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## Ignition Sensitivity of Hybrid Mixtures

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### Abstract

This paper reports on experimental and theoretical investigations into the ignition sensitivity of four gas-dust hybrid mixtures consisting of either methane or propane combined with either starch or polyethylene. The experimental work was done using a Hartmann tube to determine minimum ignition energy (MIE) and a Godbert-Greenwald furnace to determine minimum ignition temperature (MIT). The test procedures were based on European standards EN 13821 and EN 50281 for dust-air mixtures for MIE and MIT, respectively, with minor modifications to accommodate testing of hybrid mixtures. The experimental results show a significant decrease in the MIE and MIT of hybrid mixtures compared to corresponding values for single-components. The increase in ignition sensitivity was apparent even with gas concentrations below the lower flammability limit, or with dust concentrations below the minimum explosible concentration. For example, the MIE of polyethylene dust decreased from 40 mJ to 5 mJ when only 1 vol% of propane was added.

Mathematical models were also used to predict the MIE and MIT of hybrid mixtures. The predicted values were compared to experimental results, which showed very good agreement. The models allow more accurate estimates for MIE and MIT of a hybrid mixture than using corresponding data for either the dust or gas component.

Keywords: dust explosion, hybrid mixture explosion, minimum ignition temperature, minimum ignition energy, ignition sensitivity.

## 1. Introduction

The explosion hazards associated with hybrid mixtures are present in process facilities that either handle or process materials in different states of aggregation. Accidents involving these types of explosions can cause failure to equipment, serious worker injuries or fatalities, environmental damage, business interruption, and sometimes complete destruction of the factory, resulting in huge financial loss and devastating consequences. It is well known that the explosion sensitivity of a hybrid mixture cannot be predicted based on the measured values of its individual components, and that hybrid mixtures can be more hazardous than their single-component values [1-6]. However, these kinds of mixtures are usually not considered in the various hazard and risk assessments even though combustible dusts are dispersed in industrial equipment containing flammable gases or solvents [7].

Moreover, research on hybrid mixtures is limited and very complicated because of the large number of complex physical processes that occur during the explosion and numerous parameters that must be considered. In order to prevent or mitigate the risks associated with hybrid mixtures, improved knowledge of their explosion behaviour is required. It is therefore important to know the explosibility parameters related to the ignition sensitivity such as minimum ignition energy (MIE) and minimum ignition temperature (MIT) of hybrid mixtures so that explosion incidents can be prevented.

The MIE is the minimum amount of electrical energy stored in a capacitor which, when released as a high voltage spark, is sufficient to ignite a fuel mixture at its most easily ignitable concentration in air. MIT is the lowest temperature of a heated surface which can ignite a fuel-oxidizer mixture at fuel concentrations within its explosible limits. An explosible atmosphere generated in an uncontrolled way in proximity to a hot surface with temperature above the actual MIT or electrical discharge above the MIE, can result in an explosion [8, 9]. Consequently, in the prevention and mitigation of explosions, it is important to quantify ignition sensitivity of fuel mixtures in order to effectively evaluate and control potential ignition sources.

The present work is a subset drawn from previously published studies by the authors [10-12] on ignition sensitivity of hybrid mixtures. The objectives of this paper are (1) to present representative examples of hybrid mixtures and to illustrate the synergistic effect of gas-dust mixtures on increased ignition sensitivity; (2) to demonstrate the value in performing experimental work with hybrid mixtures; and, (3) to exemplify the usefulness and validity of mathematical models in predicting the MIE and MIT of hybrid mixtures.

## 2. Materials and experimental work

The minimum ignition temperatures and minimum ignition energies were determined for four dust-gas hybrid mixtures consisting of either starch or polyethylene dust combined with either methane or propane gas. Since particle size has a strong influence on the explosibility parameters of dusts and hybrid mixtures, the particle size distributions of the starch and polyethylene dust samples were characterized using multi-wavelength laser diffraction particle size analysis. The results are shown in Figure 1.

