

# Determination of the Self-ignition Behaviour of Bulk Materials from Heat Storage Tests below Atmospheric Pressure

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Is a displacement of the self-ignition temperature up to higher temperatures possible by a pressure reduction? Therefore, the influence of an air pressure reduction on the self-ignition temperature of dust bulks is investigated. First, major effects which might influence the self-ignition temperature and the combustion behaviour of dust bulks under reduced pressure are described. Based on these theoretical considerations, basic experiments are carried out to determine the self-ignition temperature under reduced pressure. Thereby, the determination of the self-ignition temperature is carried out by isoperibolic heat storage tests according to DIN EN 15188:2007. The results of the investigations show that self-ignition temperatures of different bulk materials shift to higher temperatures within a pressure reduction.

## 1. Introduction

Drying of products is an essential process step in the chemical industry. Here, the vacuum drying is an often used method. To run the drying apparatus efficiently with respect to space-time yield, the drying time must be kept as short as possible. This can be achieved by using a high drying temperature, as far as the stability of the product allows this procedure. With increasing drying temperature, however, the risk of a spontaneous self-ignition of the bulk material rises. Spontaneous combustion is caused by the oxidation reaction of oxygen molecules on the particle surface of the solid bulk. The resulting self-ignition occurs due to insufficient heat dissipation itself. The phenomenon of self-ignition of bulk materials was extensively studied in the past in terms of a variety of factors, such as the sample geometry, the porosity of the bulk, the air convection around the bulk etc.. In industrial environments, most commonly performed preventive measures to avoid a self-ignition while drying are currently the temperature reduction and the reduction of the oxygen content in the drier air through partial inertization. Alternatively, reducing the oxygen volume concentration can be achieved by a reduction of pressure. By reducing the pressure, different questions arise regarding the self-ignition behaviour. Thereby, two different approaches might be possible:

By the reduction of the pressure, the oxygen volume concentration will be reduced. Due to the oxygen reduction, the reactivity and thereby the heat production in the bulk material will be inhibited which should lead to a displacement of the self-ignition temperature to higher temperatures. If this assumption is correct, releasing higher drying temperatures for vacuum dryers also becomes possible from a process safety perspective.

On the other hand, an effect accompanying pressure reduction is a decreasing thermal conductivity of the bulk material. Due to the pressure reduction and a decreasing number of molecules, heat dissipation in the bulk material will not be as good as at ambient pressure, which might lead to a decreasing self-ignition temperature and a more critical state with regard to the self-ignition of the bulk material.

The aim of the investigation of the self-ignition behaviour under reduced pressure is to find out which of the described effect will dominate.

## 2. Theoretical introduction

### 2.1 Theory of the self-ignition

An initially self-heating and subsequently probable spontaneous combustion is caused by the exothermic gas-solid reaction of oxygen molecules on the surface of each dust grain. At ambient temperatures, these

reactions proceed across the dust layer, since the cavities of the bed are filled with air. Due to this exothermic process, heat is produced throughout the bulk.

A self-ignition will occur, when the heat production within the bulk is greater than the heat dissipation. In the combustion reaction, the molecules need to contact one another in order to react with each other. The more oxygen molecules react with each solid grain within a certain time interval, the greater the rate of combustion will be. Consequently, the combustion rate is highly dependent on the concentrations of the flammable substance and oxygen (Rempe and Rodewald, 1993).

## 2.2 Heat transfer in bulk material

The thermal conductivity in bulk materials is made up of different types of heat transfer and is therefore also called effective thermal conductivity (see Eq(1)). It consists of the thermal conductivity of the particles ( $\lambda_p$ ), the thermal conductivity of the fluid ( $\lambda_f$ ) and the bulk porosity ( $\psi$ ). The thermal conductivity by radiation can be neglected at low temperatures.

$$\lambda_{effektiv} = f(\lambda_p, \lambda_f, \psi) \quad (1)$$

The thermal conductivity of the fluid phase (gas) in a bulk is pressure dependent and can be calculated with the Knudsen equation (Eq(2)) (Heinemann et al., 2007).

$$\lambda_{gas} = \frac{\lambda_{gas,0} \cdot \psi}{\left(1 + \frac{2}{p}\right)} \quad (2)$$

