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Investigation on the Explosion Characteristics of an Aluminum Dust−Diethyl Ether−Air Mixture

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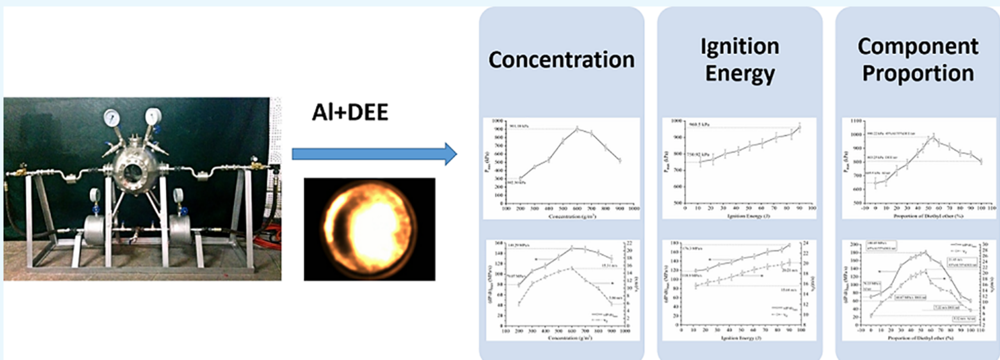

ABSTRACT: In this work, the explosion characteristics of an aluminum (Al)−diethyl ether (DEE)−air mixture were investigated in a 20 L spherical vessel. The effect factors of the explosion characteristics considered were fuel concentration, component proportion, and ignition energy. With the increasing concentration of the mixed fuel (Al/DEE = 1:1), the maximum pressure (P_{max}), the maximum rate of pressure rise ($(dP/dt)_{max}$), and the flame propagation speed (ν_F) exhibit an inversely "U-shaped" curve. The maximum P_{max}, $(dP/dt)_{max}$, and ν_F values are 901.2 kPa, 148.3 MPa/s, and 15.3 m/s, respectively, corresponding to an optimum concentration of 600 g/m^3. The P_{max}, the $(dP/dt)_{max}$, and the ν_F increase with the addition of DEE when the proportion of DEE is below 55% but have a decrease tendency when the proportion of DEE is over 55%. As the explosions of Al and DEE were mutually promoting, the studied explosion characteristics of the Al−DEE−air mixture are obviously higher than those of pure Al or DEE in air. The minimum ignition energy (MIE) of the Al−DEE−air mixture is 1.9 mJ, between the MIE of Al and DEE. With the increase of ignition energy, P_{max}, $(dP/dt)_{max}$, and ν_F all increase, while the minimum explosion concentration presents a linear decreasing trend. This work could provide significant scientific evidence for evaluating the explosion risk of the Al−DEE−air mixture.

1. INTRODUCTION

The mixed fuels consisting of combustible liquid, solid, or gas are commonly used in industry. Research studies about the explosion of mixed fuels mainly focus on the development and the severity of the explosion.[1−4] Compared with simple fuels, there have been only fewer studies on the explosion behavior of solid−liquid mixtures in air. In recent years, the interaction between the components of mixed fuels has received increasing attention. It was found that the mixtures were more reactive than all the other simple hybrid mixtures in the complex hybrid system.[5−8] In view of the higher explosion risk, it is necessary to study the explosion of complex hybrid mixtures. Fuel air explosive (FAE) is one kind of hybrid mixture that consists of solid, liquid fuel, and air. To improve the explosion performance of the FAE, flake aluminum powder is considered to mix with liquid fuels in some factories.

Al powder is widely used in the military for propellants in aerospace devices, rockets, and missiles.[9,10] With a high energy density and good compression-ignition characteristics, DEE can be used as an engine ignition accelerant or as an alternative fuel for diesel engines.[11] In addition to their broad applications, Al and DEE are dangerous due to their flammability and explosive nature.[12−19] In the past few decades, explosions of aluminum dust clouds have posed serious threats to the safety of human life and property.[20,21] As reported, the volatile and flammable DEE could generate a large variety of peroxide species when exposed to air and light, which may lead to a more serious explosion.[22] It is conceivable that the explosion caused by an Al−DEE−air mixture would bring serious consequence to the factories if there were no

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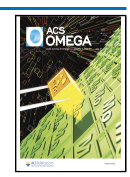