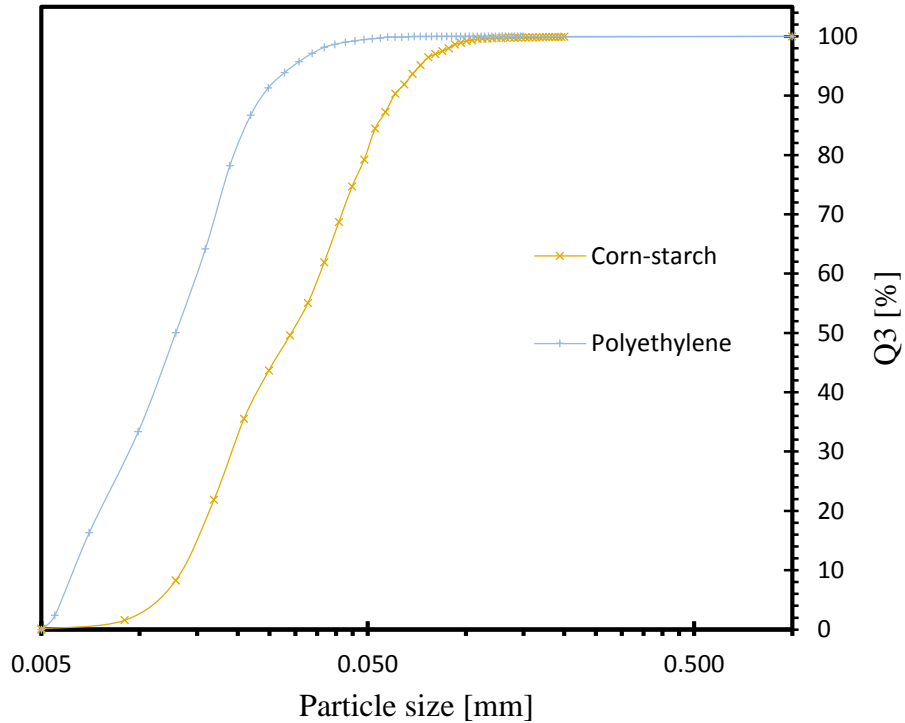


Figure1: Particle size distribution of dust samples.

To reveal the dust particle morphology, scanning electron microscopy (SEM) images of the two dust samples were taken at the same magnification (Figure 2). The images give an indication of particle shape and porosity. It can be seen that the polyethylene particles agglomerate, which may affect the global settling velocity of the material, depending on how easily the agglomerations are broken up during dispersion.

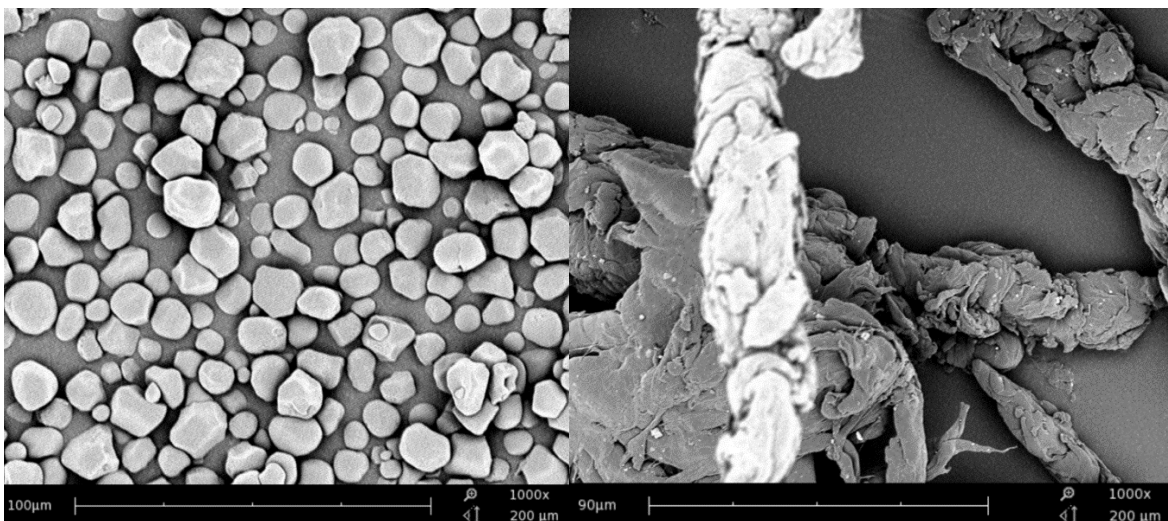


Figure 2: SEM images of starch (left) and polyethylene (right) dust samples.

Table 1 provides data on particle size distribution and chemical properties of the combustible dusts, including elemental analysis, median particle sizes, moisture content and heat of

combustion. Table 2 lists the basic thermodynamic, ignition and combustion properties of the gases used.

Table 1: Properties of starch and polyethylene dust samples.

Dust Sample	Particle Size ( $\mu\text{m}$ )		Volatile Content (% wt)	Moisture Content (% wt)	Calorific Value (MJ/kg)	Elemental Analysis (% wt)				
	d50	d90				C	H	O	S	N
Starch	14	21	93.77	0.50	15.30	44.3	6.3	48.9	0.4	0.0
Polyethylene	34	84	98.67	0.68	39.68	86.0	14.0	0.0	0.0	0.0

Table 2: Properties of methane and propane [13, 14].

Property	Methane	Propane
	$\text{CH}_4$	$\text{C}_3\text{H}_8$
Molecular formula	$\text{CH}_4$	$\text{C}_3\text{H}_8$
Purity (%)	99.87	99.00
Density ( $\text{g}/\text{cm}^3$ )	6.6E-4	4.93E-4
Molecular weight ( $\text{g}/\text{mol}$ )	16	44.1
Explosible range (vol%)	4.4–17	1.7–10.8
Melting point ( $^{\circ}\text{C}$ )	–161	–187
Specific heat capacity ( $\text{J}/\text{mol}\cdot\text{K}$ )	35.69	73.60
Boiling point ( $^{\circ}\text{C}$ )	–182.5	–42.1
Heat of vaporization ( $\text{kJ}/\text{mol}$ )	–74.87	–103.80
Maximum explosion pressure (bar(g))	8.1	9.8
Maximum experimental safety gap (mm)	1.14	0.92
Temperature class	T1	T1
Explosion group	IIA	IIA
Heat of combustion (MJ/kg)	55	50
Heat of combustion ( $\text{kJ}/\text{mol}$ )	–286	–890
Adiabatic flame temperature (K)	2226	2267
Maximum burning velocity ( $\text{cm}/\text{s}$ )	39	45

## 2.1. Test procedure for minimum ignition energy

Dust cloud minimum ignition energy was measured using an electric spark igniter in the Hartmann tube and following test procedures provided in EN 13821 [15]. In order to test hybrid mixtures, the apparatus was modified as shown in Figure 3. The combustion chamber consists of a 1.2-L glass tube, provided with a mushroom-shaped dust dispersion system. All experiments were carried out under the same initial conditions as specified in EN 13821.