In Eq(2)  $\lambda_{gas,0}$  is the thermal conductivity of the free gas in  $W/(m \cdot K)$ ,  $\psi$  the porosity,  $p$   $\frac{1}{2}$  the gas pressure at half of the full gaseous thermal conductivity (Pa) and  $p$  the gas pressure (Pa). By the pressure reduction, the number of molecules, which are responsible for the heat transport, is reduced. The mean free path of the molecules is thereby antiproportional. Both effects, reducing the number of particles and the increase of the mean free path compensate one another at first, so that a pressure reduction does not affect the thermal conductivity of the gas noticeably down to a pressure of approximately 300 mbar. An influence is achieved, when the mean free path of the gas molecules is equal to the pore size of the dust layer, since this circumstance has a direct effect on the thermal conductivity of the bulk (Meyer and Schiffner, 1989) (see Figure 1). The thermal conductivity of the solid particles ( $\lambda_p$ ) will not be influenced by a pressure reduction. This theoretical approach could be confirmed by measuring the thermal conductivity of a carbon black bulk at different pressure stages (see Figure 1) during experiment with the Hot Disc Method. The results are shown in Figure 1.

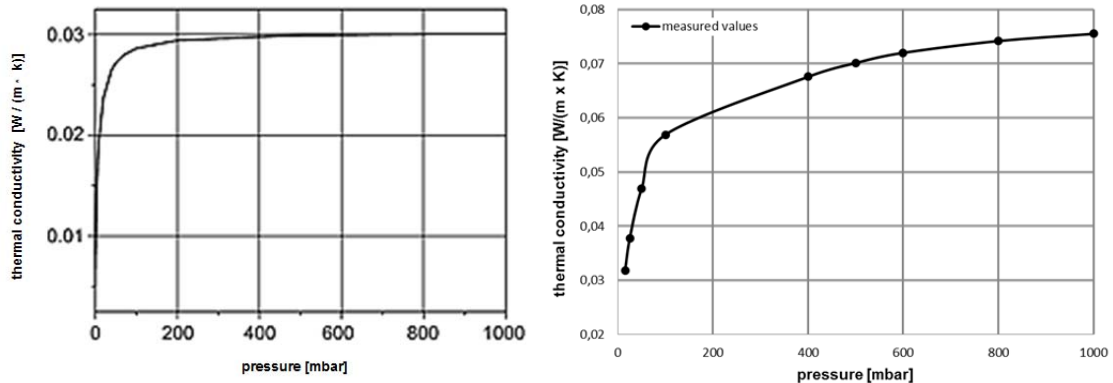


Figure 1: left picture: Theoretical approach of the pressure dependence of the thermal gas conductivity by Smoluchowski (Heinemann et al., 2007). Right picture: Experimentally determined thermal conductivity of a carbon black bulk at different pressure stages.

It could be shown, that in accordance to the theory, the effective thermal conductivity of the bulk material decreases within a pressure reduction. Due to the fact that the thermal conductivity of the particles is not influenced, the reduction is attributed to the reduced thermal conductivity of the gas phase. Yet, whether this effect represents the main part for a displacement of the self-ignition temperature remained to be investigated in experiments.

### 2.3 Dependence of oxygen volume fraction with regard to the self-ignition temperature

Schmidt et al. (2002) already investigated the spontaneous ignition behaviour of dust and solids under reduced oxygen volume fraction. Thereby, the oxygen reduction was achieved by a partial inertization of the drying chamber at ambient pressure. In this study, various substances, various  $V/A$  - ratios and the burn-off behaviour of the bulk were analysed. The authors came to the conclusion, that by reducing the oxygen volume fraction, the auto-ignition temperature can be shifted to higher temperatures. The displacement of the self-ignition temperatures is not uniform and different for each test substance. The self-ignition temperature remains dependent on the geometric constraint conditions ( $V/A$  ratio). The time of complete burning off extended with decreasing oxygen volume fraction. A comparison of the results of Schmidt et al. and the results of this paper is useful since both methods, partial inertization as well as a pressure reduction, cause a decrease of the oxygen volume concentration.

