Emission Control of Particles and Gaseous Pollutants with a High-Temperature Granular Bed Filter†

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Abstract

The combined collection of fine dusts and the sorption of gaseous pollutants at high temperatures have been experimentally and theoretically investigated. The results of the fractional separation efficiency measurements and the experimentally determined breakthrough curves have revealed that excellent \( \text{SO}_2 \) and \( \text{HCl} \) sorption characteristics exist under conditions which also yield high particle collection efficiencies. New concepts have been developed to describe the measured fractional collection efficiency curves in the inertia dominated size range, which are based on particle trajectory calculations using a simple flow model. During the sorption, conversions of over 80% can be achieved when using a reactive bed consisting of pellets agglomerated from fine limestone particles. The application of such beds in a counter-current mode can even yield solid conversions of nearly 100%. The mathematical prediction of the conversion time behaviour under differential conditions and the breakthrough characteristics in a fixed bed are in reasonable agreement with the experimentally measured data.

1. Introduction

The application of granular bed filters for hot gas cleaning has become an increasingly popular topic. The use of reactive bulk media additionally opens up the possibility of combining the filtration with a process which extracts specific gases by means of catalytic conversion or sorption within the bed. The industrial realization of such simultaneous particle collection and gas sorption processes will then offer small and average-sized plants a technique with which hot flue gases can be effectively cleaned, even at temperatures of up to 1000°C, whilst functionally exploiting the waste heat involved.

The particle collection behaviour is characterized by means of systematic fractional separation efficiency and pressure loss measurements in dependence of the chief operational parameters. The sorption process of gaseous components is characterized with the aid of experimentally determined conversion time curves under differential conditions, and by the evaluation of breakthrough curves of the acidic gaseous components \( \text{SO}_2 \) and \( \text{HCl} \) using an integral reactor. The investigations were conducted at temperatures of up to 800°C.

2. Results of the particle collection measurements

Systematic separation efficiency measurements at room temperature for particles in the size range between 0.01 \( \mu \text{m} \) and 10 \( \mu \text{m} \) have allowed the influence of the chief filtration parameters (e.g. filter face velocity, filter thickness, type and size of the bed grains and the electrostatic charging of the up-stream particles) to be extensively quantified. The influence of the filtration duration is of decisive importance to the assessment of such a filter's operational behaviour. In general, it may be assumed that the separation improves with the quantity of dust collected. Nevertheless, unfavourable operation (filter medium grain size \( d_K \geq 1 \text{ mm} \), filter face velocity \( u_0 > 0.2 \text{ m/s} \)), in conjunction with high dust concentrations can lead to a breaching which consequently causes the filtration characteristics to deterio-
rate enormously. In view of both the separation and the pressure-loss (and hence energy consumption), the room-temperature collection would appear more favourable at low filter face velocities \( u_0 < 0.1 \text{ m/s} \) and with small bed grains \( d_K < 0.5 \text{ mm} \). This is demonstrated in Fig. 1, in which the mass-specific penetration \( E_3 \), calculated from the measured fractional efficiencies, and the inlet particle size distribution has been plotted for three different filter medium grain sizes as a function of the measured pressure drop. One can observe that after a short run-up period, the finest grain bed shows the highest total separation efficiency for any specific pressure loss and thus, the lowest emission values.

The influence of the temperature on the particle collection has been investigated up to temperatures of 800°C. Theoretically, the transport conditions for extremely small particles \( (x < 0.5 \mu\text{m}) \) improve towards the grain's surface with increasing temperature as a result of the increasing particle diffusion coefficient. Conversely, a poorer collection performance is to be expected with increasing temperature for larger, inertia-dominated particles \( (x > 0.5 \mu\text{m}) \) due to increasing drag forces, since the dynamic viscosity of the fluid increases with the temperature, hence impairing the transport. This anticipated theoretical temperature influence was verified for particles in the inertia dominated regime by experimentally determined grade efficiency curves.

The high-temperature, long-term filtration influence (and therefore the effect of the filter loading) is demonstrated in Fig. 2. In addition to the fractional separation efficiencies, this graph also incorporates the mass-specific total separation efficiency \( E_3 \) calculated from the grade efficiency and the raw gas particle distributions, together with the pressure loss \( \Delta p \), measured for each respective dust loading. Despite relatively coarse bed grains \( d_K = 0.66 \text{ mm} \) and a comparatively high filter face velocity \( u_0 = 0.25 \text{ m/s} \), the fractional separation efficiency increased extremely rapidly, to remain at nearly 100% for all particle grades.

At ambient temperatures, similarly good collection characteristics could only be attained from bed grains smaller than 0.3 mm with significantly lower filter face velocities. The more favourable dynamic behaviour at higher temperatures is to be traced back to an improved adhesion to the individual surfaces of...
the grains. Under such conditions, a dense dust layer forms on the filter's surface in a manner similar to the dust cake of a bag filter, so that even for substantially higher operational raw gas dust concentrations of \(c_e = 2.6 \text{~g/m}^3\), clean gas values of less than \(1 \text{~mg/m}^3\) may be achieved under standard conditions. As a result, the filter face velocity of granular bed filters may be substantially increased at higher temperatures. This will allow the filter area to be correspondingly reduced, cutting both the filter unit's required floor space and investment costs, although at the expense of higher pressure losses.

