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On the explosion and flammability behavior of mixtures of combustible dusts



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ABSTRACT

In the work presented in this paper, the explosion and flammability behavior of combustible dust mixtures was studied. Lycopodium, Nicotinic acid and Ascorbic acid were used as sample dusts.

In the case of mixtures of two dusts, the minimum explosive concentration is reproduced well by a Le Chatelier's rule-like formula, whereas the minimum ignition energy is a linear combination of the ignition energies of the pure dusts.

An unexpected behavior has been found in relation to the explosion behavior and the reactivity. When mixing Lycopodium and Nicotinic acid or Ascorbic acid, the rate of pressure rise of the mixture is much higher than the rate of pressure rise obtained by linearly averaging the values of the pure dusts (according to their weight proportions), thus suggesting that strong synergistic effects arise; but it is comparable to that of the most reactive dust in the mixture.

The observed behavior seems to be linked to the presence of minerals in the Lycopodium particles which catalyze oxidation reactions of Nicotinic acid and Ascorbic acid, as suggested by TG analysis.

In the case of mixtures of three dusts, a similar behavior is observed when the concentration of Lycopodium is twice that of the other two dusts.

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1. Introduction

In chemical processes, a high number of accidents are imputable to the explosion of combustible dusts causing failures to equipment, injuries and damage to people and surrounding environment.

In recent years, owing to the possibility of formation of hybrid mixtures (i.e., mixtures of flammable gas/vapor and dust) at industrial scale, attention has also been paid toward the basic understanding of their behavior with the objective

of both preventing the occurrence and mitigating the consequences of such events (Abuswer et al., 2013; Amyotte et al., 2010; Dufaud et al., 2008, 2009; Garcia-Agreda et al., 2011; Di Benedetto et al., 2012; Khalili et al., 2012; Kosinski et al., 2013; Sanchirico et al., 2011).

Results on the explosion of hybrid mixtures show that mixing combustible dust with flammable gas or vapor may significantly increase the flammability (Khalili et al., 2012; Kosinski et al., 2013; Sanchirico et al., 2011) and the explosion severity (Amyotte et al., 2010; Dufaud et al., 2008, 2009;

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Garcia-Agreda et al., 2011; Kosinski et al., 2013) with respect to each mixture component.

In food, pharmaceutical and wood industries, pigment and dyes industries, and chemical manufacturing processes, powder products are frequently handled giving rise to possible mixtures of dusts in air. Some examples are mixtures of paracetamol/microcrystalline cellulose, instant cappuccino and coffee, polymers with different molecular weight during the co-polymer manufacturing.

In the case of dust mixtures, most of the studies are focused on the effect of mixing a combustible dust with an inert solid acting as suppressant (Dastidar et al., 1997, 1999; Amyotte, 2006; Eckhoff, 2003; Dufaud et al., 2012). The flammability and explosion behavior of mixtures of two or more combustible dusts has been scarcely studied. The most detailed works on dust mixtures were performed many years ago by Nagy et al. (1968) and Jacobson et al. (1962, 1964).

Nagy et al. (1968) measured the flammability and explosion parameters for 181 typical industrial mixtures like, for example, blasting machine dust (mixtures of quartz, hematite, magnetite and alpha iron). Jacobson et al. (1962, 1964) tested dust mixtures of the plastic industries (copolymers) and metal dusts used in titanium plants (for example, titanium, carbonaceous material, silicon dioxide, manganese and iron). However, from these data, it is not possible to derive the reactivity and the flammability of the mixture with respect to the single dust. Also, it is not clear if mixing one combustible dust with another combustible dust at a concentration lower than the minimum explosive concentration (MEC) would result in the explosion of the mixture.

More recently, Denkevits and Dorofeev (2005, 2006) studied the explosion behavior of fine graphite dust, tungsten dust and their mixtures. They showed that the mixture burns faster than both pure graphite and pure tungsten dusts.

Dufaud et al. (2012) investigated the flammability and the explosibility of various solid/solid mixtures, observing different behavior: the properties of the mixture uniformly ranged between those of the pure compounds or the explosibility of the mixture decreased with respect to the components.