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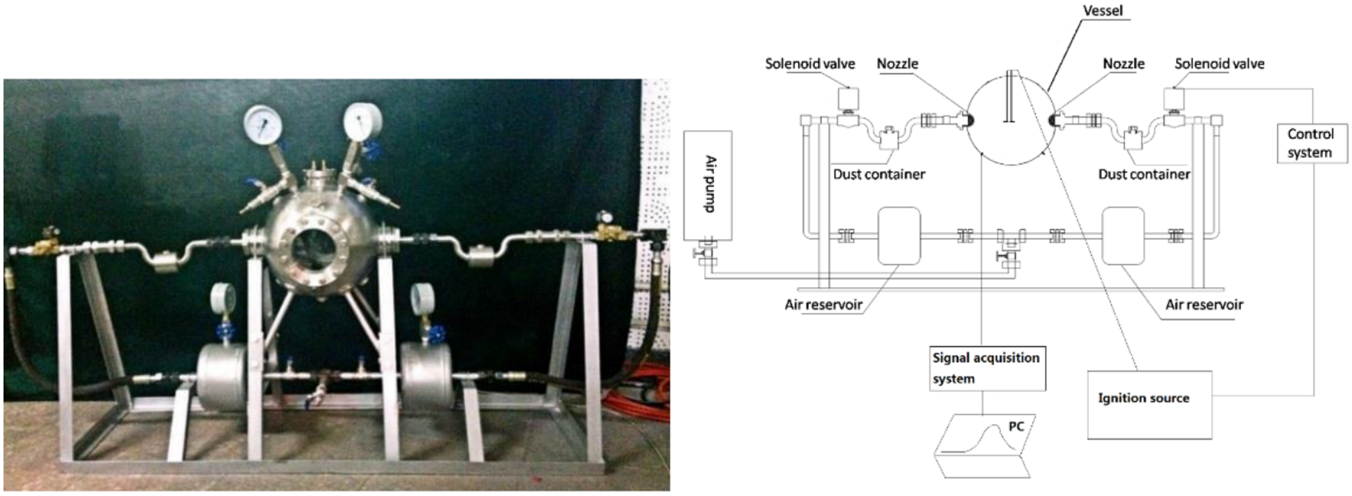

Figure 1. Experimental setup.

proper safeguards. In order to prevent the accidental explosions occurred during the production process in these factories, the research on the explosion characteristics of the multiphase mixed fuel is necessary.

In recent years, studies on the explosions of Al dust mixed with flammable gases were reported. Yu et al. indicated that both microdiffusion flame and asymmetric flame existed during the explosion of Al−H_2−air mixtures.[23,24] The addition of hydrogen resulted in a more continuous flame front and a faster flame propagation velocity. For the explosion overpressures and the pressure rise rates of the hybrid mixtures, it was reported that the tested explosion characteristics of the Al−H_2−air mixture were higher than those of the H_2−air mixture and Al dust−air mixture.[25,26] Zhang et al. proposed that the addition of aluminum dust reduced the lower flammability limit of gaseous epoxypropane and increased the maximum rate of combustion pressure rise of the gaseous epoxypropane/air mixtures.[27] James et al. recorded the propagation of constant-pressure flames of hybrid aluminum/methane/oxidizer mixtures using high-speed cameras.[28] The flame speed first declined then remained constant with the increasing aluminum concentration in a lean oxygen atmosphere. While in an excess oxygen atmosphere, the flame speed increases with the increasing aluminum concentration before it became constant.

As the interaction between the multiphase components is quite complex, very few research has been done on the mixtures consisting of Al powder and liquid fuel. Liu et al. established a two-phase (gas−solid) flow and detonation model to explore the heterogeneous detonation characteristics of aluminum dust/JP-10/air mixtures. Results showed that the detonation velocity and temperature of hybrid mixtures are both higher than those of aluminum dust/air mixtures and JP-10/air mixtures.[29] Liu et al. measured the explosion characteristics of RDX/aluminum powder/nitromethane/air mixtures.[30,31] The maximum rate of pressure rise and maximum pressure of RDX/aluminum powder mixtures both first increased and then decreased with the increasing mass fraction of RDX. The maximum pressure of RDX/aluminum powder/nitromethane/air mixtures continuously decreased with the increasing mass fraction of RDX but increased with the raising mass fraction of Al. Yao et al. found that the explosion pressure of the aluminum dust/diethyl ether/air mixture was more sensitive to the ambient pressure, while the lower flammability limit of the mixture was more sensitive to the ambient temperature.[32] Liu et al. discussed the interactions between the components of Al−DEE−air by analyzing the lower flammability limits of the mixture under different proportions.[33] The addition of aluminum obviously raised the lower flammability limit of the hybrid mixture as the aluminum power absorbs the heat to crack, which was released by the reaction of DEE.

