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FIRST VERIFICATION OF THE MODIFIED HARTMANN-APPARATUS WITH ALUMINUM AND MAGNESIUM PARTICLES

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Abstract

The objectives of the present investigation are test and characterization of the modified Hartmann-Apparatus after constructional changes. Initial studies showed leaks between the steel tube and the acrylic glass tube at which particles reached outwards (1). The Hartmann-Apparatus offers a good possibility for assist studies for large scale experiments.
The aim of this study is identifying the explosion limits of different materials with defined particle size and particle surface area. In the present work aluminum and magnesium particles are used to investigate the flame velocity dependent on the air ratio.

Introduction

Dust explosions are fortunately rare occurrences, but they have the potential to kill. In the UK the analysis of dust explosions between the years 2000 and 2010 had shown 295 events of fire, explosion or fire & explosion. 55 of those incidents had happened in the metal industry (2). In the US 10-20% of the examined events could be attributed to metal dusts reactions (3) (4).This large number may be reduced if potential hazards are found and avoided.

To characterize the explosion and the ignition potential of metal dust a number of laboratory tests have been developed (for example: classification test, minimum ignition temperature test, lower explosion limit test etc.). (5). Beside the 20 l-ball and the 1 m³ container, the Hartmann-Apparatus is one standard method to determine combustion velocities of dust-air-mixtures (6). The benefits of the Hartmann-Apparatus are the small amount of necessary material, small space requirement, the easy usage and a fast characterization of dust-air-mixtures. Furthermore the small sample consumption benefits low test costs. Especially in the beginning of test series, before large scale experiments, it is an important aspect to curtail the space parameter.
In the present work, the flame propagation of aluminum and magnesium particles was investigated in Fraunhofer ICTs modified Hartmann-Apparatus (7) (1).

Experimental equipment

The modified Hartmann-Apparatus is built of a vertical opened up steel tube with an acrylic glass tube inside (volume approx. 1.2 l) and a hinged lid at the top. At the bottom an atomizer device is mounted to steadily disperse dust of particles in the reservoir.

Figure 1 and Figure 2 show the schematic design and a picture of the used Apparatus respectively.

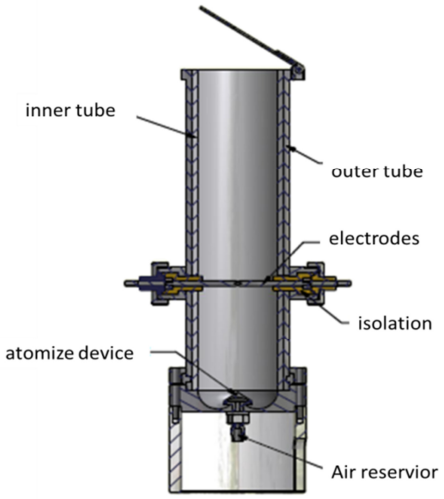

Figure 1: Schematic design of the modified Hartmann-Apparatus.

Figure 2: Picture of the modified Hartmann-Apparatus at the Fraunhofer ICT.

Test Procedure

In order to interpret the behavior of the different particle materials, certain characteristics like specific surface area (method after Brunauer-Emmett-Teller theory, Quantachrome Instruments), particle diameter (laser diffraction, Mastersizer 2000 Malvern Instruments Ltd, UK) and pre-oxidation rate

(thermal gravimetric analysis, Netzsch STA 449C Jupiter thermo-microbalance) have to be determined in the beginning.
With the resulting data the air ratio of the different dust-air-mixtures were calculated (Table 2). The air ratio λ is the amount of actually available air in relation to the amount of air required for a complete combustion.

$$\lambda = \frac{m_{Air,actual}}{m_{Air,required}}$$

For tests in the Hartmann-Apparatus the weighed test samples were deposited at the bottom of the tube and dispersed with a blast of air (from a 50 ml reservoir at 7 bar pressure). After a triggered ignition delay time the completely build up dust-air-mixture was ignited by a discharge spark between two electrodes in the center of the tube (parameters: 1 mm and 5 kV). Afterwards the reaction behavior was observed using a color high-speed camera (Phantom v.9.1 color; Vision Research).

