PARTICLE BREAKAGE IN THE CYCLONES OF FLUIDIZED BED SYSTEMS

Marvin Kramp* Anja Püttmann† Andreas Thon‡
Ernst-Ulrich Hartge** Joachim Werther††

*Hamburg University of Technology, m.kramp@tuhh.de
†Hamburg University of Technology
‡Hamburg University of Technology
**Hamburg University of Technology
††Hamburg University of Technology

PARTICLE BREAKAGE IN THE CYCLONES OF FLUIDIZED BED SYSTEMS

Anja Püttmann, Marvin Kramp, Andreas Thon, Ernst-Ulrich Hartge, Joachim Werther
Institute of Solids Process Engineering and Particle Technology, Hamburg University of Technology
Denickestrasse 15
21073 Hamburg, Germany

ABSTRACT

A breakage model is developed to predict the change in particle size distribution of brittle materials that undergo mechanical stress in gas cyclones. The breakage probability depends on the cyclone inlet gas velocity, solids load, particle size, and a material-specific constant. The model is validated with data from experiments on a laboratory gas cyclone with aluminum oxide and iron oxide. The results are applied to calculate the particle size distribution of a chemical looping combustion process with iron ore as oxygen carrier.

INTRODUCTION

Minerals are increasingly treated in fluidized bed systems. The application of interconnected fluidized beds in mineral processing is well established, e.g. alumina calcination or iron ore direct reduction (Lurgi Circored and Circofer process) (1). Recently fluidized bed systems are also employed in new technologies to reduce the emission of carbon dioxide from fossil fuel combustion into the atmosphere. The chemical looping combustion (CLC) process with inherent carbon dioxide capture is composed of two fluidized beds with a metal oxide traveling as an oxygen carrier between the air and the fuel reactor (2). Carbonate looping is a technology for post-combustion CO_2 capture from power plants by means of CaO in a system of two fluidized bed reactors (3).

All these fluidized bed systems involve gas cyclones for solid-gas separation. Cyclones have been identified as a major source of mechanical stress on the particles causing particle attrition or breakage (4). The main consequence of comminution in fluidized bed processes is the reduction of the average particle size and the generation of fines that cannot be kept in the system and lead to a loss of valuable material. The particle size distribution (PSD) determines all relevant mechanisms in a fluidized bed system, such as elutriation and entrainment (5), conversion and selectivity of reactions (6), and heat transfer from bed to inserts (7). Furthermore the quality of the solid product can be directly affected (e.g. in mineral processing).

In order to predict the quantitative extent of breakage in fluidized bed processes a breakage model has been developed that can be applied in the context of population balances. The breakage function is based on the approach by Hill und

Ng (8, 9) and is assumed to depend on material properties only. The breakage probability, however, is also accounted for by physical variables such as the gas velocity in the cyclone inlet, solids volume concentration, and particle size. Thus the breakage model can be extrapolated to describe attrition behavior under different operating conditions.

To determine the model parameters attrition tests for two brittle minerals, aluminum oxide and iron oxide, were carried out in a laboratory sized gas cyclone. Conducting the tests in the technical apparatus under consideration ensures that the applied stress mechanism is identical with the one that leads to breakage in the technical fluidized bed system.

The breakage model has been implemented in the flowsheet simulation tool SolidSim (10) and applied to the calculation of the solids inventory of a CLC process with iron ore as oxygen carrier.

THEORY

In computer-aided simulations of processes containing solids processing steps population balance equations are used to follow the evolution of the PSD from one equipment unit to another. This is essential for process design and optimization. The continuous number-based population balance equation that describes breakage phenomena is

$$\frac{dn(v)}{dt} = \int_v^\infty b(v,w)S(w)n(w)dw - S(v)n(v) \quad (1)$$

where *n(v)* is the number density function. This breakage equation states that the rate of change of the number of particles between particle volume *v* and *v+dv*, *n(v)dv*, is the net result of generation due to breakage of particles larger than *v* and loss of particles of size v by breakage into smaller sizes. The number-based specific rate of breakage, *S(v)*, is the number fraction of particles broken from volume *v* per unit time. The number-based breakage function, *b(v,w)dv*, is the number of particles formed between *v* and *v+dv* divided by the number of particles broken from volume *w*. Hill and Ng (8) present a discretization method for the breakage equation that provides an output discretized PSD that has the same zeroth moment (total number of particles) and first moment (total mass of particles) as that obtained from the continuous population balance equation.