## 3. Experimental investigation

### 3.1 Determination of the self-ignition temperature

The determination of the self-ignition temperature of various test items was investigated by isoperibolic heat storage tests, using a 1000 ml wire basket (cube) according to DIN EN 15188 and UN Transport Regulation Test N.4. Here, the test items were held at a specific storage temperature for 24 h in a convective hot-air type chamber with an inner volume of 115 L. The test is considered positive in terms of self-heating, when the test item was heated above the set storage temperature during the experiment by at least 60 K. For the investigation of the influence of a reduced pressure, the isoperibolic heat storage test was used. Due to the changing conditions by different pressure stages, the criteria of self-ignition, 60 K temperature increase, was used but the time of temperature increase (24 h) was not considered.

### 3.2 Experimental equipment

To determine the self-ignition temperature a vacuum drying chamber was used. This included an air inlet at the bottom and an air outlet in the upper part of the chamber. The airflow should be previously preheated by means of copper tube. The data acquisition must be carried out with suitable software to determine the self-ignition temperature within the bulk. In the standard, it is mentioned that alternative test configurations may be used to achieve the required test conditions. For the experimental determination of the self-ignition temperature, a vacuum drying chamber from Binder GmbH<sup>®</sup> with an inner volume of 115 L was used. The drying chamber is subsequently optimized with a PT 100, and seals that are suitable for temperatures up to 600 °C. This optimization was necessary because the investigations of the self-ignition temperature combustion can cause high temperatures inside the drying chamber. In order to detect the incoming air stream, a flowmeter is installed in front of the air inlet valve. This is designed for flow rates from 0 to 500 l/h. The flowmeter is powered by the compressed air network. The compressed air has a constant pressure of approximately 8 bar<sub>g</sub> and is dry fed into the compressed air network. The use of dry air is an important requirement in the determination of the self-ignition temperature since the relative humidity has also an influence on the self-ignition temperature (Lohrer, 2005). To safeguard the apparatus against pressure oscillation, a pressure reducer was installed upstream, thereby ensuring that the air flow is defined and stationary for the flowmeter. For the reduction of air pressure within the drying chamber, a vacuum pump of Vacuubrand<sup>®</sup> (PC 3000 series) was provided with a rated output of 4.9 m<sup>3</sup>/h. In order to keep the pressure loss as low as possible, a metal pipe from the output of the vacuum chamber downstream off the pump was made, equipped with a 50 cm long cooling jacket. The cooling jacket is to cool down any hot gases that could arise. Thus, overheating or damage to the vacuum pump is avoided. Figure 2 shows the experimental setup for the investigation of the self-ignition temperature under reduced pressure.

### 3.3 Experimental investigations and results

For the determination of self-ignition temperature under reduced pressure, the substances Carbon Black (Norit CN 4) and wheat flour (type 405) were investigated at different pressure stages.

The self-ignition temperature could be shifted to higher temperatures in the experiments by way of pressure reduction (Figure 3 and Figure 4). The error bar of the determined self-ignition temperature is +/- 2 K. By the studies of the two test substances it could be overserved, that the shifts of the self-ignition temperatures were not uniform with regard to pressure reduction.

