nanofluid preparation. Firstly, the required chemicals (CNTs and SDBS) were accurately weighed using a high-precision Sartorius M5P analytical balance. Subsequently, the SDBS was blended with diesel fuel and placed in an 1800 ultrasonic bath for 15 minutes. In the second step, CNTs were added to the SDBS-diesel fuel mixture, followed by sonication using a Hielscher UP200s ultrasonic probe operating at 60% amplitude for one hour. The sonication process aimed to break agglomerates of CNTs and ensure uniform dispersion of both the nanomaterials and surfactants within the diesel fuel. Table 2 shows the main properties of the diesel fuel and the resulting fuels, where “D” and “S” denote diesel fuel and SDBS, respectively.

The selection of CNTs and SDBS concentrations for this study were based on the criteria established by Gallego et al. [29]. Their findings indicated that a 100 mg/L dispersion of CNTs in diesel fuel stabilized with SDBS maintains a high degree of stability over time. This mitigates the agglomeration and sedimentation of nanomaterials, which could otherwise lead to operational problems within the experimental apparatus used (Cetane ID 510 of Herzog-PAC), which incorporates a conventional fuel injection system, including a fuel filter, a high-pressure pump, and a Bosch common rail injector, as detailed in the following section.

Table 2. Effect of nanomaterials on the combustion characteristics of diesel fuel.

Property	Unit	Diesel fuel (D)	D+SDBS (S)	D+S+CNTs 100 mg/L	Standard
Kinematic viscosity at 40 °C	cSt	2.399	2.7182	2.426	EN ISO 3104
Derived Cetane number	-	51.98	50.82	52.12	ASTM D7668/EN 16715
Density at 15 °C	kg/m³	837	837.5	838.0	EN ISO 3675
Lower heating value	MJ/kg	43.2	42.0	43.7	EN 12937
Smoke point	mm	19.75	18.77	20.74	EN 3014
Wear Scar diameter (WSD)	µm	187.5	199.5	296.4	ISO 6508-1

2.2. Autoignition Tests

A Herzog-PAC Cetane ID 510 equipment was utilized to perform the autoignition assessment. This equipment has a Constant Volume Combustion Chamber (CVCC). It allows to determine the Derived Cetane Number (DCN) of diesel-type fuels, following ASTM D7668 and EN 16715 standards. The equipment comprises a 0.423 L CVCC, an external heating source, intake and exhaust valves, a fuel delivery system, and temperature and pressure sensors. Additionally, the Cetane ID 510 includes a Bosch fuel injection system with common rail technology, which allows injection pressures up to 1500 bar (in the case of the current study, the pressure was adjusted at 1000 bar). This device performs 20 consecutive fuel injections within a set period, generating pressure-time curves. To ensure data reliability, the initial five tests are discarded to eliminate any potential influence from residual traces of previously tested fuels.

The combustion chamber was supplied with synthetic air composed of 79% nitrogen (N₂) and 21% oxygen (O₂). In addition, dual-mode combustion tests were conducted using synthetic gas bottles containing air and a prescribed amount of low-reactivity fuel to attain the target energy ratio. The

secondary fuels tested were methane (CH₄), hydrogen (H₂), and ammonia (NH₃), following the method described in [11,24]. Accordingly, when secondary fuels were introduced, the diesel injection duration was reduced to offset the energy supplied by the secondary fuel, aiming to maintain constant energy delivery. This approach, also used in [11,24], allows for a fair comparison when these fuels are used in real engines operating under dual mode, by keeping the total energy supply constant, the engine power remains comparable, subject only to expected minor differences in thermal efficiency.

The experimental parameters outlined in **Error! Reference source not found.** were considered to evaluate the influence of CNTs dispersions on diesel fuel. In addition, three temperatures were selected. The first one, 590 °C, is the value used to determine the DCN according to the mentioned standards, and 535 °C and 650 °C are useful to observe the effect of temperature, providing indicative values of potential temperature at the end of the compression stroke of a compression ignition engine. Percentage energy substitution ratios of 10%, 20% and 40% were used. However, the synthetic H₂/air mixture with 40% of hydrogen (energy content) was not tested, as the supplier does not provide it for safety-related reasons. Furthermore, testing with NH₃ was limited to an energy substitution ratio of 40%. This decision was based on the premise that a lower concentration, due to the poor combustion performance of ammonia, would not allow for highlighting the role of this fuel on autoignition.