The experimentally observable breakage function is expressed as a linear combination of theoretical breakage functions for various numbers of child particles (9):

$$b_{ij}^{comp} = \sum_p w_p b_{ij}(p) \quad (2)$$

where ε_p are weighting factors expressing the probability that *p* child particles are formed when one particle is broken. The number-based breakage function, b_{ij}, is the number of child particles that fall in interval *i* from breakage of a particle in interval *j*.

For the breakage rate, S_i, the approach is based on the work of Weichert (11). He assumes that material defects which are statistically distributed in size and location on the particle surface determine the particle strength. It is more likely to find defects on the surface of larger particles than on small ones which explains the higher mechanical strength of small particles. Based on Weibull statistics and breakage mechanics he proposed for the breakage rate of round particles

$$S = 1 - \exp(-S_c \cdot d_p{}^2 \cdot W_m^z) \qquad (3)$$

Here S_c is a material constant, d_p denotes the particle diameter, and W_m is the mass-based specific energy with which the particle is pressed against a plane. For z Weichert (11) gives the relation $z = (2 + k)/5$ where k is a parameter that represents the distribution of defects on the particle surface.

With consideration of the stress acting on the particles in the cyclone details of which may be found elsewhere (12) the following relationship for the breakage rate S_i for a particle of the size dp_i is obtained:

$$S_i = 1 - \exp(-S_c \cdot d_{p,i}{}^2 \cdot u^z \cdot (1 - \varphi)) \qquad (4)$$

with the solids occupancy ratio in the strand that forms on the gas cyclone wall (13)

$$\varphi = \frac{b_{inlet} \cdot \mu_{inlet}{}^n \cdot \frac{\rho_g}{\rho_s}}{d_p \cdot (1 - \varepsilon) \cdot c} \qquad (5)$$

where b_{inlet} is the width of the gas cyclone inlet, μ_{inlet} denotes the solids load in the gas flow entering the cyclone. ε is the voidage ratio of the densest packing of particles. c and n are a model parameter with $0<c<1$ and $n>0$.

Relation 4 assumes that breakage only occurs due to particle-wall collisions.

EXPERIMENTAL

The experimental setup for the investigation of cyclone attrition is shown in figure 1. It was developed by Reppenhagen and Werther (14) to study attrition mechanisms inside cyclones. In this work, however, the experimental procedure was changed to investigate particle breakage caused by the mechanical stress in cyclones. The cyclone is operated in the suction mode. The gas flow in the tube attached to the cyclone inlet is loaded with solids which are added via a vibrating feeder. The separate feeding of solids allows for independent variation of gas inlet velocity, u, and solids volume concentration, c_v. Solids that are collected in the underflow hopper after their pass through the cyclone can be reintroduced as feed material by exchanging the underflow and feeder hoppers. The cyclone overflow is connected to a sieve and filter in series. The sieve has a mesh width of 25 µm. After each pass the sieve is disconnected from the cyclone and cleaned by sucking air through its bottom. This way all the fines that were still stuck to the sieve were transported through its meshes into the filter. After each pass a sample was taken from the underflow hopper and the filter was exchanged. Figure 2 gives an example of the experimental results obtained with iron ore. The mass density distribution of particle size is shown for the material before and after 15 passes for given operating conditions (gas velocity at inlet, u, and solids volume concentration, c_v). Initially the change in the mass density distribution of particle size is large. It rapidly decreases during the first cyclone passes and reaches a steady-state after about five passes. Weaker or pre-damaged particles break during the first passes until in the steady-state the ore is homogenous with respect to particle strength. It has been shown that the change in particle size in each cyclone pass after five passes can be described by one breakage matrix. For determining the breakage matrix the solids in the filter were distributed evenly among all particle size intervals smaller than 25 µm by calculation as after the cleaning procedure no particles were left on the sieve.