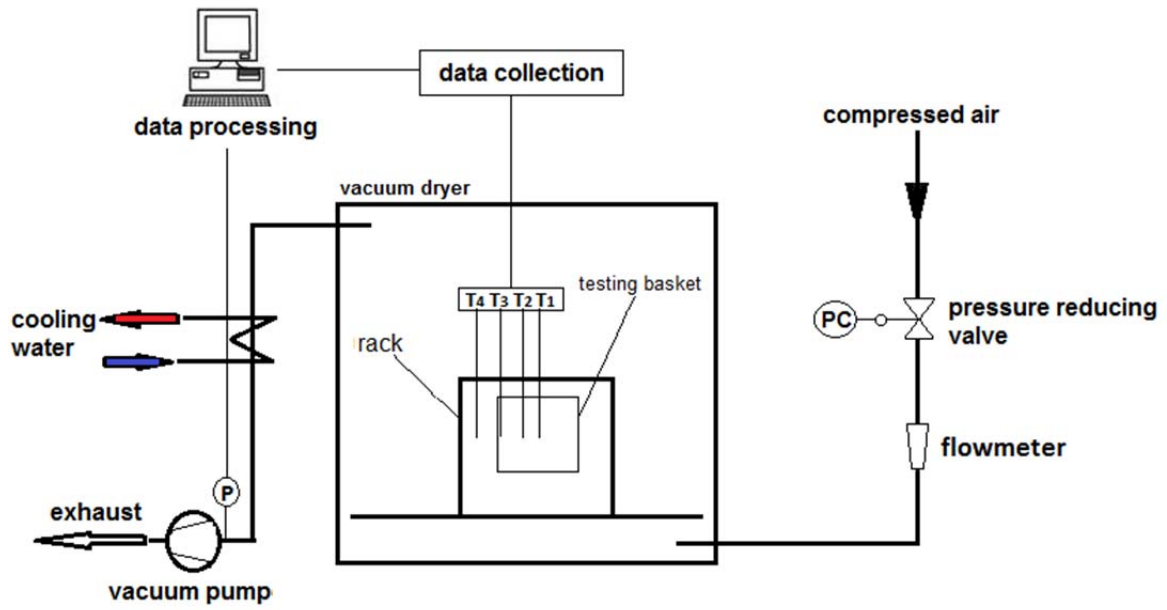


Figure 2: Experimental test set up for the investigation of the self-ignition behaviour under reduced pressure (consilab set up)

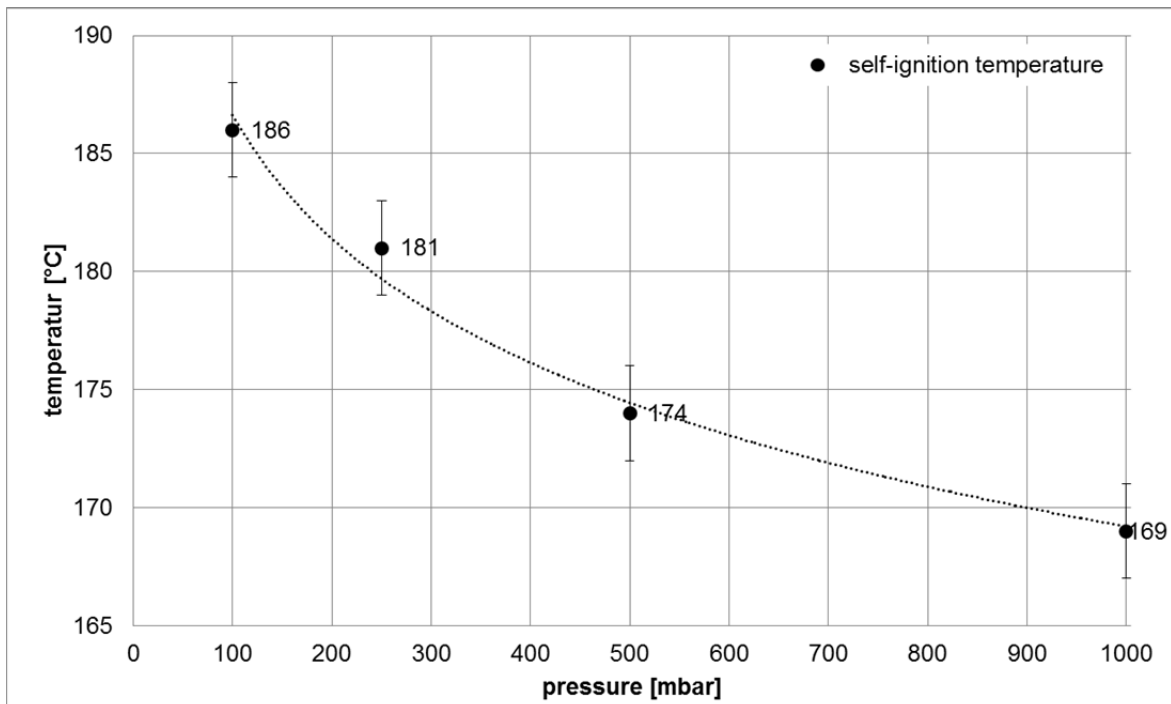


Figure 3: Dependence of the self-ignition temperature at different pressures stages for wheat flour.