In this work, the typical explosion characteristics (i.e., the maximum pressure, the maximum rate of pressure rise, the flame propagation speed, the minimum ignition energy, and the minimum explosion concentration) of the Al−DEE−air mixture were investigated. The effect factors of these explosion characteristics considered in this work were fuel concentration, component proportion, and ignition energy. The present experimental work employs an experimental method recently introduced in previous work in which the different phase components were well mixed and distributed before the ignition. The objective of the present work is to evaluate the explosion risk of hybrid Al−DEE−air mixtures.

2. EXPERIMENTAL DETAILS

2.1. Experimental Apparatus. The tests were performed in a 20 L closed spherical vessel. As shown in Figure 1, the experimental system is composed of a vessel with two symmetrical nozzles, a high-voltage electric ignition system, a control system, a venting pump, a transient pressure measurement system, and a data acquisition system. The details of the two nozzles have been described in our previous work.[34] A pair of tungsten electrodes are installed in the center of the vessel, connected with the high-voltage electric spark generator. The ignition energy was calculated by $1/2\ CU^2$, where C and U are the capacitance and voltage obtained from an electric igniter, respectively.

First, the Al powder and the liquid DEE were weighted separately. After filling the Al powder sample into the containers, the DEE was injected into the Al powder sample by using a syringe. In that way, they were per-mixed and filled into the containers. Furthermore, the pneumatic system and the specially designed nozzles provide turbulence disturbance, which allows the aluminum powder and DEE droplets to be more evenly distributed in the vessel. The pressure of the compressed air was set to 0.4 MPa, and the duration time was

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50 ms. The explosion vessel was partially vacuumed to −0.035 MPa to ensure that the fuel was ignited at standard atmospheric pressure (1 atm). Once the solenoid valves were opened, the compressed air pushed the mixed samples into the reaction vessel to form flammable clouds. The electric igniter was initiated after a delay time of 65 ms.[34] The explosion occurred after the ignition, and the experimental data were collected by the data acquisition system. The vessel and containers were cleaned, ready for the next test.

2.2. Materials. The aluminum powder used for explosion experiments was tested using a laser particle analyzer of Sympatec Gmbh. The particle diameter distribution is shown in Figure 2. The average diameter and the Sauter mean

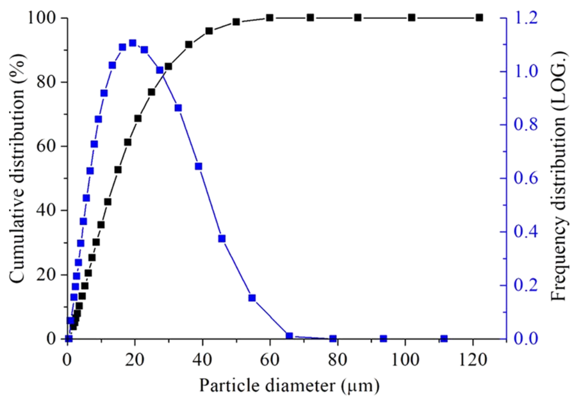

Figure 2. Particle diameter distribution of Al.

diameter of aluminum powder were 14.28 and 7.66 μm, respectively. The morphology of the aluminum powder was observed through scanning electron microscopy. Figure 3 shows the lamellar shape and the typical size of aluminum powder. The physical properties of diethyl ether are given in Table 1. As shown in Table 1, the auto-ignition temperature of DEE is 160 °C and the flash point is −45 °C. The flammability limit of DEE−air ranges from 1.9 to 36% under standard conditions.[35] The minimum ignition energies of Al and DEE are 15 and 0.33 mJ, respectively.

2.3. Data Processing. The mixture explosion experiments were repeated three times under the same conditions. In each test, the explosion pressure evolution was obtained using four pressure sensors (Kistler, Switzerland) installed on the wall of the vessel. The average value of the explosion pressure recorded using the four pressure sensors was calculated for the sake of analysis. The error of the results was composed of two parts: one is the error generated in the experimental process (5%), and the other is the error of data processing. The error of data processing was determined by the standard deviation of the data obtained from the four pressure sensors.