Table 3. CVCC operating conditions.

Parameter	Level 1	Level 2	Level 3
Fuel blends	Diesel	Diesel and SDBS at 200 mg/L	Diesel, SDBS and CNTs at 100 mg/L
Tested temperature (°C)	535	590	650
Initial absolute pressure (bar)	21		
Synthetic air (%Vol.)	21% O ₂ and 79% N ₂		
Fuel injection pressure (bar)	1000		
Injection time (μs)	2500		

The energy substitution percentage (E_{gas}) is calculated through equation (1), where m_{gas} is the total mass of injected gaseous fuel (kg), LHV_{gas} is the lower heating value of the gaseous fuel (MJ/kg), m_{fuel} is the total mass of injected diesel fuel (kg), and LHV_{fuel} its lower heating value (MJ/kg).

$$E_{gas} = \frac{m_{gas} \times LHV_{gas}}{m_{gas} \times LHV_{gas} + m_{fuel} \times LHV_{fuel}} \times 100 \quad (1)$$

Finally, the autoignition process exhibits two distinct stages, each associated with a specific ignition delay time (ID). Following the criteria established by Lapuerta et al. [40], the ID was defined as the time interval between the start of fuel injection and the initiation of the main combustion phase. This initiation was determined by the intersection of zero with the secant line connecting one-half and one-quarter of the maximum pressure derivative (dp/dt). An example of ID is shown in Figure 1, where diesel fuel results are displayed at 535 °C.

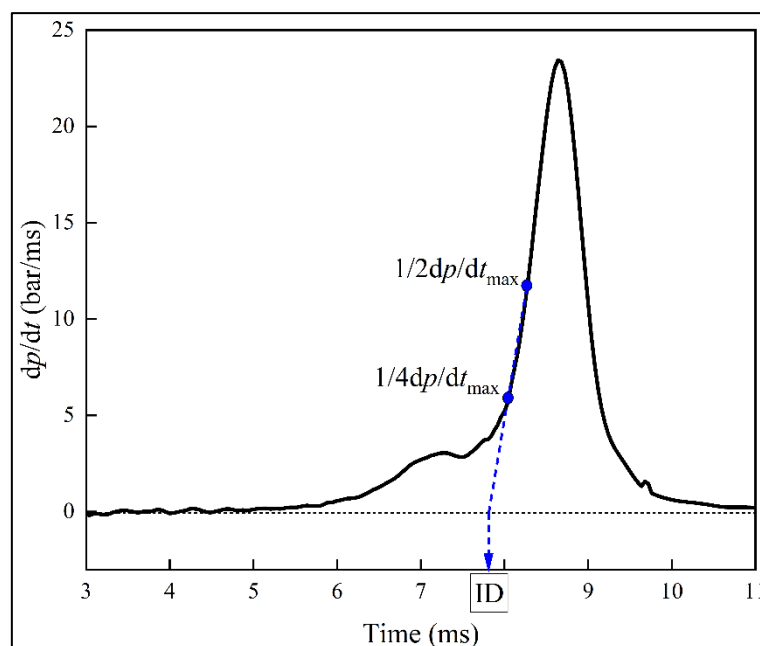


Figure 1. ID definition obtained from diesel fuel tests at 535 °C.

3. Results and Discussion

3.1. Effect of CNTs on the Ignition Delay

Figure 2 show the ignition delay (ID) results obtained for the three low-reactivity fuels used. Each graph represents the average of 15 replicates, with 95% confidence intervals determined using Student's t-distribution. The deviations within these confidence intervals are negligible compared to the observed variations in ID values, thereby confirming the significance of the trends discussed herein.

In general, ID values decrease when temperature increases, a trend consistent with the model by Henein and Bolt [41] based on the Arrhenius equation. At the lowest tested temperature (535 °C, Figure 2a), dual-fuel operation with methane and ammonia led to a marginal reduction in ID. While these low-reactivity fuels typically delay ignition by inhibiting chemical kinetics, and ultra-lean local mixtures as a consequence of a very enlarged mixing period [24], the observed results suggest that such effect is offset by changes in the thermodynamic properties of the charge. Specifically, the addition of these gases modifies the heat capacity of the mixture, which can slightly accelerate the physical delay period. Conversely, hydrogen at a 20% energy substitution ratio increased the ID, suggesting that under these conditions, hydrogen exerts a limited chemical-kinetic effect on ignition reactions, delaying the start of combustion relative to the diesel baseline [42].