Results and Discussion

Particle characterization:

The first material was aluminum powder made by Sibthermochim (Russia) using electrical wire explosions. The particle size of d(0,5) = 4,28 µm was measured by laser diffraction, its surface area by BET method resulted to a value of 4,18 $m^2\ g^{-1}$. The particle size calculated from the surface area measurements by BET assumed sphere like particles (Equation 1) provides 5.3 nm. From there we assumed that the powder consist of agglomerated nano sized particles (Figure 3).

Equation 1

$$\boldsymbol{d_{BET} = \frac{6}{\rho\, A_{BET}}}$$

The second material was commercially available magnesium powder supplied by Alfa Aesar. The particle size of d(0,5) = 38,120 µm was measured by laser diffraction, its surface area by the BET method resulted to a value of 5,123 $m^2\ g^{-1}$.

Figure 3 and Figure 4 show the SEM analysis of the magnesium and aluminum particles. In comparison to the sphere like structure of the aluminum particles, the magnesium particles have a plate like structure with a bumpy surface. Agglomeration could be observed in the case of aluminum particles.

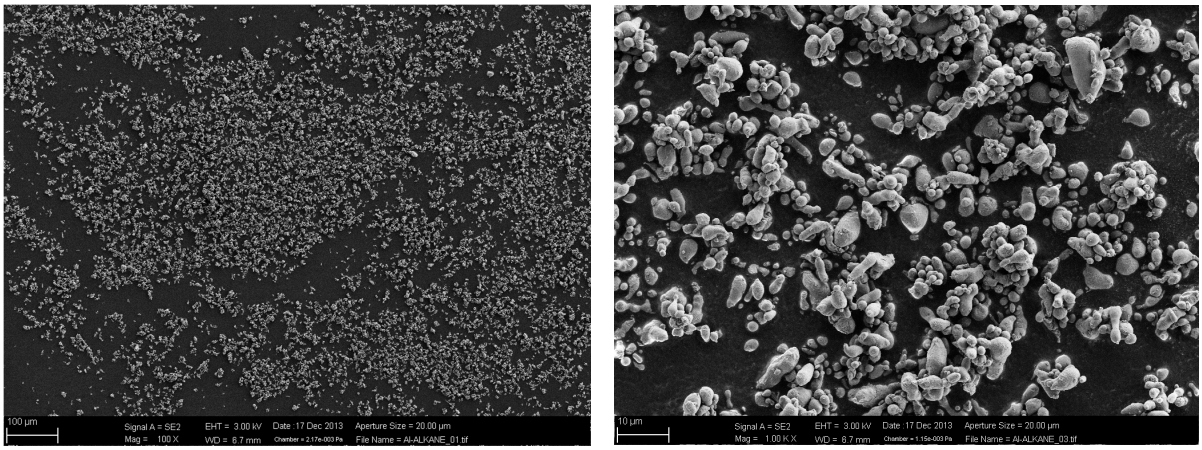

Figure 3: SEM micrographs of aluminum particles (Sibthermochim, $d_{0,5}$=4,25 µm; SEM magnification: 100x and 1000x).