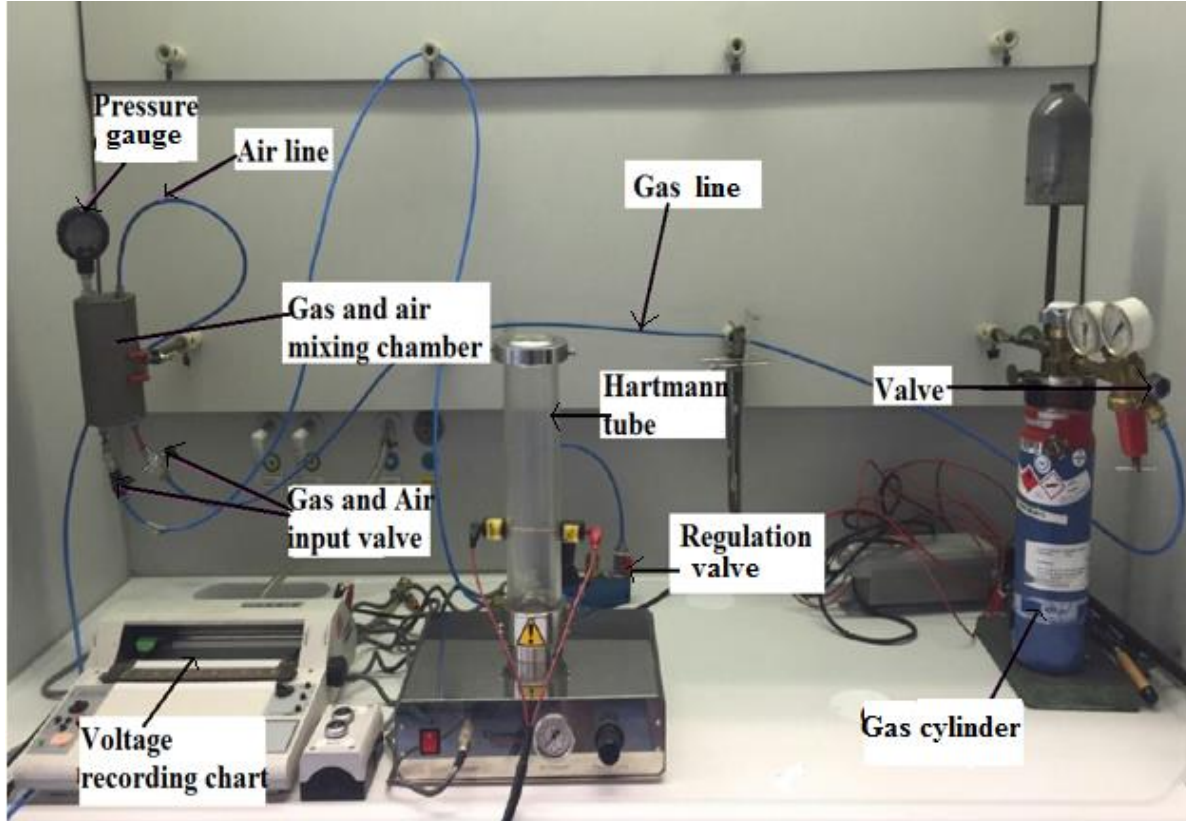


Figure 3: Image of MIE experimental setup.

The required amount of dust was placed in the mushroom part in the dust dispersion system. The dust dispersion was triggered by a compressed air blast at 7 bar(g). The air blast generates considerable turbulence and results in the creation of a dust cloud. A spark was passed between two electrodes. In accordance with the test protocol, the spark gap was varied from 2–6 mm, to achieve the desired spark discharge energy. The minimum ignition energy lies between the highest energy at which ignition fails to occur ( $E_1$ ) for ten successive attempts to ignite the dust-air mixture and the lowest energy at which ignition occurs ( $E_2$ ) within up to ten repeated attempts. For comparison of MIE among different hybrid mixtures, a single-value estimate ( $E_s$ ) was determined using Eq. 1 [15]:

$$\log E_s = \log E_2 - I[E_2] \cdot \frac{(\log E_2 - \log E_1)}{(NI+1) \cdot [E_2] + 1} \quad (1)$$

Where  $I[E_2]$  is the number of tests with successful ignition at energy level  $E_2$  and  $(NI + 1) \cdot [E_2]$  stands for the total number of tests at the energy level of  $E_2$ .

For hybrid mixture testing, the Hartmann apparatus was modified to allow the input of combustible gas. At first, the required gas concentration (below the lower flammability limit) was mixed with air in the gas-air mixing chamber. Contrary to dust testing whereby the dust is dispersed by compressed air, in the case of hybrid mixture testing, the dust was dispersed by a blast of flammable gas-air mixture. The gas concentration was determined using Dalton's law of

partial pressures. In all cases, the test gas concentration was below the lower flammability limit of the gas.