Regarding the addition of CNTs at 535 °C, the overlapping error bars with the baseline indicate high uncertainty, suggesting that these differences may not be statistically significant. Nonetheless, the trends observed for the diesel-CNTs blend and methane at the largest replacement (40%) show a slight shortening of the ID, implying a localized benefit that disappears under other fuels and energy substitution ratios.

Furthermore, Figure 2b and Figure 2c demonstrate that CNTs does not enhance ignition delay process at 590 °C and 650 °C. At these higher temperatures, the accelerated chemical kinetics diminish any potential influence of the nanomaterials. Although CNTs possess a high specific surface area, their slow decomposition rate allows them to act as a heat sink during the pre-ignition phase. This thermal energy absorption locally reduces the temperature, counteracting any effects the CNTs might otherwise exert on the ID [43].

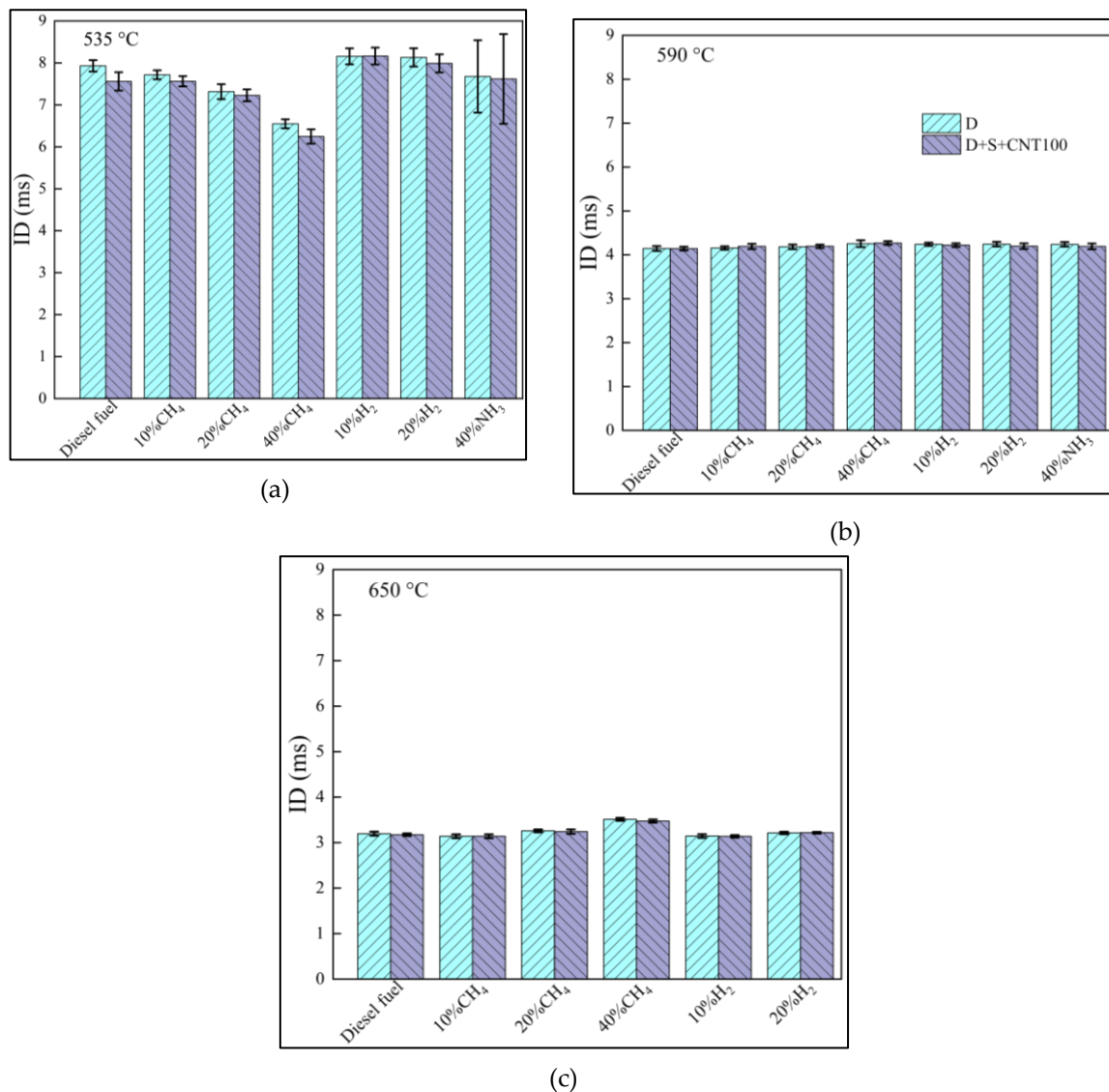


Figure 2. Ignition delay for dual-mode combustion with methane, hydrogen and ammonia at 535 °C (a), 590 °C (b) and 650 °C (c).

3.2. Effect of CNTs on the Combustion Rate

Figure 3 shows the instantaneous pressure gradient for methane at different temperatures, which for practical purposes, is directly proportional to the heat release rate (HRR), given the constant volume of the device (CVCC). The figure depicts the first stage in which pressure gradually increases due to a slow heat release, in the case of diesel fuel, particularly with the lowest input temperature (535 °C). This phenomenon is induced by low-temperature chemical reactions of the long alkyl chains found in diesel fuel (LTHR, low temperature heat release). Once the temperature reaches a threshold value, the main combustion stage starts [44].