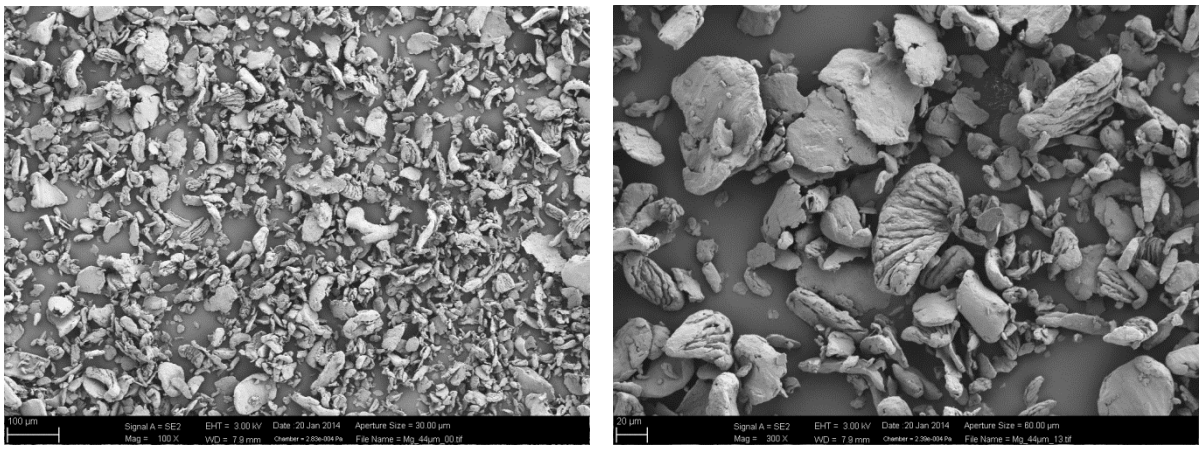

Figure 4: SEM micrographs of magnesium particles (Alfa Aesar, d(0.5) = 38.120 µm; SEM magnification: 100x and 300x).

Reaction of metal particles in a gaseous atmosphere is in most cases a highly exothermic process. In most cases it is an oxidation reaction, but reactions with nitrogen, carbon dioxide or other gases are also possible. Three conditions have to be fulfilled to get combustion of metal particles (8):

- The metal particle itself has to be combustible
- Oxidizer has to be available
- The ignition temperature of the metal particle has to be reached (Table 1) or an ignition source of sufficient energy has to be present.

To get a dust explosion three additional conditions have to be fulfilled: (9)

- The dust must have a particle size distribution that will propagate flame.
- The dust must be in suspension in the atmosphere with a sufficient amount of oxygen to sustain combustion.
- The dust concentration must be within the explosive range.

According to the VDI guideline 2263 “a dust is considered explosive if there will be a flame propagation after ignition and the dust-air mixture will result in a pressure rise in a closed vessel” (6).

The metal oxides, alumina and magnesia are thermodynamically more stable than the pure metals. Therefore the thin natural oxide layer is formed at the surface of the particles and protects the particle against further oxidation. On the other hand the same oxide layer thickness contributes to a higher oxide layer – metal - ratio in the case of small particles and therefore less metal contributes to the oxidation reaction and the released heat.

The pre-oxidation rate of the particles was measured by thermal gravimetric analysis (TGA) in previous studies (10) (11) (12). Equation 2 and Equation 3 are showing the underlying oxidation reactions of aluminum und magnesium in air.

Equation 2

$$4Al + 3O_2 \rightarrow 2Al_2O_3$$

Equation 3

$$2Mg + O_2 \rightarrow 2MgO$$

The theoretical mass increase due to the formation of oxides is 89 % for aluminum and 66 % for magnesium (Table 1). The observed mass increase of aluminum in the TGA-measurements until 1500°C is 85 %. Thus the pre-oxidation is approx. 4,49 %. For magnesium the observed mass increase of 63% in the experiment until 1000 °C results in a pre-oxidation rate of approx. 5 %. These results are in good agreement with the measurements of heat of combustion with a determinedpre-oxidation rate of 4,61 % for aluminum and 4,24 % for magnesium particles.

Table 1: General and measured data of aluminum und magnesium particles (8).

	Aluminum particles	**Magnesium particles**
General data		
Molar mass [g/mol]	26,98	24,31
Density [g/cm³]	2,70	1,74
Melting point [°C]	660	651
Ignition temperature of layer / dust [°C]	280 / 530	410 / 610
Combustion heat [kJ/g]	-31,05	-24,75
Mass incerase [%]	89	66
Measured data		
Specific surface [m²/g]	4,18	5,12
Average particle diameter [µm]	4,28	38,12

Combustion heat [kJ/g]	-29,62	-23,70
Pre-Oxidation rate on basis of combustion heat measurements [%]	4,61	4,24
Mass incerase [%]	85,10 (1500 °C)	63 (1000 °C)
Pre-Oxidation rate on basis of TGA measurements [%]	4,49	5

Experimental Investigation in the Hartmann-Apparatus:

The dust concentration in the experiments was calculated by the mass of dust divided by the chamber volume. It was varied over a range, from 300 to 850 g/m³ in the case of aluminum and from 300 to 1100 g/m³ in the case of magnesium. Weighted mass and the thus resulting dust concentrations and air ratio values are combined in Table 2.

Table 2: Overview about weighted mass, calculated dust concentration and Air ratio.