### 2.1.1. Test procedure for minimum ignition temperature

The experimental setup consisted of a Godbert-Greenwald (GG) furnace which is commonly used to determine the MIT of dust clouds. In contrast to EN 50281 [16], the length of the reaction cylinder used in the present study was twice the specified length of 42 cm.

For the test with only dust, the EN 50281 test procedure was followed. For gases, and hybrid mixtures, the equipment was modified as shown in Figure 4. All experiments were carried out under the same initial conditions in accordance with EN 50281.

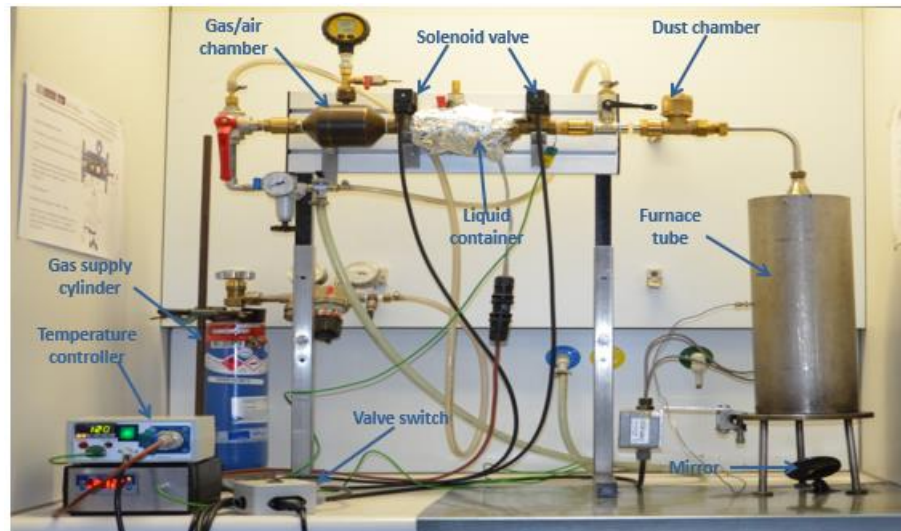


Figure 4: Image of MIT experimental setup.

The MIT for each of the individual hybrid components was first determined. The furnace tube was heated and fixed at the desired temperature, and the weighed amount of dust was placed in the dust chamber. The dispersion reservoir was pressurized with air in the range of 0.1 to 0.5 bar(g) and the dust sample was then dispersed through the furnace tube by a blast of air. The ignition criterion was a visible flame at the bottom of the furnace opening. Both the pressure (0.1 to 0.5 bar above atmospheric pressure) and the mass of dust (0.1 to 0.5 g) were varied until “vigorous” combustion was observed. The conditions in which the most vigorous combustion was observed were taken as “worst-case”. These conditions were maintained for the remaining tests which were performed at successively lower furnace temperatures until flames were no longer observed after ten consecutive tests at the same temperature. The prescribed difference in temperatures between ignition and no ignition was 5 °C. The lowest temperature at which an ignition with a flame occurred was taken as the minimum ignition temperature.

For the tests with gas, the GG furnace was modified by installing a gas feed line to the air reservoir as shown in Figure 4. The same experimental principle as explained for dust was used for the gas test. The only difference in this case was that the air was premixed with the combustible gas in the air reservoir and the dust chamber was left empty. The composition of the gas mixtures was determined based on partial pressures. The chosen pressures were between 0.1 to 0.5 bar above atmospheric pressure and the concentrations were also within the explosible range of individual substances.

For the test with hybrid mixtures, the same experimental principle as explained before was used. In this case, the procedure followed was a combination of the test methods for dust and for gas as individual components. Once the MIT was obtained, further testing was performed at 5 °C temperature increments below the MIT. Dispersion pressure and concentration were varied to test for ignition.

### 3. Results and discussion

#### 3.1. Minimum ignition energy of hybrid mixtures

The determined MIE values for the starch and polyethylene dust samples were 41 mJ and 116 mJ, respectively. The ignitability of a dust cloud is strongly dependent on particle size distribution. Finer particles have greater specific surface area which allows more rapid devolatilization in the presence of an ignition source, and therefore results in a more readily ignitable mixture [17]. This was observed in the test results for dusts with similar volatile contents, where the MIE for starch with median particle size of 14 µm was found to be three times lower than that of polyethylene with median particle size of 34 µm. The lowest achievable ignition energy of the device used in this work was limited to 4 mJ. Thus, the MIE for propane or methane could not be measured directly, as their values lie well below 4 mJ. Hence, the MIE values for methane of 0.28 mJ and propane of 0.25 mJ were taken from the literature [18].

For the next step, the MIE of hybrid mixtures was determined using results from the individual-component dust and gas tests. The main focus of the hybrid mixture testing was to verify if the addition of a non-flammable concentration of gas could decrease the MIE of the dust. A description of how the MIEs of dusts and hybrid mixtures were obtained was presented in Section 2.1. The lower flammability limit of the gases (methane = 5 vol% and propane = 2.0 vol%) were initially determined at the same test conditions used to estimate MIE for dusts. After obtaining the MIE of the individual dust, concentrations of the gases (methane = 1.0 vol%, 2.0 vol%, 3.0 vol%, 4.0 vol%, and propane = 0.6 vol%, 1.0 vol%, 1.4 vol% and 1.7 vol%) were added to the dust and the experiments were performed at an ignition energy below the MIE of the dust.