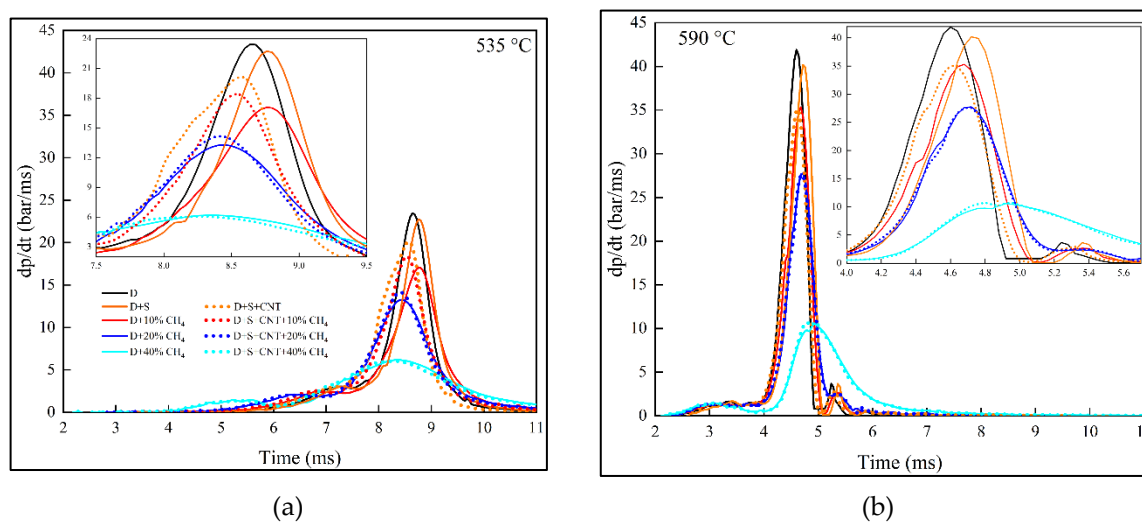
Additionally, Figure 3 shows that the use of SDBS surfactant resulted in a reduction of approximately 3% of the pressure peak with respect to diesel at 535 °C, 590 °C and 650 °C. Furthermore, at the same temperatures, using a blend of diesel, SDBS and CNTs, the pressure peak showed an additional decrease of 15% and 12% compared to both diesel and diesel-SDBS blends, respectively. Although literature reports generally suggest an increase in peak pressure due to shorter ignition delays and faster combustion rates when nanofluids are used [45], the results indicate a contrary tendency, since the ignition delay is shortened negligibly (as is shown in Figure 2) and, pressure peak is reduced.

This phenomenon is attributed, first, to the relatively slow decomposition process of the CNTs during combustion and, second, to CNTs can limit the fuel evaporation process, which occurs because fuel droplets become trapped within the CNTs. Consequently, the liquid fuel surrounding the CNTs is the first to absorb heat and evaporate. Subsequently, the trapped liquid fuel must absorb heat from the hot walls before vaporizing. Furthermore, the CNTs possess a thick, multi-walled structure and are heated more slowly than the surrounding fuel. This delay prolongs the evaporation process, increasing the quantity of premixed fuel and thus elevating the chamber pressure [46].