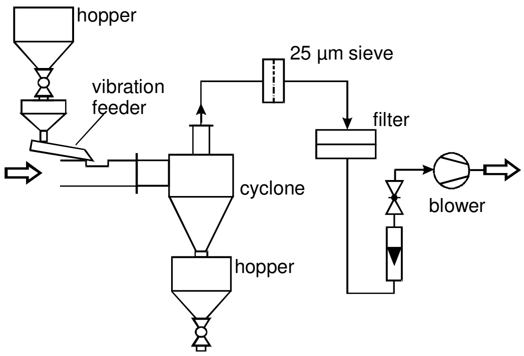

Figure 1: Experimental setup for breakage experiments (Reppenhagen and Werther, 14)

The gas cyclone had a main diameter of 90mm. Breakage experiments were conducted for aluminum oxide for particle sizes between 50 and 250 µm and iron oxide for particle sizes between 100 and 400 µm. For the experiments small fractions of the PSD were used to make the production of child particles visible. For iron ore most experiments were conducted with a particle size fraction of 250-315 µm. In the case of iron ore it is interesting to investigate coarser fractions: First, the feed material for the direct iron ore reduction is relatively coarse (up to 1 or 2 mm). Second, in the CLC process with solid fuel the particle size of the oxygen carrier must be considerably bigger than the size of the produced ash particles. This is necessary for mechanical separation of the ash from the oxygen carrier to remove it from the system while the oxygen carrier is recycled. Gas inlet velocities of 15, 20, 25, and 30 m/s were investigated. The solids load was varied between 0.1 and 2.5 kg/kg.

RESULTS AND DISCUSSION

Determination of model parameters

The model parameters were determined for two metal oxides, aluminum oxide and iron oxide. It was found that the change in PSD could be described best if theoretical breakage functions for small, a medium, and a high and very high number of child particles are combined. The performance of the composed breakage equation was insensitive to the specifically chosen number of child particles for the theoretical breakage functions. Equation 2 was specified to

$$b_{ij}^{comp} = w_1 b_{ij}(p=3) + w_2 b_{ij}(p=8) + w_3 b_{ij}(p=20) + w_4 b_{ij}(p=150) \qquad (6)$$

The values for the probability, w_p, with which three, eight, twenty, and 150 child particles are formed when a parent particle is broken are given in table 1 for two aluminum oxides and iron oxide. Table 1 also shows the material constants, S_c, in equation 4 for all materials that are used to calculate the breakage rate.

	w_1	w_2	w_3	w_4	S_c	n	c	z
equation	6	6	6	6	4	5	5	4
iron oxide	62.0%	15.8%	16.7%	5.5%	2.97E-11	0.5	0.2	3
aluminum oxide	76.0%	3.2%	18.8%	2.0%	9.00E-10			

Table 1: Experimentally determined model parameters

In order to simulate the experimental findings *n* in equation 5 was determined to 0.5 and *c* to 0.2. The exponent *z* for the gas velocity u in equation 4 was found to be 3.

Validation of the model

Figure 2 shows a comparison of the measured and the calculated mass density distribution for iron ore after being exposed to mechanical stress in the cyclone fifteen times. The results are presented for two different particle size fractions. It shows that the model can predict the change in the PSD well. In figure 3 the measured and calculated mass density distribution for aluminum oxide after fifteen exposures to mechanical stress in the laboratory gas cyclone is shown.