In Figure 4, a typical pressure profile with time during an explosion test is shown, where P_{max} is the maximum pressure

Table 1. Physical Properties of Diethyl Ether

formula	$C_2H_5OC_2H_5$	Al
calorific value	33.9 MJ/kg	30.4 MJ/kg
density	713 kg/m^3	2700 kg/m^3
boiling point	34.4 °C	2056 °C
flash point	−45 °C	
explosion limits	1.9−36%	37−50 g/m^3
minimum ignition energy	0.33 mJ	15 mJ
stoichiometric air fuel ratio (mass basis)	11.1	4.23
auto ignition temperatureheat of combustion	160 °C2752.9 kJ/mol	645 °C822.9 kJ/mol
cetane number	>125	

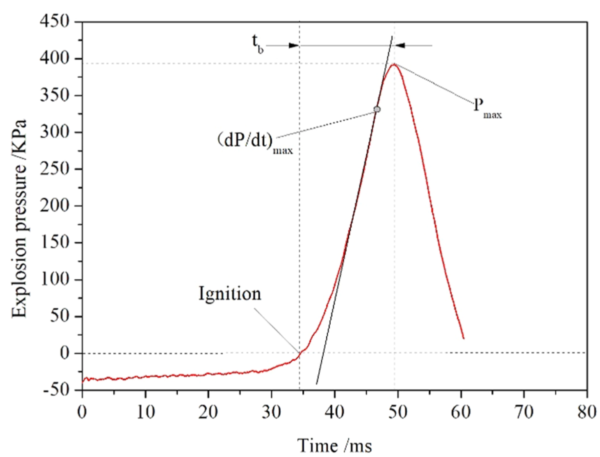

Figure 4. Typical pressure profile as a function of time during mixture explosion.

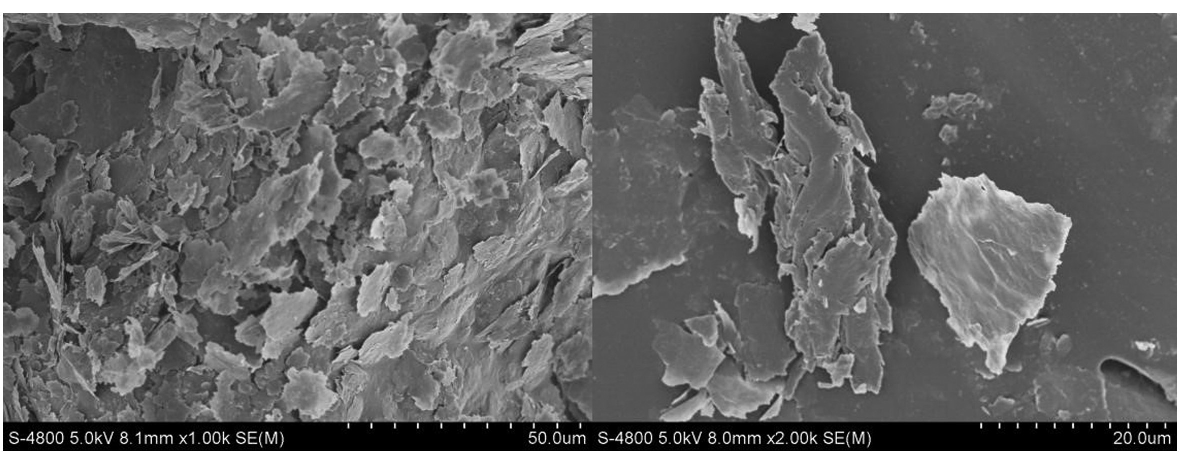

Figure 3. Scanning electron microscopy images of Al powders.

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and $(dP/dt)_{max}$ is the maximum rate of pressure rise. The burning time (t_b) is the time interval between the time corresponding to the ignition point and the time corresponding to the maximum explosion pressure (P_{max}) point. Therefore, the flame propagation speed (ν_F) can be calculated by t_b and the radius ($R_{20L-Sphere}$) of the explosion vessel.[36] The equation is as follows (eq 1):

$$\nu_F = \frac{R_{20L-Sphere}}{t_b}(m/s) \tag{1}$$

3. RESULTS AND DISCUSSION

3.1. Influence of Mixture Concentration. The influence of fuel concentration on the explosion characteristics of the mixtures was investigated first. The mass ratio of Al to DEE in the Al−DEE−air mixture was fixed at 1:1, and the ignition energy was set to 10 J. The range of the fuel concentration varied from 300 to 900 g/m^3.