3. Theoretical calculation of grade efficiencies

Model concepts already exist with which the separation curves for dusts within the diffusion range may be adequately predicted\(^3\), provided that the influence of the deposited dust mass may be neglected (i.e. for short filtration periods). In the inertia range, however, this is only possible with extensive restrictions. As a result, a new model concept has been developed which describes the collection process as a combination of fluid flow through the pores of the filter bed and stagnation flow around a spherical collector. With the aid of particle trajectory calculations, fractional separation efficiencies could be calculated which, in practice, are in good agreement with measured efficiencies. This is demonstrated in Fig. 3 for a limestone bed and particle sizes between 0.01 \(\mu\text{m}\) and 10 \(\mu\text{m}\). In addition to the measured fractional separation efficiencies (dots), the curves of Tardos\(^4\) and Goren\(^5\) have been included, together with the results yielded by the pore model. The calculated grade efficiencies coincide not only at ambient conditions with the measured data, but also exhibit a reasonable agreement with the experimentally determined curves at elevated temperatures. In contrast, however, the theoretical description of the filtration kinetics is extremely sophisticated, and has not yet been accomplished.

4. Results of the Sorption of SO\(_2\) and HCl

The investigations into the sorption of SO\(_2\) and HCl have verified that the operational conditions relevant to the particle collection will also yield an effective pollutant gas sorption. The sorbents used incorporated raw limestone and pellets agglomerated from fine limestone dust. The size of this parent dust proves to be a key parameter for the achievement of high solid conversion rates and hence, maximal sorbent exploitation. Figure 4 exemplarily illustrates breakthrough curves for SO\(_2\) for various parent grain sizes through granular beds of limestone pellets (agglomerated using a rotary disk unit to a size of \(d_K = 1.9 \text{~mm}\)) at a filter face velocity of \(u_o = 0.25 \text{~m/s}\) (air as carrier medium).

![Fig. 4 Influence of the parent grain size \(x_{S0,3}\) on the penetration behaviour of SO\(_2\) for three different limestone pellet beds (pellet size \(d_K = 1.9 \text{~mm}\)) at a filter face velocity of \(u_o = 0.25 \text{~m/s}\) (air as carrier medium)](image-url)
gas concentration $C_0 = 1500$ ppm) then, in the case of the finer fraction $F_1$, 70% of the solids may be converted before an effective filtration ceases after 7 hours of operation.

The actual size of the pellets themselves is only of subordinate relevance. This becomes evident from Fig. 5, which illustrates the breakthrough characteristics of HCl through beds of two different raw limestone fractions and agglomerated pellet sizes. In this instance, the pellets were fabricated from limestone $M$ 1000 powder ($X_{50.3} = 9.8 \mu m$) and the filtration was conducted at 500°C. In the case of the raw limestone, it is evident that the attainable solid conversions are by no means adequate for industrial purposes. This does not stem from a financial assessment (since in comparison with other sorbents such as Ca(OH)$_2$, limestone is indeed a cheap raw material), but rather from the problems involved in disposing of the residues.

At temperatures of around 500°C, a comprehensive sorbent exploitation may be attained by utilizing limestone pellets, which offer average solids conversions of over 90%. As a result of the low porosity of raw limestone ($\varepsilon_s = 9.5\%$), the diffusive transport into the grain is not only impaired from the beginning of the reaction, but ceases completely at low conversion ratios due to the formation of product layers. The limestone pellets, however, possess a larger, more easily accessible internal surface, so that an almost total reaction can occur. Even at high solid conversions, the large pore volume of the pellets ($\varepsilon_s = 45\%$) still warrants a diffusive transport into the core of each sorbent particle.

The theoretical description of the single grain sorption kinetics (differential conditions) for HCl is accomplished by means of an elementary shrinking-core model. The sulfation reaction, however, requires the application of a model which also takes into account the pellets' internal pore radius distribution. The layout of a granular bed filter depends on the solution of the mass balance for the packed bed reactor, which may be accomplished using two different mathematical procedures. One can either solve the partial differential equation under consideration of the axial dispersion using a finite difference method, or apply the method of characteristics, in which axial dispersion is neglected. Since both approaches deliver nearly identical solutions, the influence of the axial dispersion can be excluded. The comparison between measured and calculated breakthrough curves for both HCl and SO$_2$.
yield an adequate conformity.

The calculated solid conversion profiles within a 20-cm-deep CaO pellet bed for SO₂ (inlet concentration \( C_0 = 1500 \) ppm) is demonstrated in Fig. 6. The kinetic parameters for the reaction model (i.e., reaction rate constant and the gas diffusion coefficient through the product layer) were implemented with those values with which both the temporal single grain solid conversion behaviour (measured under differential conditions), and the breakthrough characteristics (measured in an integral reactor) could be described.

The operational life of even relatively thin beds is in the order of many hours. In spite of the fact that the CaSO₄ product layer which forms during the reaction progressively retards the reaction, the solids may nevertheless be practically totally converted. The steep solid conversion profiles allow non-converted bed substance to remain available to the reaction, attaining low emission values. The countercurrent application of this technique hence gives access to an unique possibility of being able to practically totally exploit the sorbent of a dry flue-gas cleaning process. This is especially important with respect to the increasing necessity of minimizing industrial waste.

A comparison between the results of the particel collection and the sorption of the gaseous components has revealed that the sorption of the gases is indeed possible under similar operational conditions to the filtration of the dispersal phase. The mutual influences between the sorption and the particle collection, incited by bed structure modification through the chemical reaction or the lodging of inert dust, are generally insubstantial. This has been verified by means of separation efficiency measurements with both fresh and fully exhausted pellets, and from measured breakthrough curves through beds loaded with inert quartz dust.

**References**

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