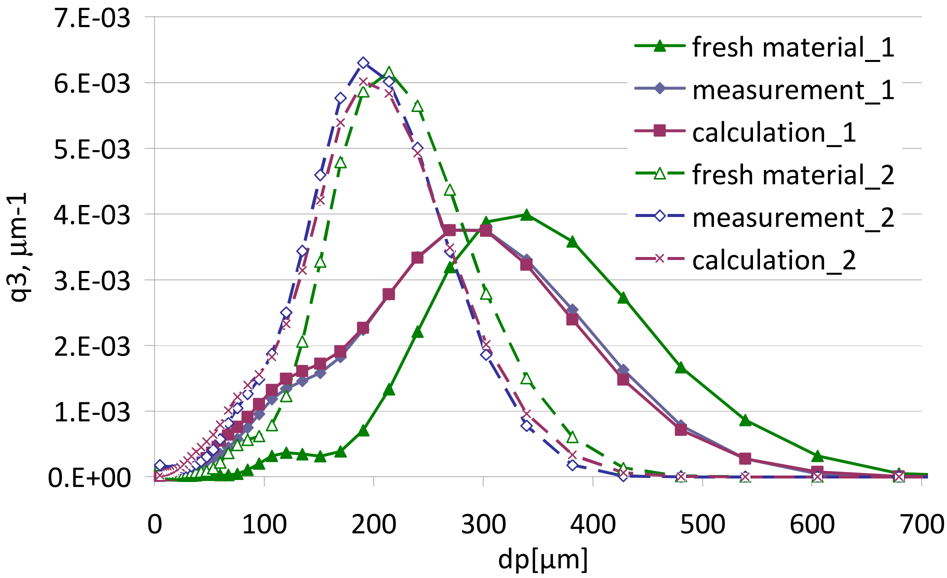

Figure 2: Measured and calculated mass density distribution after 15 passes through the gas cyclone for iron ore. Original size fractions: Index 1: 160-200µm, index 2: 250-315µm. Operating conditions: gas velocity at cyclone inlet: 25m/s, solids load: 0.1 kg/kg

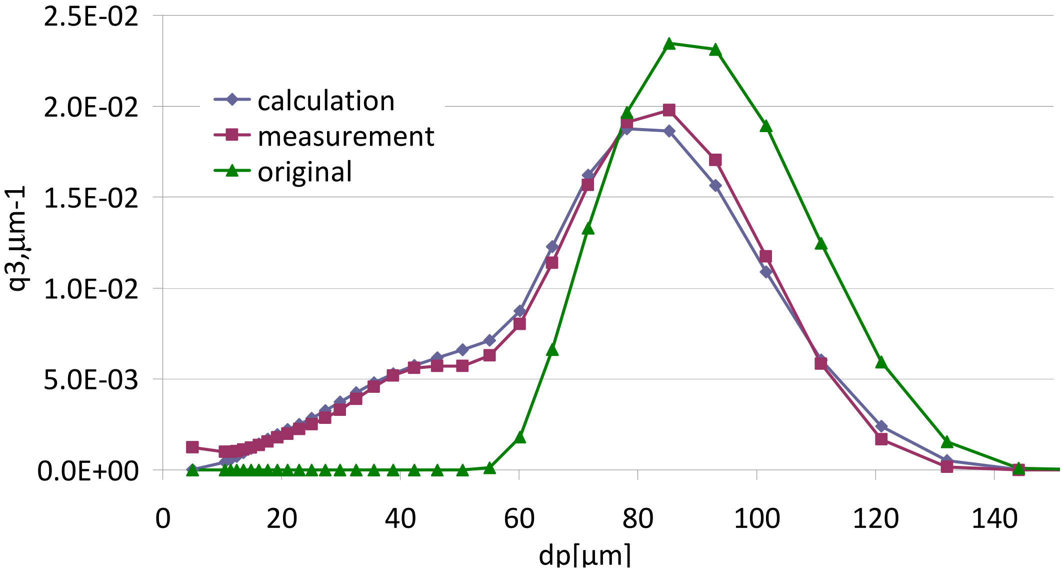

Figure 3: Measured and calculated mass density distribution after 15 passes through the gas cyclone for aluminum oxide. Original size fraction 80-112µm. Operating conditions: gas velocity at cyclone inlet: 25m/s, solids load: 0.5 kg/kg