As shown in Figure 5, the maximum value of P_{max} is 901.2 kPa, corresponding to 600 g/m^3. When the fuel concentration is lower than 600 g/m^3, there is sufficient oxygen in the vessel. P_{max} depends on the amount of heat released by the reaction between fuel and oxygen. The more fuel there is, the more energy released during the explosion process, resulting in a greater explosion pressure. Thus, the P_{max} increases with the increasing fuel concentration. While the fuel concentration exceeds 600 g/m^3, there is finite oxygen in the vessel. Instead of participating in the combustion reaction, the excess fuel will absorb a part of energy to impede the reaction. Therefore, the higher fuel concentration leads to a greater loss of energy, resulting in a lower explosion pressure. As a consequence, the P_{max} shows a downward trend.

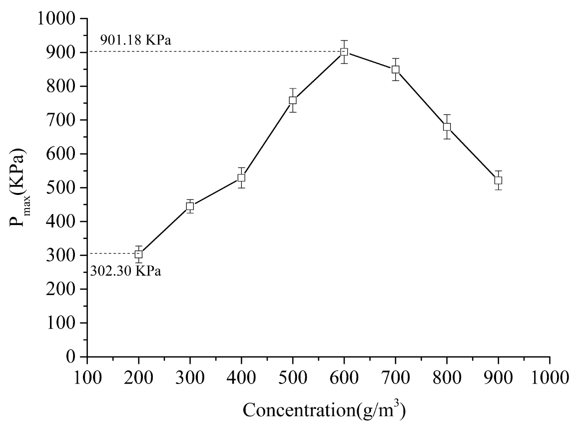

Figure 5. P_{max} vs mixture concentration.

In Figure 6, the maximum value of $(dP/dt)_{max}$ is 148.3 MPa/s, and the maximum value of ν_F is 15.3 m/s, corresponding to an optimum concentration of 600 g/m^3. As shown in the figure, the variation tendency of $(dP/dt)_{max}$ is consistent with that of P_{max}. When the fuel concentration is below 600 g/m^3, the $(dP/dt)_{max}$ mainly depends on the rate of chemical reaction during explosion. The more fuel takes part in the reaction, the greater rate of chemical reaction it is. The $(dP/dt)_{max}$ increases with the increase of fuel concentration. When the concentration is above 600 g/m^3, the amount of fuel participating in the chemical reaction remains constant since the amount of oxygen in the vessel is constant. Excess fuel makes the rate of chemical reaction slow down by consuming heat and hinders the diffusion of oxygen. Thus, the rate of chemical reaction will not increase with the increasing fuel concentration.

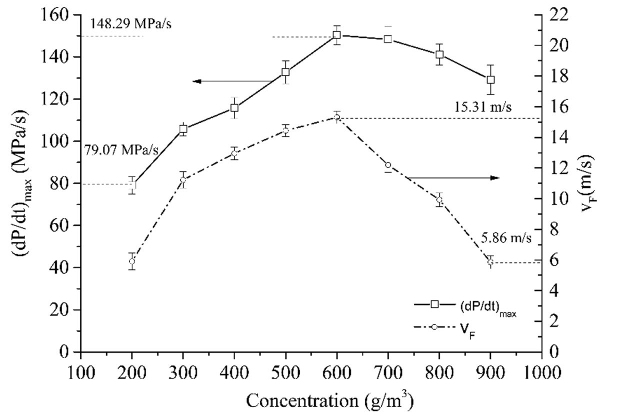

Figure 6. $(dP/dt)_{max}$ and flame propagation speed vs concentration.

The increase of the fuel concentration shortens the distance between particles, improving the heat transfer efficiency. As a result, the burning time of the flammable mixture shortens and the ν_F increases. However, the distance between particles could not be infinitesimally small because of repulsive forces between molecules. When the concentration is higher than 600 g/m^3, the influence of the distance change is weak. Excess fuel hinders the heat propagation and oxygen diffusion. Thus, the ν_F begins to decrease. The rate of chemical reaction directly affects ν_F, and the increase of chemical reaction rate makes ν_F faster.

By analyzing the influence of the concentration on the explosion characteristics, the maximum values of the explosion parameters are obtained. The extreme values of P_{max}, $(dP/dt)_{max}$, and ν_F reflect the explosion severity of Al−DEE−air mixtures. The results can be used to assess the explosion risk and provide data basis for the design of protective measures.

3.2. Influence of Ignition Energy. The effect of the ignition energy on the explosion characteristics of the Al−DEE−air mixture was investigated in this section. The mass ratio of Al to DEE was fixed at 1:1. The fuel concentration was fixed at 600 g/m^3, and the ignition energy varied from 12 to 90 J.