Despite the negative effect of CNTs described above for just diesel fuel, their incorporation also resulted in increases of pressure peak when methane is used in dual combustion mode. Figure 3a demonstrates a rise in the pressure peak (around 8%) and in the burning rate, which was evident at 535 °C for blends of diesel fuel, SDBS, CNTs and methane at energy percentage substitution of 10%. This is attributed to the prolonged fuel evaporation time, which increases the amount of premixed fuel. Consequently, the increased availability of the reactive mixture raises the in-cylinder pressure (particularly in the presence of methane) thereby offsetting the initial effect [1].

For instance, Arslan, et al. [21], employed a diesel engine working in dual mode and reported a 2% increase in pressure relative to base fuel when a blend of diesel fuel, CNTs at 50 mg/L, and natural gas was used. This result was achieved using replacement percentages between 24% and 32%. However, Atelge et al. [47] state that when utilizing diesel fuel, CNTs at concentrations of 30, 60 and 90 mg/L and biogas with energy replacement percentages between 7.1% and 8.7%, the pressure peak progressively could decrease as the concentration of CNTs increases. They showed that, in a diesel engine running in dual combustion mode, the CNTs chemical shorted the premixed burning phase at full load.

On the contrary, Figure 3 b and Figure 3c reveal a negligible effect of the nanomaterials on combustion behavior at 590 °C and 650 °C, respectively, despite the use or not of dual-fuel conditions.



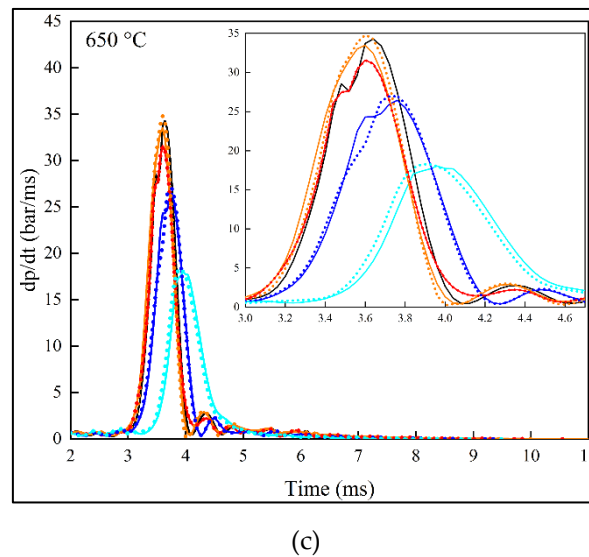


Figure 3. Instantaneous pressure gradient curves (dp/dt) with carbon nanotubes with methane at 535 °C (a), 590 °C (b) and 650 °C (c).

Besides, Figure 4 shows the instantaneous pressure gradient profiles for combustion utilizing CNTs in dual-mode combustion with hydrogen at energy substitution percentages of 10% and 20% across different temperatures. Similar trends observed with methane were achieved, characterized by a reduction in peak pressure with increasing hydrogen energy substitution levels. This phenomenon can be attributed to pressure losses resulting from the displacement of oxygen by hydrogen within the combustion chamber, subsequently diminishing the availability of oxidants for the combustion process leading to unburnt hydrogen [48]. Peak pressures were generally higher with hydrogen compared to methane, primarily attributed to the wider flammability limits [8] and enhanced mass diffusivity, which facilitates improved gas entrainment into the liquid fuel jet [11]. However, as illustrated in Figure 4c, at 650°C the use of hydrogen results in pressure curves that might reach levels close to diesel fuel pressure, except at a 20% energy substitution percentage.