Mass [g]	Dust concentration [g/cm³]	Air ratio (without pre-oxidation)	Air ratio (with pre-oxidation)
		Aluminum	
0,4	333,3	1.6	1,13
0,6	500	0,79	0,75
0,8	666,6	0,59	0,56
1,0	833,3	0,47	0,44
		Magnesium	
0,4	333,3	1,60	1,68
0,7	583,3	0,91	0,96
1,0	833,3	0,64	0,67
1,3	1083,3	0,49	0,52

Analysis

In Figure 5 and Figure 6 series of example pictures are shown taken from the high speed recording of the experiments with 1 g magnesium particles and 0.6 g of aluminum particles dispersed inside a 1.2 l volume. The video sequences were shot at 4202 frames per second (fps) and the temporal difference between the selected images is 3.57 ms.

It is clearly visible that the reaction of aluminum particles start later and the combustion is more inhomogeneous than the reaction of the magnesium particles. Due to the inhomogeneous combustion

of the aluminum particles the obtained data from the modified Hartmann-Apparatus are very sparse. Three measurements are needed to make an informed statement.

In the next step the flame propagation rate was determined by analyzing the brightness of each frame of high-speed recording which was integrated to one single line of a frame (using the software "Avicor"). The single lines were put in series to a new image. Resulting images of the examples are shown in Figure 7 and Figure 8 with time scale on the x-axis and frame height on the y-axis. The brightness is coded in a color.

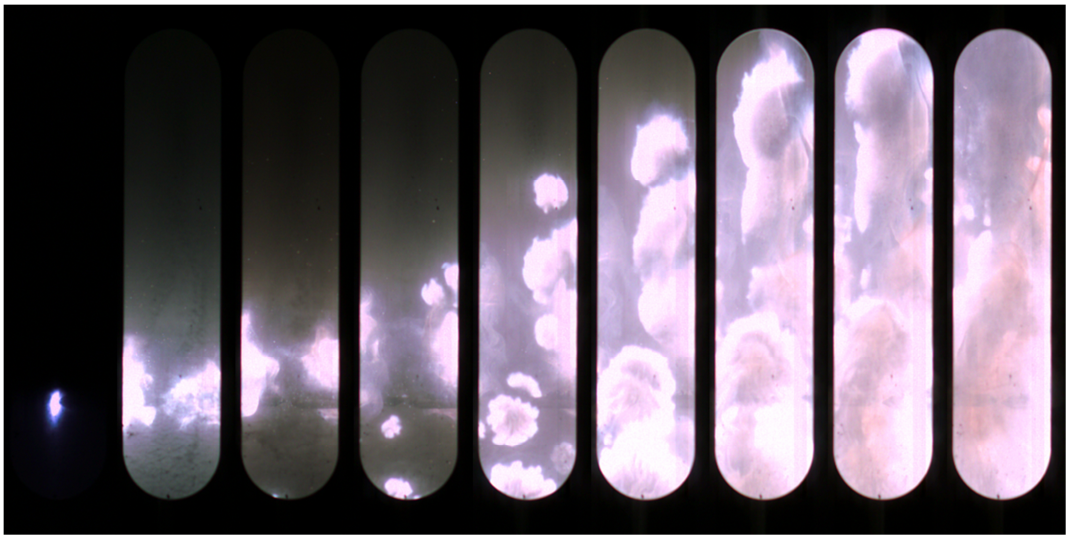

Figure 5: Series of aluminum particles combustion in the modified Hartmann-Apparatus.