As shown in Figure 7, the relationship between P_{max} and ignition energy is a clear linear positive correlation. P_{max} increases from 750.9 to 960.5 kPa when the ignition energy

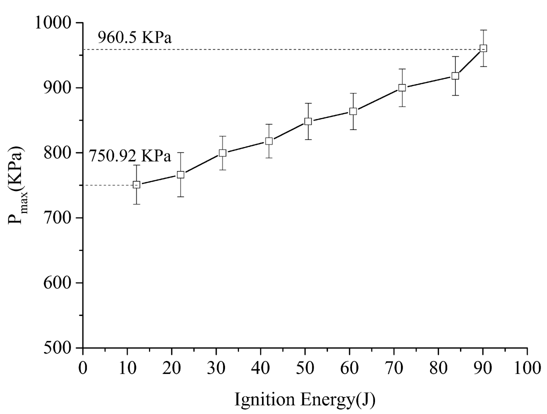

Figure 7. P_{max} vs ignition energy.

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increases from 12 to 90 J. The ignition energy is just an inducer for the chemical reaction, which increases the heat to some extent at the initial stage of the reaction, rather than the amount of heat released during the explosion process. Thus, the changing tendency of P_{max} with the increasing ignition energy is not obvious.

Figure 8 shows that both $(dP/dt)_{max}$ and ν_F are positively correlated with ignition energy. When the ignition energy

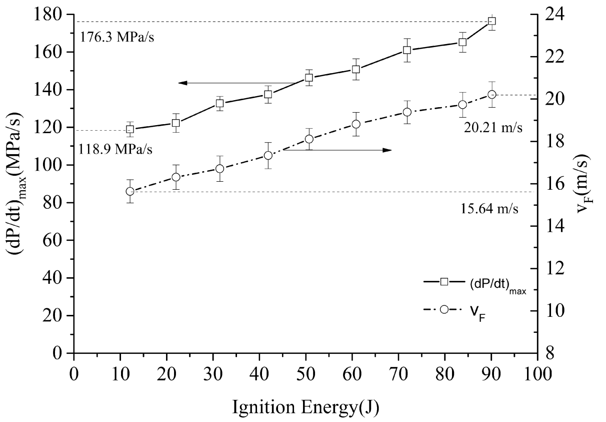

Figure 8. $(dP/dt)_{max}$ and flame propagation speed vs ignition energy.

varies from 12 to 90 J, the $(dP/dt)_{max}$ increases from 118.9 to 176.3 MPa/s, and the ν_F increases from 15.6 to 20.2 m/s. The increase of ignition energy promotes the production of free radicals. Hence, the rate of chemical reaction is accelerated. Furthermore, the increase of ignition energy leads to a rise of the ambient temperature in the vessel, which enhances the volatility of the mixture. The more volatiles participating in the reaction, the faster the reaction rate is. As a result, the burning time is shortened. With the increase in ignition energy, the volume of the ignition space becomes larger and the turbulence intensity increases, which improves the combustion efficiency. Therefore, the $(dP/dt)_{max}$ and the ν_F increase with the increasing ignition energy.

The results provide the evidence that the increasing initial ignition energy rises the risk of Al−DEE−air mixture explosion. In conclusion, the ignition sources must be eliminated as far as possible in the process of industrial production and transportation.

3.3. Influence of the Mass Ratio of Aluminum Dust to Diethyl Ether. A series of tests were conducted to analyze the effect of the mass ratio of aluminum dust to diethyl ether on the explosion characteristics. The mixture concentration is 600 g/m^3, and the ignition energy is set to 90 J. The proportion of DEE varied from 0 to 100% with a 10% step.

As shown in Figures 9 and 10, the variation tendency of P_{max}, $(dP/dt)_{max}$, and ν_F with the increasing proportion of DEE is similar. The maximum values of P_{max}, $(dP/dt)_{max}$, and ν_F are 980.2 kPa, 180.5 MPa/s, and 21.5 m/s, respectively. It is worth mentioning that the P_{max}, the $(dP/dt)_{max}$, and the ν_F of Al/DEE/air mixture explosions at any mass ratio are higher than those of pure Al/air mixtures or pure DEE/air mixtures.