Figures 5 and 6 present MIE test results for various hybrid mixtures as well as a comparison with an empirical model (Eq. 2). Derivation and validation of this model is provided in [12]:

$$\text{MIE}_{\text{hybrid}} = \frac{(\text{MIE}_{\text{dust}})}{(\text{MIE}_{\text{dust}}/\text{MIE}_{\text{gas}})^{\frac{c}{c_0}}} \quad (2)$$

Where  $C$  is the gas volume concentration (vol %);  $C_0$  is the gas concentration (vol %) leading to the lowest MIE, and  $MIE_{dust}$  and  $MIE_{gas}$  are the MIEs of dust and gas respectively. Note: this mathematical model is valid only if,  $C \leq C_0$ .

Each plot (Figures 5 and 6) shows the MIE of the hybrid mixtures on the ordinate and gas concentration on the abscissa. The square represents the experimental hybrid MIE of dust with propane, triangle represents the experimental hybrid MIE of dust with methane, the dashed line indicates the empirical model estimation of the hybrid MIE with propane and the solid line represents the computational estimation of the hybrid MIE with methane. Error bars based on an error and uncertainty analyses are plotted on the experimental results. The total quantifiable error for experimental work was found to be 8.1% [12].

Figure 5 shows the results obtained for starch mixtures with propane and methane. It was generally noticed that the MIE of starch decreased with the addition of a small amounts of gas. Various gas concentrations below the LEL were mixed with the dust and tested for ignition at energy levels below the MIE for the dust. With respect to starch, a series of tests were performed below the MIE by adding gas in concentrations below the LEL as shown in Figure 5. It was noticed that the ignition energy decreased with increased gas concentration. For example, the MIE of starch decreased from 40 mJ to 22 mJ, 15 mJ and 4.3 mJ when methane concentrations of 1 vol%, 2 vol% and 4 vol% were added. A similar trend was also observed for starch-propane mixtures.

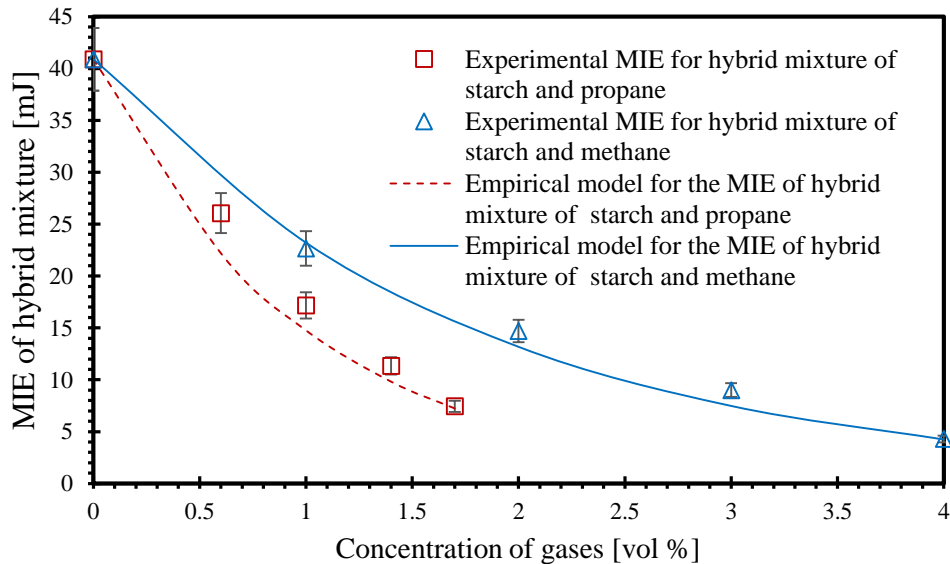


Figure 5: Experimental results and empirical prediction of the minimum ignition energy of hybrid mixtures of starch mixed with propane or methane as a function of gas concentration.

Similar trends were observed for polyethylene mixed with methane and propane. The MIE of the hybrid mixture significantly decreased with the addition of non-explosible gas concentrations as shown in Figure 6. This reflects the fact that the ignition sensitivity of a hybrid mixture is higher than that of the individual dust. The MIE of hybrid mixtures with polyethylene dust decreased by 93% with 1.7 vol% of propane and 96% with 4 vol% of methane, compared to



the MIE of the dust alone. It was also noticeable that the experimental results were in good agreement with the results obtained from the empirical model.