Simulation of the PSD of a CLC process

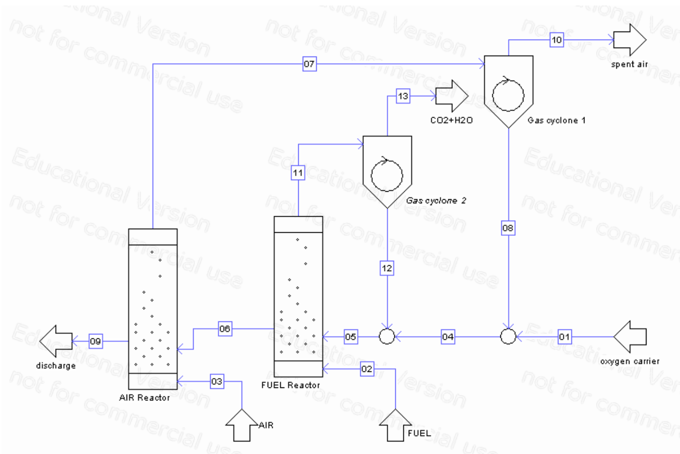

Figure 4: Flowsheet of CLC process in SolidSim

After validation the breakage model was integrated into the flowsheet simulation software for solids processes SolidSim [10]. This tool was used to calculate the PSD of the CLC process with hematite as an oxygen carrier taking breakage due to mechanical stress in the gas cyclones as well as attrition due to bubble-induced material movement in the fluidized beds into account. Figure 4 shows the SolidSim flowsheet of the CLC system. It is composed by two reactors, an air and a fuel reactor. An oxygen carrier circulated between the reactors, transferring oxygen necessary for the fuel combustion from the air to the fuel. The system is designed as a circulating fluidized bed with a bubbling fluidized bed on the return side. The air reactor is operated as the riser. The fuel combustion takes place in the bubbling fluidized bed. The main design and operating parameters are given in table 2. So far CLC processes have been only operated on the laboratory scale. That is why we assumed the respective dimensions.
For the description of the fluid mechanics of the bubbling fluidized bed the model by Werther and Wein (15) is applied, which provides means for the calculation of the height-dependent values of bubble size, bubble rise velocity, visible bubble flow and bubble volume fraction. The elutriation correlation suggested by Tasirin and Geldart (16) is used. The riser is divided into a dense bottom zone and an upper dilute zone. The dense bottom zone is modelled like a bubbling fluidized bed. In the upper dilute zone the solids volume concentration decays exponentially with height (17). Here, an elutriation correlation suggested by Colakyan and Levenspiel (17) was applied. The resulting solids recirculation flux was 40kg/(m²s). The separation effects of the gas cyclones are modeled by the Trefz and Muschelknautz model (18).

	diameter, mm	gas velocity, m/s	pressure drop, Pa	solids load,kg/kg
air reactor	100	7	10	
fuel reactor	300	0.3	2140	
cyclone 1	240	30	323	0.96
cyclone 2	225	10	200	0.05

Table 2: Design and operating parameters

In the fluidized bed bubble-induced attrition was taken into account. For this the model of Reppenhagen and Werther (14) was applied. The respective attrition constant was determined experimentally according to procedure described by Reppenhagen and Werther (14).

Table 3 shows the calculated solid in and out mass flows. The oxygen carrier flow to the system is adjusted so that the solid mass losses in spent air and flue gas are compensated and the discharge from the air reactor is zero. The catalyst loss from the

solids mass flow, g/h		
feed	spent air	flue gas
12.4	8.3	4.1

Table 3: Calcluated solids mass flows

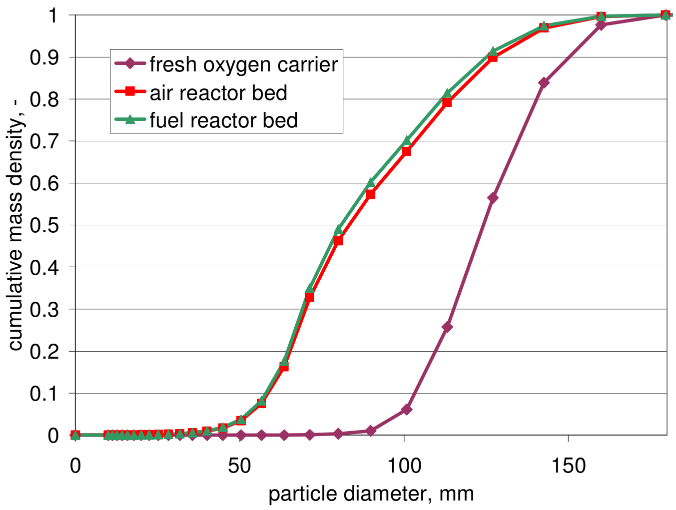

Figure 5: Calculated cumulative mass density distribution in CLC system

system adds up to 300 g/d which is equivalent to 2% of the total inventory of the system. The resulting costs for fresh oxygen carrier have to be considered in the design of a technical plant. The simulation shows in which part of the process an improvement of the solids recovery system would be most effective.

From catalytic processes it is known that the PSD that stabilizes in the reactor under steady-state conditions may differ significantly from the PSD of the fresh catalyst that is fed to the system. Figure 5 shows that this is the case for the simulated CLC process as well. The bed PSDs with a median of 80µm are considerably finer than the PSD of the fresh oxygen carrier with a median of 125µm. Consequently a performance prediction for the process on the basis of the PSD of the fresh catalyst feed is would be misleading.