When the proportion of DEE in the mixture is lower than 55%, the addition of DEE increases the P_{max} of the Al−DEE−air mixture. The reason is that DEE has a higher combustion heat at the same mass when compared with aluminum powder. The higher the proportion of DEE, the more heat will be released. Another reason for the increase in the heat release is

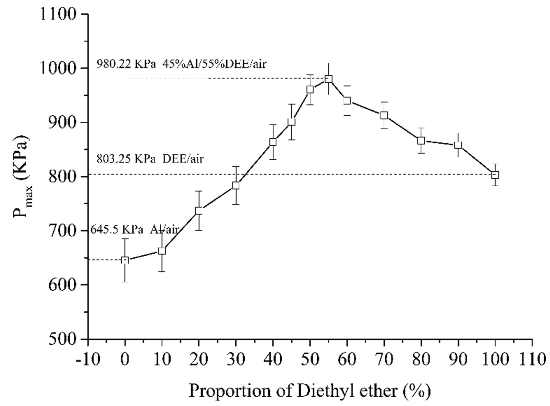

Figure 9. P_{max} vs the proportion of diethyl ether.

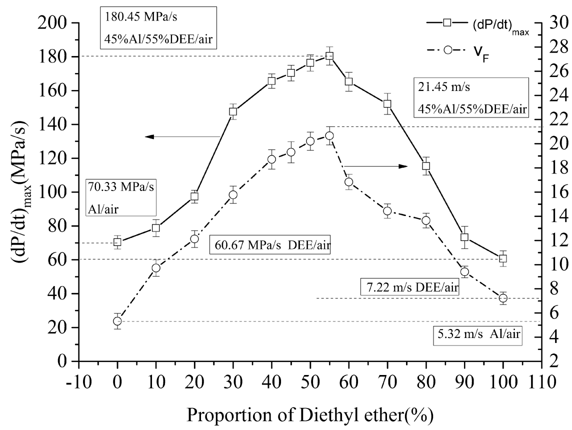

Figure 10. $(dP/dt)_{max}$ and flame propagation speed vs flame propagation speed.

the rise of the participation rate of Al in the reaction. The heat released by the reaction of DEE contributes to crack the oxide layer on the surface of Al powder and accelerate the melting of Al powder. In that case, more Al powder takes part in the chemical reaction. Additionally, Al powder could react with CO_2 and H_2O produced by DEE oxidation, which in turn promotes the combustion reaction of the DEE. When the proportion of DEE in the mixture is higher than 55%, the DEE involved in the reaction will not increase as the amount of oxygen in the vessel is limited. The excess DEE consumes heat to vaporize, reducing the participation rate of Al powder during the combustion process. Therefore, the P_{max} decreases with the increasing proportion of DEE.

The DEE act as a medium to transfer heat between Al particles when the proportion of DEE is lower than 55%. The continuity of Al powder is improved, and the heat transfer efficiency between particles is accelerated with the addition of DEE. The combustion of DEE provides more energy, making the ignition of Al powder easier and accelerating the combustion rate of Al powder. The heat released from the combustion of Al powder increases the temperature of unburned fuel, causing a thermal expansion and promoting the flame propagation. In that case, the $(dP/dt)_{max}$ and the ν_F of the Al−DEE−air mixture increase with the increasing proportion of DEE. At a high proportion of DEE, the drops of DEE will enfold Al powder, isolating the contact between Al and oxygen and reducing the participation rate of Al powder in the reaction. As the combustion rate of DEE is faster than that of Al powder, the flame front of Al powder cannot catch up

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with that of DEE. The Al powder will increase the heat capacity of the Al−DEE−air mixture and make the flame speed slow down. Thus, the $(dP/dt)_{max}$ and the ν_F of the Al−DEE−air mixture decreased.

By comparing the explosion characteristics of the Al−DEE−air mixture, Al−air mixture, and DEE−air mixture, it is found that the complex hybrid mixtures present a greater risk of explosion than the simple hybrid mixtures. Hence, when setting the monitoring of the concentration of Al powder and DEE and the prevention of the explosion during production and storage, the interaction effect between Al and DEE should be taken into consideration.

3.4. The Minimum Explosion Concentration (MEC) and the Minimum Ignition Energy (MIE). The lowest fuel concentration that can propagate an explosion is often regarded as the MEC. In this section, the MECs of Al−DEE−air mixtures were measured under different ignition energies. The mass ratio of Al to DEE was fixed at 1:1.