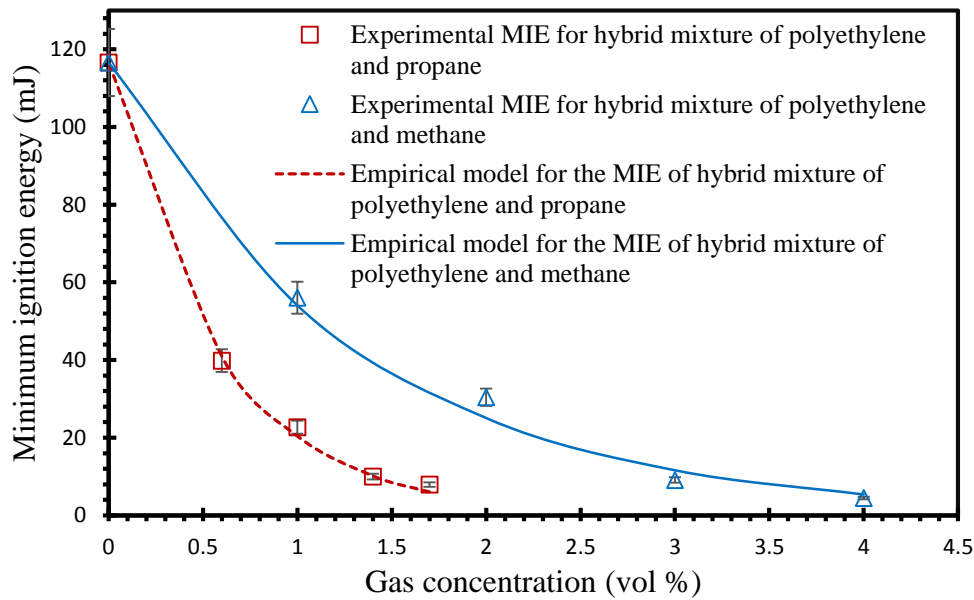


Figure 6: Experimental results and empirical prediction of minimum ignition energy of hybrid mixtures of polyethylene mixed with propane or methane as a function of gas concentration.

The results obtained from this work are consistent with the work done by Franke et al. [19]. These authors observed that with the addition 0–3 vol% of methane to coal dust, the MIE of the hybrid mixture was reduced by one to two orders of magnitude.

### 3.2. Minimum ignition temperature of hybrid mixture

The MIT of hybrid and non-hybrid mixtures was investigated in the modified GG furnace as explained in Section 2.2. The results were compared with a mathematical model to predict the MIT of hybrid mixtures. Initially, the MITs for individual dust and gas samples were determined. The MITs for methane, propane, starch and polyethylene were found to be 600 °C, 500 °C, 380 °C and 340 °C, respectively.

With respect to the dusts, the MIT is influenced by various parameters such as particle size, moisture content, and volatile content. The results illustrate that dusts with higher volatile content ignite at lower temperatures. This could be explained based on the phenomenon that dusts with higher volatile content produce more combustible gas at the same conditions, which contributes to gas phase combustion. The volatile content as well as other parameters of the dust samples is presented in Table 1. A comparison between the volatile content of the dusts (polyethylene = 98.67 wt% and starch = 93.77 wt%) and the MIT results (starch = 380 °C and polyethylene = 340 °C) reveals that dusts with lower volatile contents have higher ignition temperatures as seen for starch and polyethylene. With respect to the gases (methane and propane), it was noticed that methane had a higher MIT compared to propane. This trend may be attributed to their basic chemical and physical properties such as heat of combustion as presented in Table 2. For example, materials with higher heats of combustion have higher ignition temperatures as seen in the case of

methane and propane, which have heats of combustion of  $-286$  kJ/mol and  $-890$  kJ/mol, and MITs of  $600$  °C and  $500$  °C, respectively.

With respect to hybrid mixtures, Figures 7 and 8 show the MIT test results of dust-gas mixtures. The X symbolizes the MIT of hybrid mixtures, the square symbolizes the MIT of dust and the circle symbolizes the MIT of gas. The experimental results were compared with a mathematical model (Eq. 3) as indicated by a ‘triangle’ in each plot. Details and validation of this model are provided in [10, 11]:

$$T_{i,\text{hybrid}} = T_{i,g} \left( \frac{T_{i,d}}{T_{i,g}} \right)^{\frac{C_g}{C_d}} \quad (3)$$

Where  $T_{i,g}$  is the MIT of gas,  $T_{i,d}$  is the MIT of dust,  $C_g$  is the gas concentration in the mixture (methane =  $19.7$  g/m<sup>3</sup> at 3 vol%, propane =  $20.1$  g/m<sup>3</sup> at 1 vol%), and  $C_d$  is the dust concentration in the mixture (starch =  $82$  g/m<sup>3</sup>, polyethylene =  $87$  g/m<sup>3</sup>).

To verify the accuracy of the model, error bars based on the experimental uncertainty of 6.3% for the hybrid mixture test in the GG furnace were plotted on the experimental values. An error and uncertainty analysis is discussed in [10].

Figure 7 shows the results for the MIT of hybrid mixtures of dusts (starch and polyethylene) and gas (methane). It can be seen that the MIT of hybrid mixtures is lower than those of gas-air mixtures. For example, methane with a MIT of  $600$  °C decreased to  $570$  °C with the addition of  $82$  g/m<sup>3</sup> of starch, which is below the minimum explosible concentration of  $145$  g/m<sup>3</sup>. Similar results were observed when polyethylene dust was added to methane. It can also be seen that the presented models give a very good prediction of the MIT of a hybrid mixture of dusts and methane.