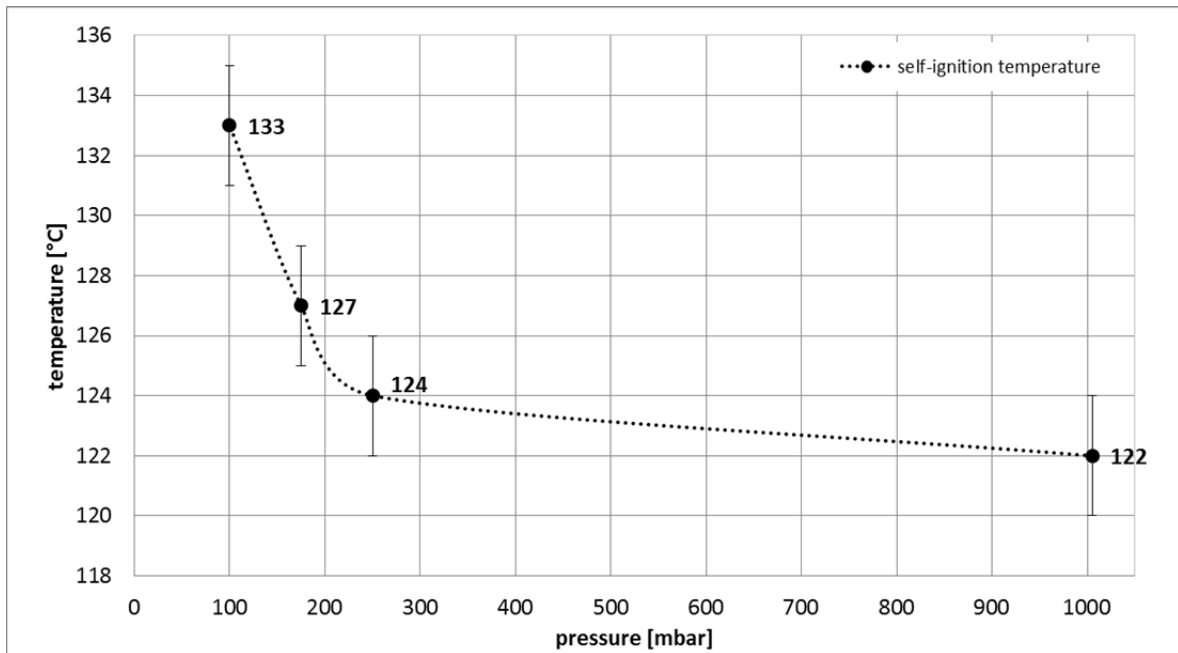


Figure 4: Dependence of the self-ignition temperature at different pressures stages of carbon black.

It could also be observed, that the pressure reduction does not prevent self-ignition. At a reduced pressure, self-ignition could also take place whereby the combustion behaviour of the bulk material is slowed down considerably as a function of oxygen volume concentration. These circumstances could be observed by both investigated materials. Furthermore, the knowledge was obtained that the air flow rate also has an impact on the combustion behaviour. This is largely responsible for the transport of oxygen to and into the bulk material. In Figure 5 two different courses of combustion after self-ignition of wheat flour at different pressure stages (995 mbar and 100 mbar) are shown. First, the drying of the substances could be seen. Thereby, the resulting drying plateau (approximately 45 °C at 100 mbar and 70 °C at 995 mbar) is lower at lower pressure. Subsequently, the self-ignition behaviour could be detected. The course under a reduced pressure showed a slower combustion in comparison to the combustion near ambient pressure.