In this work, we aim at studying the effect of adding a combustible dust to one or two combustible dusts on the flammability and explosion behavior. We chose to work with Lycopodium and Nicotinic acid, as these dusts are used as worldwide standard for explosion tests, and Ascorbic acid, which is an important industrial product.

Then, we investigated the explosion behavior of mixtures made of these three dusts.

Explosion tests were run in the 20L explosion vessel at different dust concentrations and relative amounts. Differently from standard tests (ASTM E1226), where two chemical igniters (2 × 5 kJ ignition energy) are used as ignition source, in this work a spark ignition was used to trigger the dust cloud. This allowed to avoid explosion overdriving (Dastidar et al., 2001).

We measured the maximum pressure (P_{max}), the maximum rate of pressure rise ($(dP/dt)_{max}$) and the minimum explosion concentration (MEC). We also performed measurements of the minimum ignition energy (MIE) in the Modified Hartmann Tube Apparatus (MIKE3). We tested the following mixtures: Lycopodium–Nicotinic acid/air, Ascorbic acid–Nicotinic acid/air, Lycopodium–Ascorbic acid/air and Lycopodium–Nicotinic acid–Ascorbic acid/air.

Table 1 – Dust particle size.

	Nicotinic acid	Ascorbic acid	Lycopodium
Percentile diameter, μm			
d(0.1)	5		28
d(0.5)	32	44	38
d(0.9)	93		51

Table 2 – Elemental analysis of Lycopodium.

Element	wt%
C	59.3
H	8.1
O	21.9
N	2.4
S	<0.1
Ash	7.0
Humidity	1.2

2. Materials and methods

2.1. Materials

As materials, we chose dusts used either as worldwide standard for explosion tests (i.e., Lycopodium and Nicotinic acid) or as important industrial product (i.e., Ascorbic acid). Dusts were supplied by Sigma Aldrich and were used as received. The particle size of the dusts is reported in Table 1.

Nicotinic acid ($\text{C}_6\text{NH}_5\text{O}_2$) was previously characterized by laser diffraction granulometry using di-ethyl ether as disperdant solvent (Malvern Instruments Mastersizer 2000); scanning electron microscopy (Philips mod. XL30); simultaneous TG/DSC analysis in N_2 flow (TA Instruments SDTQ600). Results (i.e., mean particle size = 32 μm ; humidity = 0 wt%) were presented in a previous paper (Garcia-Agreda et al., 2011). The particle density of Nicotinic acid is 1.473 g/cm^3 .

Ascorbic acid ($\text{C}_6\text{H}_8\text{O}_6$) has median particle size of 44 μm . The particle density is about 1.65 g/cm^3 .

The composition of Lycopodium spores was determined by elemental analysis carried out in EA Eurovector analyzer. The chemical formula obtained is $\text{CH}_{1.639}\text{O}_{0.277}\text{N}_{0.035}\text{S}_{0.001}$. Results reported in Table 2 revealed the presence of 7.0 wt% of ash. The particle density is about 1.18 g/cm^3 . The Lycopodium structure was also analyzed by High Resolution Field Emission Scanning Electron Microscopy (HR-FESEM, AURIGA Zeiss). SEM images reported in Fig. 1 show that particles are substantially mono-sized, with a mean diameter of about 40 μm . The Lycopodium spores appeared to be covered with an ornamentation of small round particles (Fig. 1b and c). The EDS analysis of the spore surface recognized as main mineral components: Al, Mg, K.

Metal analysis of Lycopodium was made by ICP-MS spectrometry (Agilent 7500 CE) of dissolved sample previously mineralized by microwave digestion. The results reported in Table 3 show the presence of Ca e Mg in concentration higher than 1000 mg/kg, Al, Mg and Ni of the order of 1000 mg/kg, Cu, Fe, Na, Mn, Zn, Ag, Ba in the range 30–70 mg/kg while other elements in lower concentration.

2.2. Experimental setup

Experiments were performed using the standard 20L apparatus manufactured by Adolf Kühner AG (CH). A scheme of the setup is shown in Fig. 2. The equipment is made of a stainless

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