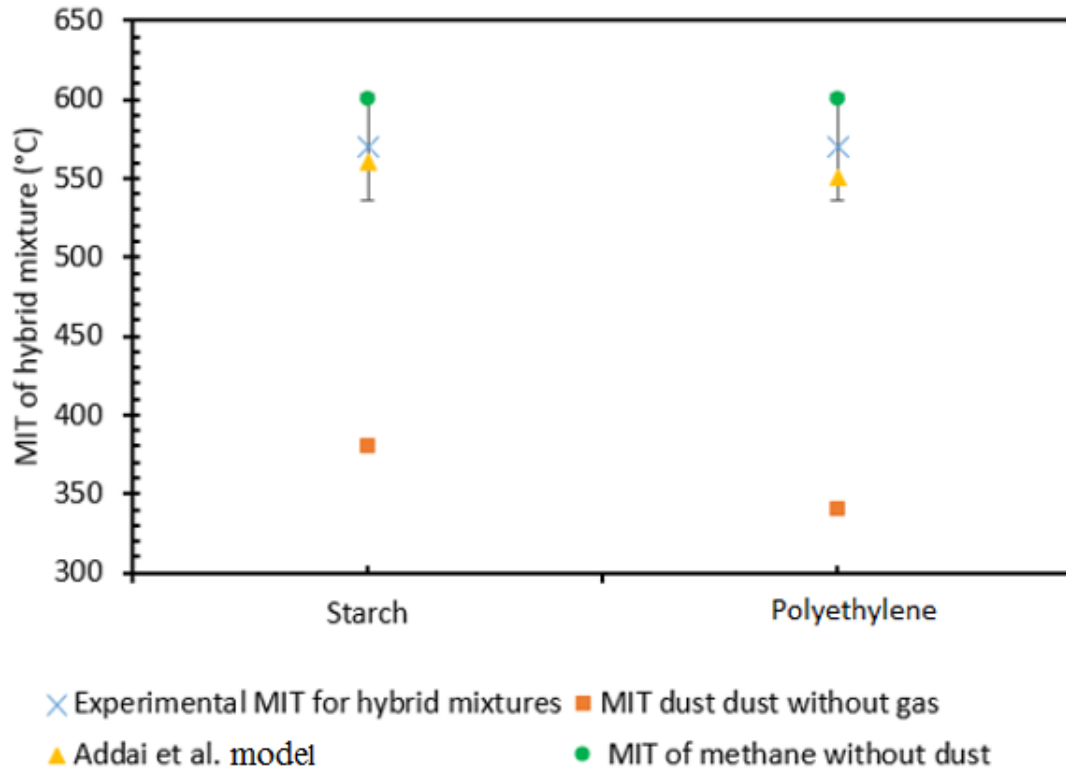


Figure 7: Ignition temperature of hybrid mixture of dusts (starch and polyethylene) with methane and comparison with a mathematical model.

Furthermore, Figure 8 presents the results from hybrid mixtures of propane and the dusts. It can also be seen that the MIT of propane decreases with the addition of a non-explosible concentration of dust. For instance, the MIT of propane decreased from 500 °C to 450 °C when a non-explosible concentration of starch (82 g/m<sup>3</sup>) was added. This behaviour was also noticed when a non-explosible concentration of polyethylene was added to propane at a temperature below its MIT. It was further observed that the calculated results from the presented model gave a good prediction of the MIT of hybrid mixtures within experimental uncertainty.

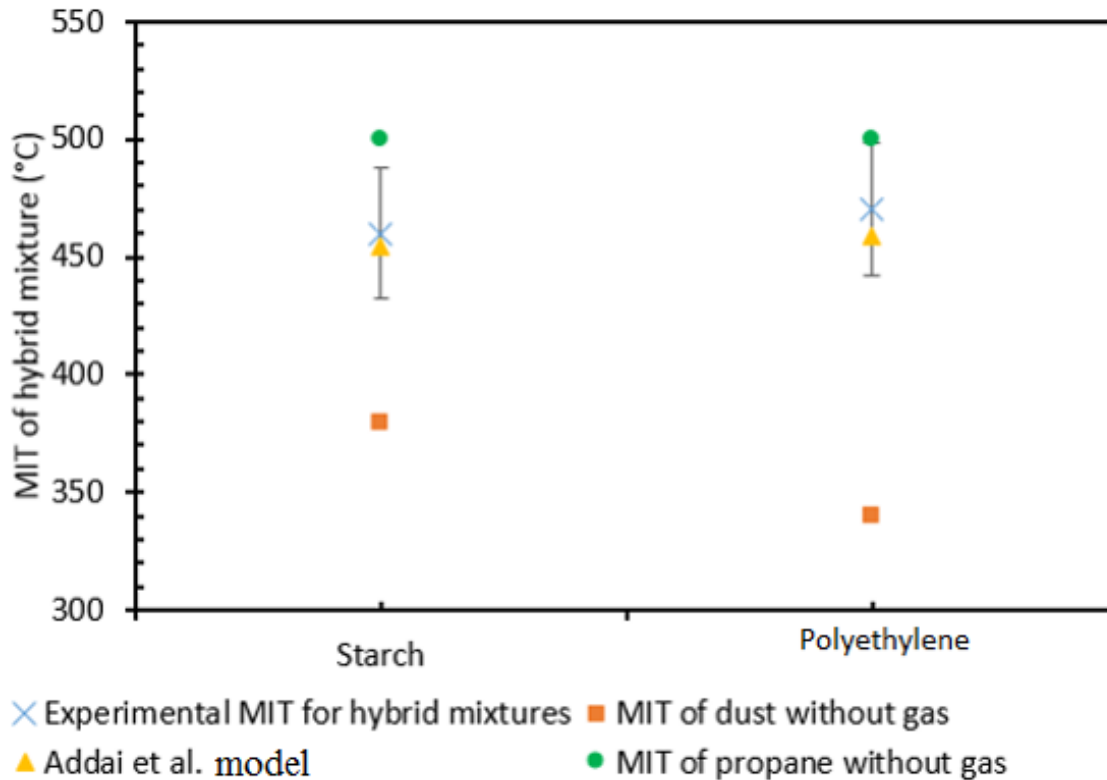


Figure 8: Ignition temperature of hybrid mixture of dusts (starch and polyethylene) with propane and comparison with a mathematical model.