The incorporation of CNTs resulted in marginal enhancements to the pressure profiles at 590 °C and 650 °C, as is shown in Figure 4b and Figure 4c, respectively. Nevertheless, in the case of 535 °C, pressure peak rise of 6% was obtained at 10%, and 20% energy substitution percentage. This phenomenon is attributed to the ID reported above in Figure 2. Furthermore, Manigandan, et al. [16] using dual hydrogen-diesel engine operation, proposed that the inclusion of CNTs may induce higher peak pressures due to their high thermal conductivity, which promotes heat transfer within the combustion chamber. Similarly, Manigandan et al. [16] reported that CNTs dispersed in diesel fuel, when used in dual hydrogen-diesel engine operation, can lead to earlier ignition delay and higher cylinder peak pressures compared to pure diesel fuel. They attributed this to the increased heat dispersion generated by CNTs, further augmented by the potential increase in supplied OH radicals when using hydrogen.

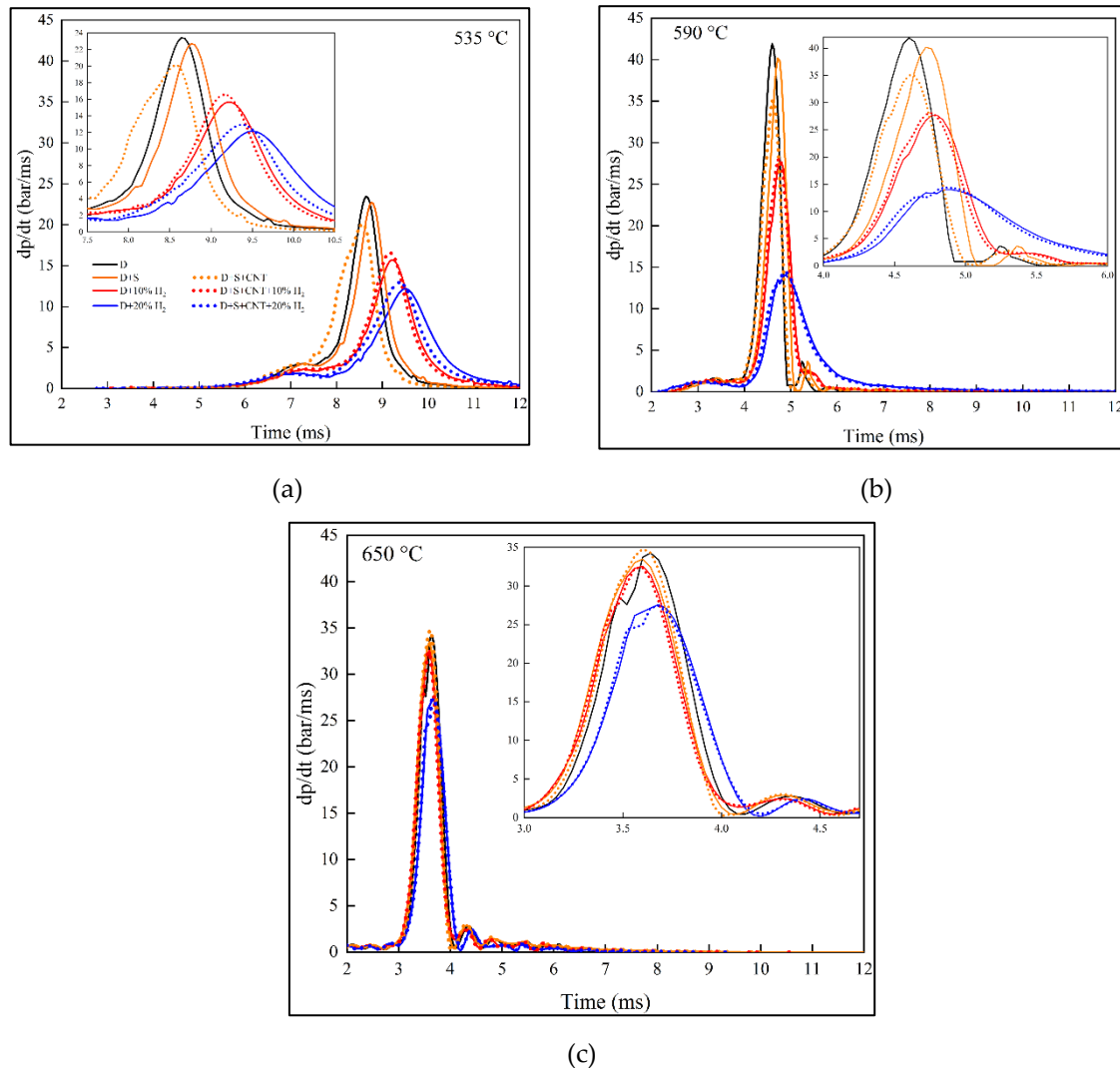


Figure 4. Instantaneous pressure gradient curves (dp/dt) with carbon nanotubes and hydrogen at 535 °C (a), 590 °C (b) and 650 °C (c).