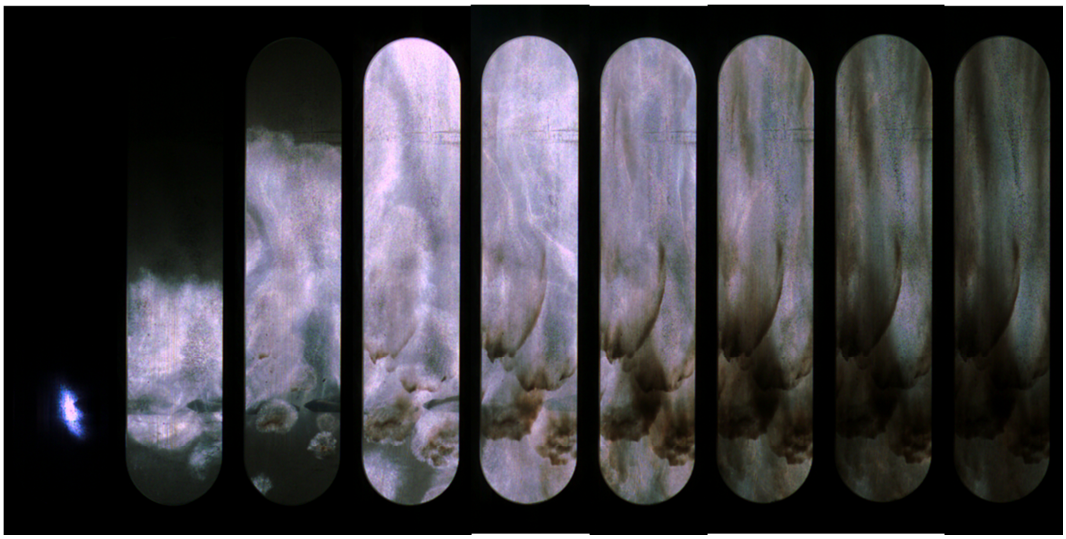

Figure 6: Series of magnesium particles combustion in the modified Hartmann-Apparatus.

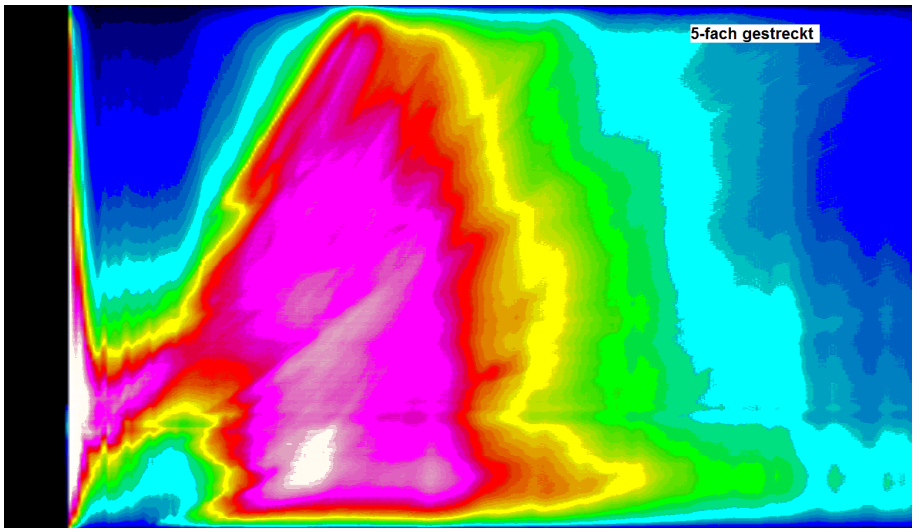

Figure 7: Evaluation of the brightness distribution of the single images in the case of aluminum particles (500 g/cm³).

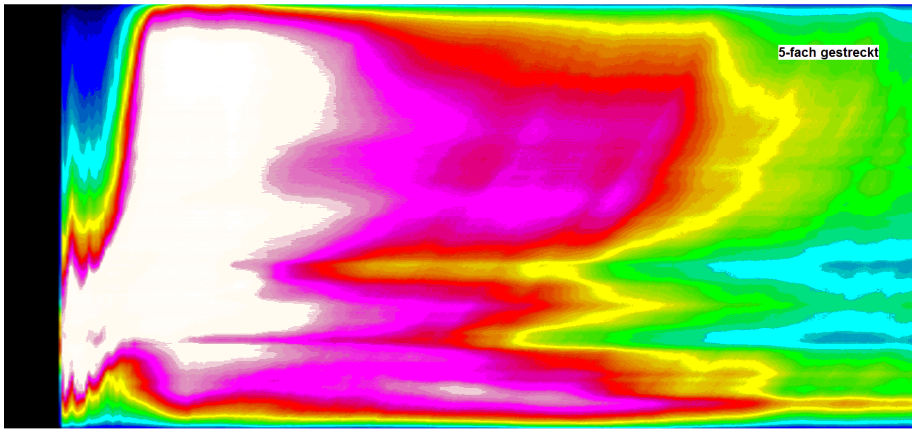

Figure 8: Evaluation of the brightness distribution of the single images in the case of magnesium particles (833,3 g/cm³).