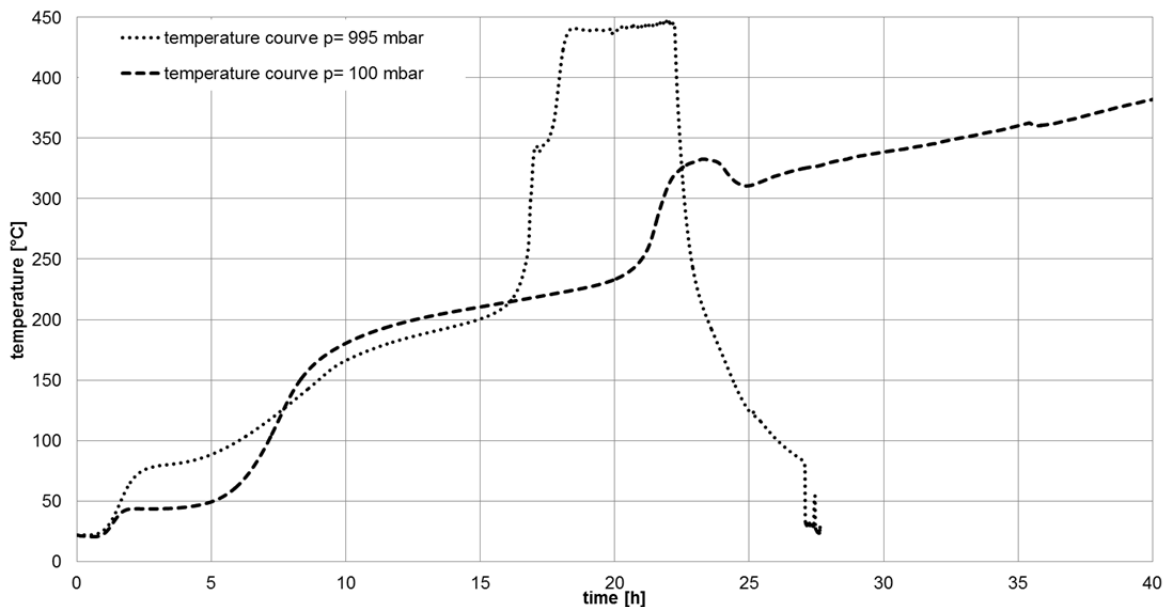


Figure 5: Combustion behaviour of carbon black at different pressures stages

#### 4. Conclusion

The general theoretical principles and the experiments show that the thermal conductivity of the fluid phase within the bulk material is reduced by pressure reduction which leads, on the whole, to lower thermal conductivity of the bulk material at lower pressures. After diminishing the oxygen concentration by pressure reduction, an increase of the self-ignition temperature could be observed. Therefore, it can be stated, that it is the heat production in the bulk material that plays the principal role for self-ignition. The decreasing thermal conductivity can be neglected on basis of the present studies.

The combustion behaviour of an ignited bulk material has the same properties as those already observed for partial inertization by Schmidt et al.. The course of the complete burning of the sample is slowed down by a pressure reduction.

Due to the slower combustion of the bulk at reduced pressures, the time needed for a complete burn-up of the bulk material increases. This leads to a smaller temperature difference between the bulk temperature and the storage temperature within a specific time span. Due to that fact, the temperature increase might not exceed 60 K within a time period of 24 h, which characterizes a self-ignition according to DIN EN 15188. Therefore, it must be pointed out, that the characterization of a self-ignition according to DIN EN 15188 is not sufficient to cover a self-ignition at reduced pressures. Thus, the experimental investigations lay in the border zone of this standard.

A self-ignition could not be completely avoided by pressure reduction. Up to now, the investigation did not show any pressure limit, beneath which a self-ignition did not take place, even though the results of the experiments point towards the existence of such a pressure limit. The results of Schmidt et al. (2002) already showed that the reduction of oxygen volume fraction can lead to an avoidance of the exothermic reaction within the bulk material, if the oxygen content is < 3 Vol.-%. Whether there is a pressure limit at which no self-ignition will be observed in bulk materials has yet to be determined in further experiments.

At this point, it must be clearly stated: If there is an oxygen intrusion into the dryer, while the bulk material is stored above its self-ignition temperature at ambient pressure, an ignition will start immediately, similar to a boiling retardation of fluid.

For the two test substances, wheat flour type 405 and carbon black, the self-ignition temperature could be increased by a reduction of pressure. The change of self-ignition temperature is not uniform for the products, but substance-specific. So far, the results of different substances showed no general rule for the shift of the self-ignition temperatures. Therefore, more fundamental research using pure, synthetic substances is required and being carried out in our laboratories.

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