Moreover, **Error! Reference source not found.** illustrates the instantaneous pressure gradient profiles for combustion utilizing CNTs in dual-mode combustion with ammonia at a 40% energy substitution percentage and at 535 °C and 590 °C. Ammonia was selected as a carbon-free energy carrier and a high-density hydrogen vector, owing to its advantages in storage, distribution, and infrastructure compared to hydrogen [16]. As demonstrated in **Error! Reference source not found.**, the introduction of ammonia significantly reduced the pressure within the combustion chamber. This reduction is attributed to the slow flame propagation speed and low chemical reactivity of ammonia. Furthermore, the hydrogen abstraction reaction of ammonia ($NH_3 + OH = NH_2 + H_2O$) inhibits ignition by consuming a substantial number of OH radicals [49]. This occurs despite the improvements in ID reported in **Error! Reference source not found.**. However, this limitation cannot be addressed by introducing CNTs into diesel fuel stabilized with SDBS, as the observed pressure profiles did not exhibit any significant enhancements. Therefore, the characteristics associated with the use of CNTs, such as catalytic activity, improved combustion properties, better mixing of the air-fuel ratio, and their high surface-to-volume ratio [50], failed to yield the anticipated results.

Despite the intent to enhance the combustion process through this additive, the outcomes were not as expected. This discrepancy may be attributed to the possibility that, under specific conditions, nanomaterials cannot effectively participate in the combustion process due to their inherent structure [51], furthermore, under the poor combustion conditions achieved in this study, CNTs failed to demonstrate the reported advantages documented in the literature. Additionally, research by

Pugazhendhi et al. [20] and Wu et al. [52] using NH_3 as the secondary fuel, suggests that nanomaterials could improve the combustion process by increasing parameters such as cetane index, heating value, and thermal conductivity of the fuel in dual-fuel combustion with ammonia. However, the properties presented in **Error! Reference source not found.** and the results discussed in this section indicate negligible or negative effects

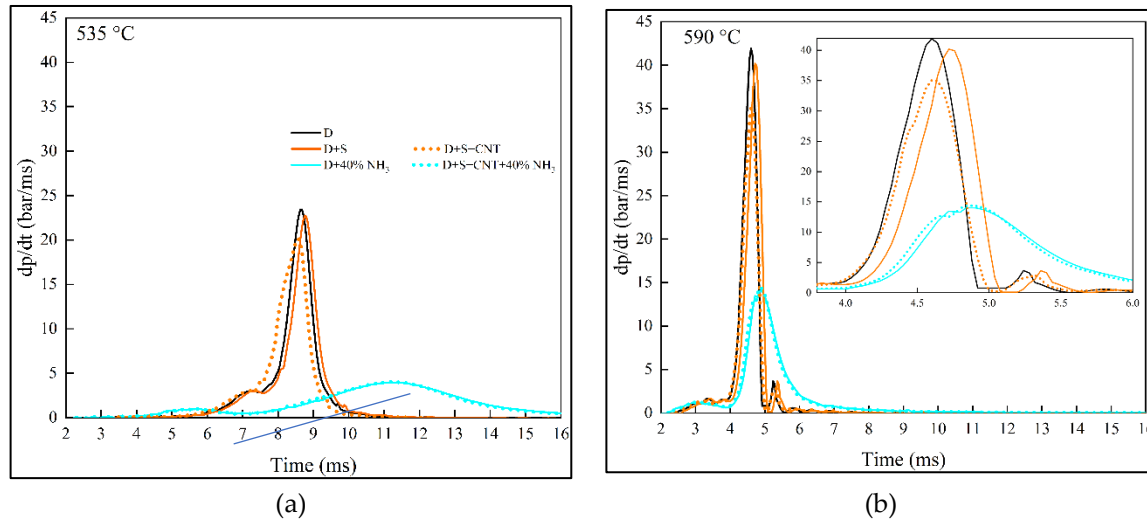


Figure 5. Instantaneous pressure gradient curves (dp/dt) with carbon nanotubes and ammonia at 535 °C (a) and 590 °C (b).