In Figure 11, the MEC decreases linearly with the increase of ignition energy. The effective collision between fuel molecules

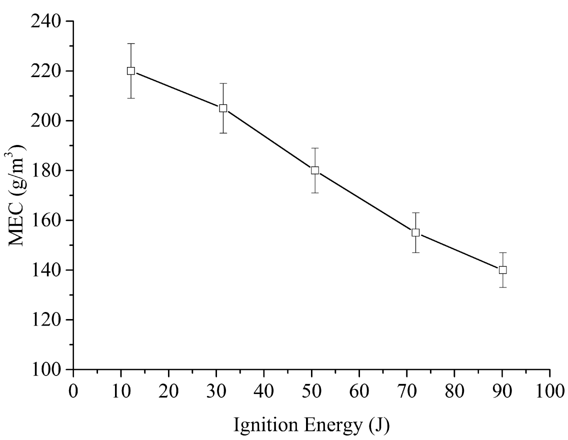

Figure 11. Effect of the ignition energy on MEC.

and oxygen molecules near the MEC is relatively low. The increase of ignition energy accelerates the thermal movement of molecules and improves the chance of collision between molecules. In this case, the concentration that would not have exploded occurs with a higher ignition energy.

The MIE refers to the minimum energy used to ignite fuel and cause an explosion at the most sensitive concentration. As shown in Figure 12, the most sensitive concentration of the mixture is 500 g/m^3, and the corresponding MIE is 1.9 mJ. The MIE exhibits a "U-shaped" curve with the increase of the concentration. When the concentration is below 500 g/m^3, the MIE decreases with the increase of fuel concentration. This phenomenon is mainly because the less fuel there is, the higher ignition energy would be needed to ignite the mixtures. When the concentration is above 500 g/m^3, the MIE shows an upward trend with the increasing fuel concentration. For these cases, excess aluminum particles act as radiators and absorb the energy; hence, the fraction of active particles for combustion reduced. In addition, excessive aluminum is attached to the tip of the electrode, which weakens the energy released by the electric spark. Therefore, the mixture is harder to be ignited than expected.

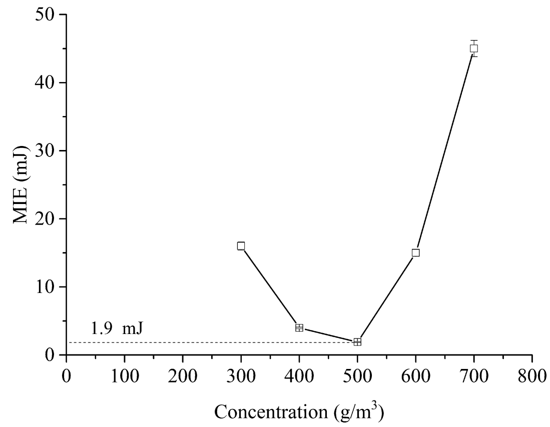

Figure 12. Effect of the mixture concentration on MIE.

4. CONCLUSIONS

This work investigates the explosion characteristics of the Al−DEE−air mixture under normal atmospheric pressure and temperature and is associated with previous studies that have studied similar mixtures in the same 20 L sphere vessel. The explosion parameters included the maximum explosion pressure, maximum rate of explosion pressure rise, flame propagation speed, MEC, and MIE. After the analysis, the conclusions are as follows:

(1) The P_{max}, $(dP/dt)_{max}$, and ν_F all exhibit an inversely "U-shaped" curve with the increasing concentration of the mixture and the increasing proportion of DEE in the mixture. The peak values of P_{max}, $(dP/dt)_{max}$, and ν_F are 901.2 kPa, 148.3 MPa/s, and 15.3 m/s, respectively. These results provide scientific evidence to the safety measures for production and storage of the Al−DEE−air mixture.

(2) As the explosions of Al and DEE were mutually promoting by improving the heat transfer efficiency between particles, the P_{max}, $(dP/dt)_{max}$, and ν_F of the Al−DEE−air mixtures are higher than the values of the Al−air mixture or DEE−air mixture. The comparison of explosion characteristics among the Al−DEE−air mixture, Al−air mixture, and DEE−air mixture indicates that there is synergistic effect between Al powder and DEE.

(3) The higher ignition energy accelerates the thermal motion of the molecules and increases the chance of collisions between molecules. Hence, the MEC presents a linear decreasing trend with the increase of ignition energy. Furthermore, the MIE of the Al−DEE−air mixture is 1.9 mJ, which is 87.3% lower than the MIE of pure Al (15 mJ). The addition of DEE plays an important role in reducing the MIE. It is worth stressing that the ignition sources must be eliminated as far as possible in the process of industrial production and transportation of Al−DEE−air mixtures.

Aimed at improving the prevention of Al−DEE−air mixture explosion during industrial production, this work provides the maximum value of P_{max}, $(dP/dt)_{max}$, and ν_F of the mixture and analyzes the influence of fuel concentration, component proportion, and ignition energy on these characteristics. The experimental results provide scientific evidence for the evaluation about the explosion risk of Al−DEE−air mixtures.

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Notes

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