The decrease in the MIT of gases upon addition of non-exposable concentrations of dusts could be due to pyrolysis and devolatilization of dusts, resulting in the addition of volatile matter to the already introduced gas. An introduction of combustible (organic) dust into the heated furnace heats the organic particles. This produces volatile matter or combustible gases. These volatile gases are then mixed with either combustible gas to increase the gaseous fuel concentration, which consequently enhances the ignitability of the mixture.

#### 4. Conclusion

This study has summarized research on ignition sensitivity of four gas-dust hybrid mixtures, consisting of either starch and polyethylene combined with either methane or propane. The experimental results affirmed that the addition of gas and dust have an influence on both minimum ignition temperature and minimum ignition energy of hybrid mixtures, even though the added concentrations are below their respective lower flammability limits or minimum explosible concentrations. The addition of combustible gases can be seen as a replacement for volatiles released from the dust during pyrolysis and hence significantly affects the ignition sensitivity of hybrid mixtures. Moreover, based on these findings, it can be concluded that the ignition sensitivity of hybrid mixtures cannot be predicted by simply overlapping the effects of the single component substances. Finally, the presented models to estimate the minimum ignition energy and minimum ignition temperature of hybrid mixtures yielded predictions that were in good agreement with the experimental results.

## Reference

1. Denkevits, A., Explosibility of hydrogen–graphite dust hybrid mixtures. *Journal of Loss Prevention in the Process Industries*, 2007. **20**(4): p. 698-707.
2. Denkevits, A. and B. Hoess, Hybrid H<sub>2</sub>/Al dust explosions in Siwek sphere. *Journal of Loss Prevention in the Process Industries*, 2015. **36**: p. 509-521.
3. Amyotte, P., et al. Determination of hybrid mixture explosion severity. in 13th International Symposium on Loss Prevention and Safety Promotion in the Process Industries, Brugge, Belgium. 2010.
4. Amyotte, P., et al., Prevention and mitigation of dust and hybrid mixture explosions. *Process Safety Progress*, 2010. **29**(1): p. 17-21.
5. Addai, E., D. Gabel, and U. Krause, Ignition and explosion behavior of hybrid mixtures of two and three components in: 4. Magdeburger Brand-und Explosionsschutztag, HS-Magdeburg Stendal: Magdeburg, 2015. 2015, ISBN 978-3-00-048960-0, 26. und 27. März.
6. Addai, E. K et al., Explosion characteristics of three component hybrid mixtures. *Process Safety and Environmental Protection*, 2015. **98**: p. 72-81.
7. Garcia Agreda, A., Study of hybrid mixture explosions. 2010, Università degli Studi di Napoli Federico II.
8. Amyotte, P.D., An introduction to dust explosions: understanding the myths and realities of dust explosions for a safer workplace. 2013: Butterworth-Heinemann, Waltham, MA.
9. Di Benedetto, A et al., On the determination of the minimum ignition temperature for dust/air mixtures. *Chemical Engineering Transactions*, 2010. **19**.
10. Addai, E. K et al., Models to estimate the minimum ignition temperature of dusts and hybrid mixtures. *Journal of Hazardous Materials*, 2016. **304**: p. 73-83.
11. Addai, E. K et al., Experimental investigation on the minimum ignition temperature of hybrid mixtures of dusts and gases or solvents. *Journal of Hazardous Materials*, 2016. **301**: p. 314-326.
12. Addai, E.K., et al., Minimum ignition energy of hybrid mixtures of combustible dusts and gases. *Process Safety and Environmental Protection*, 2016. **102**: p. 503-512.
13. Rossini, F.D., et al., Selected values of chemical thermodynamic properties. Vol. 500. 1952: US Government Printing Office Washington, DC.
14. Brandes, E., et al., Sicherheitstechnische Kenngrößen. 2003: Wirtschaftsverlag NW, Verlag für neue Wissenschaft.
15. CEN, Determination of minimum ignition energy of dust/air mixtures, European Standard EN13821, European Committee for Standardization, Brussels. 2002.
16. CEN, EN 14522 Determination of the minimum ignition temperature of gases and vapours 2005.
17. Eckhoff, R., Dust explosions in the process industries: identification, assessment and control of dust hazards. 2003: Gulf professional publishing.
18. Haase, H., Electrostatic hazards: their evaluation and control. 1977: Wiley-VCH.
19. Franke, H., Bestimmung der Minderstzudenergie von Kohlenstaub/Methan/Luft Gemisches (hybride Gemische). *VDI-Berichte*, 1978. **304**: p. 69-72.