Thus, these findings prove that CNTs do not provide substantial benefits at elevated temperatures, and their positive effects at lower temperatures are minimal. This may raise concerns about the results when used in dual-mode diesel engines operating with methane, hydrogen or ammonia given that the temperatures attained in an engine are considerably higher than those achieved in the constant volume combustion chamber (CVCC) used in this study, which suggests that the practical significance of CNTs in conventional compression ignition engines may be limited [29].

5. Conclusions

This study addresses a gap in dual-fuel combustion involving hydrogen and hydrogen carriers, such as ammonia and methane, specifically focusing on their inherent low reactivity. Based on existing literature, the dispersion of nanomaterials, particularly carbon nanotubes (CNTs), could mitigate the low reactivity characteristic of these gaseous fuels. Consequently, this fundamental research approached the autoignition characteristics of CNTs-dispersed diesel fuels within a constant volume combustion chamber (CVCC), focusing on dual-fuel operation with hydrogen (H_2), methane (CH_4), and ammonia (NH_3) across a range of energy substitution ratios and operating temperatures.

The addition of CNTs demonstrated a highly temperature-dependent behavior. At the lowest tested temperature (535 °C), a reduction in ignition delay (ID) was observed for methane at a 40% substitution ratio. However, this benefit disappeared at higher temperatures (590 °C and 650 °C). This phenomenon is attributed to the fact that, at elevated temperatures, accelerated chemical kinetics counteract the potential influence of nanomaterials. Furthermore, the slow decomposition rate of CNTs allows them to act as a heat sink during the pre-ignition phase, locally absorbing thermal energy and counteracting the potential effects of the CNTs.

The addition of CNTs negatively impacted combustion performance, evidenced by a reduction in peak cylinder pressure of up to 15% relative to diesel. This behavior confirms that the multi-walled structure of CNTs creates a physical barrier that entraps liquid droplets, thereby impeding fuel vaporization, an effect further increased by the independent pressure reduction contributed by the SDBS surfactant. While a minor performance recovery was observed with methane at 535 °C due to

enhanced thermal diffusivity, CNTs proved ineffective in enhancing the combustion of H₂ and NH₃. Specifically, regarding ammonia, the dominant inhibitory effect of its chemical kinetics could not be overcome, as the CNTs demonstrated insufficient catalytic activity to counteract this barrier.

While previous studies have highlighted the potential benefits of CNTs in diesel engines, such as enhanced heat transfer and higher surface area, our findings indicate no significant positive impact on autoignition parameters or pressure gradient under the conditions studied. These results suggest that the practical application of CNTs-SDBS in diesel engines may not be viable due to their limited influence on combustion. This conclusion is further supported by our previous publication, which showed limited viability and emission drawbacks.

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Data Availability Statement: We encourage all authors of articles published in MDPI journals to share their research data. In this section, please provide details regarding where data supporting reported results can be found, including links to publicly archived datasets analyzed or generated during the study. Where no new data were created, or where data is unavailable due to privacy or ethical restrictions, a statement is still required. Suggested Data Availability Statements are available in section “MDPI Research Data Policies” at <https://www.mdpi.com/ethics>.

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Abbreviations

The following abbreviations are used in this manuscript:

Al ₂ O ₃	Aluminum oxide
CeO ₂	Cerium oxide
CH ₄	Methane
CNTs	Carbon Nanotubes
CQDs	Carbon quantum dots
CuO	Copper oxide
CVCC	Constant Volume Combustion Chamber
D	Diesel
D+S	Diesel and SDBS blends
D+S+CNT	Diesel, SDBS and carbon nanotubes blends
DCN	Derived Cetane Number
GO	Graphene oxide
GQD	Graphene quantum dots
H ₂	Hydrogen
HC	Unburned hydrocarbons
HCCI	Homogeneous Charge Compression Ignition
ID	Ignition Delay

LHV	Low Heating Value
MWCNTs	Multi-walled Carbon Nanotubes
NH ₃	Ammonia
NTC	Negative Temperature Coefficient
RCCI	Reactivity Controlled Compression Ignition
SCCI	Stratified Charge Compression Ignition
SDBS	Sodium dodecylbenzene sulfonate
TiO ₂	Titanium dioxide
WSD	Smoke point Wear Scar diameter
ZnO	Zinc oxide

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