CONCLUSIONS

A breakage model was developed to predict the change in PSD when solids are mechanically stressed in a gas cyclone. The model predictions are in good agreement with the measurements of particle breakage taken from a laboratory sized gas cyclone for different operating conditions, particle sizes, and materials. The model can be applied to predict the particle size in fluidized bed systems that will stabilize in steady-state operation as a result of attrition and breakage on the one hand and classification in the various cyclones on the other hand.

NOTATION

b_{inlet}	cyclone inlet width, m
$b(v,w)$	number-based breakage function, -
b_{ij}	number of child particles that break from particle size interval j into i, -
c	correction factor in eq. 5
d_p	particle diameter, m
k	parameter of distribution of particle surface defects, -
S_c	material constant, -
S_i	breakage rate for particles in size interval i, -
$S(v)$	number-based specific rate of breakage for particles of volume v, -
t	time, s
u	gas velocity, m/s
v, w	particle volume, m^3

n	exponent in eq. 5	w_p	weighing factor, -
n(v)	number density function for particle volume v, -	W_m	mass-based specific energy, J/kg
p	number of child particles, -	z	parameter in eq. 3 and 4

Greek letters

ε	voidage,-	μ	solids load, kg/kg
φ	occupancy ratio, -	δ	density, kg/m³

REFERENCES

[1] J. Werther, Fluidized bed reactors, in: Ullmann's Encyclopedia of Industrial Chemistry, VCH Publishers, Weinheim, Germany, 1992

[2] A. Lyngfelt, B. Leckner, T. Mattisson, A fluidized-bed combustion process with inherent CO2 separation; application of chemical-looping combustion. Chem. Eng. Sc. 56 (2001) 3101-3113

[3] J. Ströhle, A. Galloy, B. Epple, Feasibility study on the carbonate looping process for post-combustion CO2 capture from coal-fired power plants, Energy Procedia 1 (2009), 1313-1320

[4] M. Pell, Gas fluidization, In: Handbook of Powder Technology, Elsevier, Amsterdam, 1990

[5] J. Werther, E.-U. Hartge, Elutriation and entrainment, in: W.C. Yang (Ed.), Handbook of fluidization and fluid-particle systems, Marcel Dekker Inc., New York, US, 2003

[6] J. Werther, Scale-up modeling for fluidized bed reactors, Chem Eng Sci (47) 1992, 2457-2462

[7] O. Molerus, Heat transfer in gas fluidized beds, Powder Technol (70) 1992, 1-20

[8] P. Hill, K. M. Ng, New Discretization procedure for the breakage equation, AIChE Journal (41) 1995, 1204-1216

[9] P. Hill, K.M. Ng, Statistics of multiple particle breakage, AIChE Journal (42) 1996, 1600-1611

[10] E.U. Hartge, M. Pogodda, C. Reimers, D. Schwier, G. Gruhn, J. Werther, Flowsheet simulation of solids processes, Kona 24 (2006) 146-158

[11] R. Weichert, Anwendung von Fehlstellenstatistik und Bruchmechanik zur Beschreibung von Zerkleinerungsvorgängen, Zement-Kalk-Gips (45) 1992, 1-8

[12] A. Püttmann, E.U. Hartge, J. Werther, to be published

[13] E. Muschelknautz, K. Brunner, Untersuchungen an Zyklonen, Chemie Ing. Technik (39) 1967, 531-538

[14] J. Reppenhagen, J. Werther, Catalyst attrition in fluidized-bed systems, AIChE Journal (45) 1999, 2001-2010

[15] J. Werther, J. Wein, Expansion behavior of gas fluidized beds in the turbulent regime, AIChE Symp. Ser. 90 (1994) 31-44

[16] S.M. Tasirin, D. Geldart, Entrainment of FCC from Fluidized Beds – A new correlation for the entrainment rate constants, Powder Technol. 95 (1998) 240-247

[17] D. Kunii, O. Levenspiel, Fluidization Engineering, Butterworth-Heinemann, Stoneham, MA, 1991

[18] VDI-Heat Atlas, VDI Verlag, Duesseldorf 2006

KEY WORDS

Gas cyclone, breakage, fluidized bed system, population balance, Chemical looping combustion process