Using the transition between green and yellow region of the flame spread, the flame velocity is determined by Equation 4 (7):

Equation 4

$$v = \frac{\Delta h}{\Delta t} = \frac{h_{ges} * \Delta px_y}{px_y} * \frac{fps}{px_x}$$

With h = covered track, Δt = duration of flame propagation, h_{ges} = total height,
px_y = number of pixels in y direction, px_x = number of pixels in x direction,
fps = frames per second.

Figure 9 shows the calculated flame velocity against the air ratio. Partly results were already shown in (1).

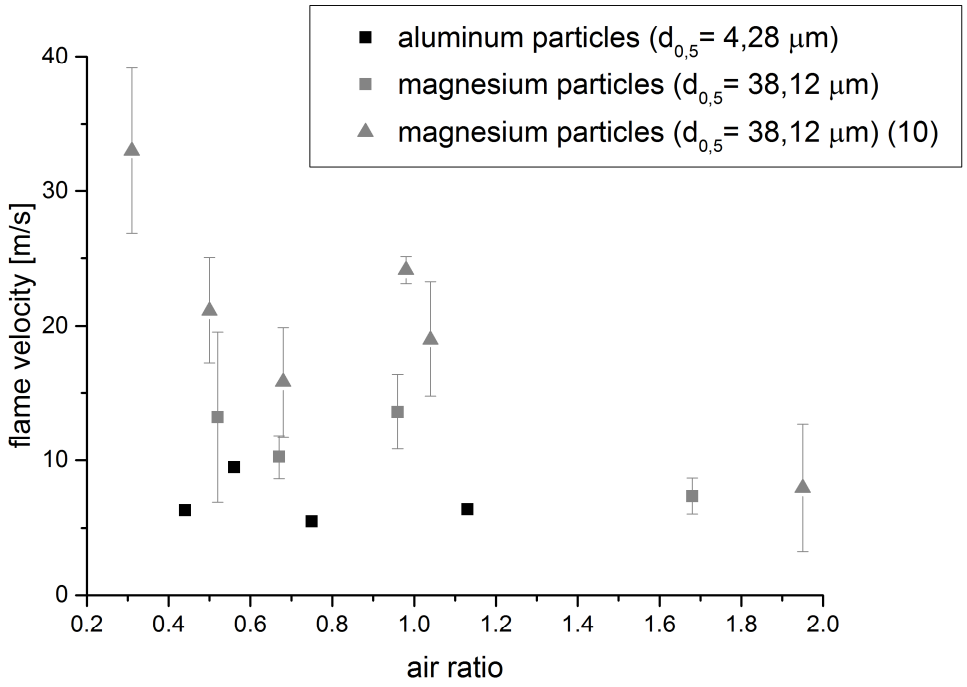

Figure 9: Flame propagation velocity dependent of air ratio (black: aluminum particles; grey: magnesium particles; triangles already published in (1)).

In the case of magnesium the combustion velocity shows a dependence of the air ratio: The lower the air ratio the higher the combustion velocity.

In the case of aluminum the combustion velocity shows no clear dependence on the air ratio. The reason for this could be the small amount of data points we could calculate from the tests.

If the calculated flame velocities of aluminum are compared with those of the magnesium particles, it is clearly visible that the magnesium particles show a higher flame velocity over the selected air ratio area.

Conclusion

The experiments with the modified Hartmann-Apparatus showed that both, aluminum and magnesium, are flammable in a defined dust-air-mixture. Magnesium particles show a higher flame propagation rate than aluminum particles in the selected air ratio range between 0.2 and 2.0. Due to the inhomogeneous combustion of the aluminum particles in the present experiments the received data of flame velocity is very rare. Therefore the increasing flame propagation rate with decreasing air ratio could not be observed for aluminum particles. More experiments have to be done with respect to the influence of material (e.g. carbides), particle size distribution, ignition delay and dust concentrations to get a wide data base for large scale experiments.

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