Residence time distribution of carbon black in granulation drums

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1. Introduction

In compliance with the protection of environment, health and work and in order to improve transportation and handling properties, a major part of industrially produced carbon black is granulated to form beads. One of the processes used is dry granulation, which particularly meets the demand for easily dispersible products for certain applications.

Dry granulation is carried out on a large scale in rotary drums; the transportation, handling and dispersing properties of the product depend on the properties of the material fed to the drum, e.g. primary particle and aggregate size and, mainly, on the process conditions of granulation. Beside fixed parameters (drum geometry), these conditions include process parameters such as throughput, drum inclination, filling ratio and the number of drum revolutions. These process parameters determine the residence time (distribution) of the carbon black in the drum, which is assumed to be the decisive parameter [1, 2]. If the residence time is too short, the result are very soft granules whereas a too long residence time may yield very hard beads with an increased percentage of broken beads.

The present work describes a systematic investigation of dry granulation of carbon black with special regard to the residence time characteristics. A novel experimental method and a model for the determination of residence time distribution of carbon black in the bead-forming drum are presented, in order to assess the potential for improvement of product quality in operating installations.

2. Experimental

The granulation tests were carried out in a continuously operated semi-technical drum (Fig. 1). The internal diameter of the beadforming drum was D = 0.52 m and the length L = 4 m. A rubber shroud inside the drum was to improve entrainment of the carbon black and to prevent caking. The weir height in the drum could be freely adjusted by two opposing slide gates. For the tests a constant weir height of 100 mm was selected. The number of revolutions of the drum could be continuously adjusted between 0 and 41 rpm. The inclination of the drum was adjusted by handwheel.

The carbon black powder, precompacted to a density of 150 ± 20 g/l was fed to the drum by a twin-screw feeder, using a gravimetric metering balance which was operated to maintain an approximately constant carbon black level (appr. 50 %); this means that it was refilled at short intervals in order to avoid the continuous increase of the apparent density of the powder. Product recycling for stabilising the bead-forming process was abandoned. Only for initiating bead formation the drum was filled with bead-shaped carbon black at the start of the test. In order to counteract fine dust emissions a suction device was installed at the drum exit in the vicinity of the bagging station. The quantity of solids removed by suction amounted up to 6 % of the throughput.

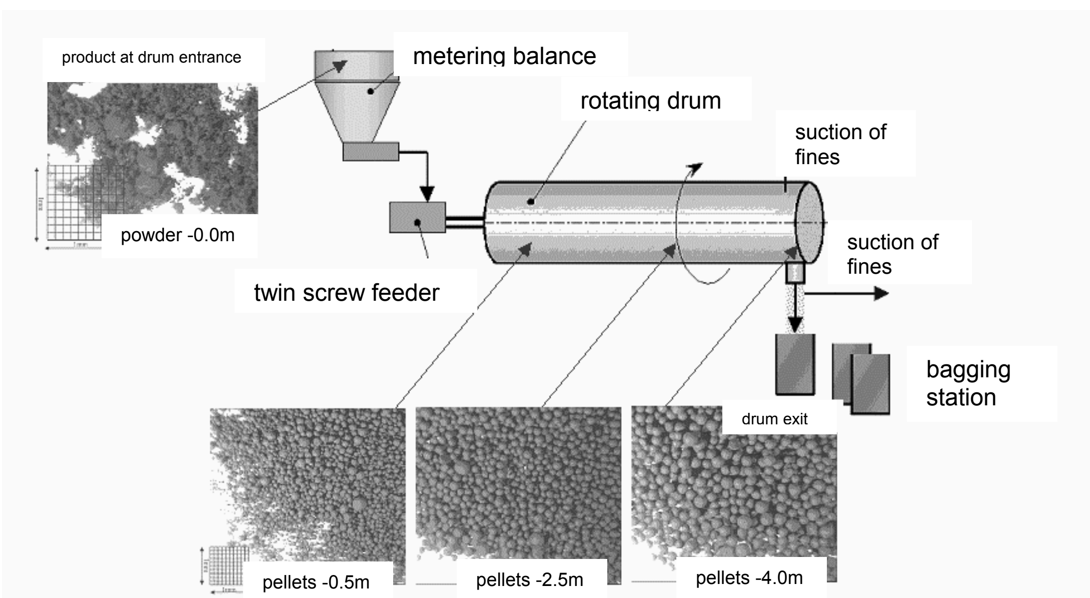

Figure 1. Schematic view of the experimental setup – bead-forming drum with metering balance, twin screw conveyor and various granulation stages

The number of revolutions was varied between 23 and 28 rpm. Within these limits a well flowing carbon black bed was observed. The drum inclination (ascent to drum exit) was made 0° and 1°. The throughput was varied between 5 and 15 kg/h. Table 1 shows the test plan.

Table 1. Plan of experiments

test No	drum throughput M [kg/h]	numbers of drum rotation n [UpM]	drum inclination α [º]
1	15	28	0
2	15	23	0
3	10	23	0
4	5	23	0
5	10	23	1

A test was carried out as follows. First, the empty drum was filled with seed material until - controlled by the inclination of the drum and the weir height - a uniform carbon black bed had formed. Subsequently the test parameters (number of revolutions and throughput) were set. The test series was started with the onset of metering. Every 20 min samples were withdrawn at the drum exit and their apparent density was immediately determined. After stationary conditions - characterised by constant apparent density - had been reached, the residence time measurement was started.

For this purpose 150 to 300 g of a labeling substance were fed to the drum entrance, whereupon samples were withdrawn at the drum exit every 3 min. The labeling substance was a carbon black powder containing appr. 4 % by weight SiO_2, the particle and powder properties of which were identical with those of the product itself. After the residence time had been measured the drum was stopped and samples were taken every 0.5 m along the drum length. Following, the drum was rotated until it was empty; in order to achieve this, the weir slides were removed and the drum exit was lowered. The carbon black quantity thus recovered was weighed.

After having completed the test, the SiO_2 content in the samples taken after the residence time measurement was determined. For this purpose, 2.5 g of the carbon black sample were mixed with 7.5 g stearic acid and compacted to form tablets which were dried at 50 °C over night; the SiO_2 content was determined by X-ray fluorescence analysis (RFA).

3. Results and discussion

3.1 Residence time of carbon black in the beadforming drum - measurement of residence time distribution

An example for the residence time distribution is shown in Fig. 2. It shows the time dependence of the SiO_2 concentration in the main and fine fractions withdrawn from the drum exit.

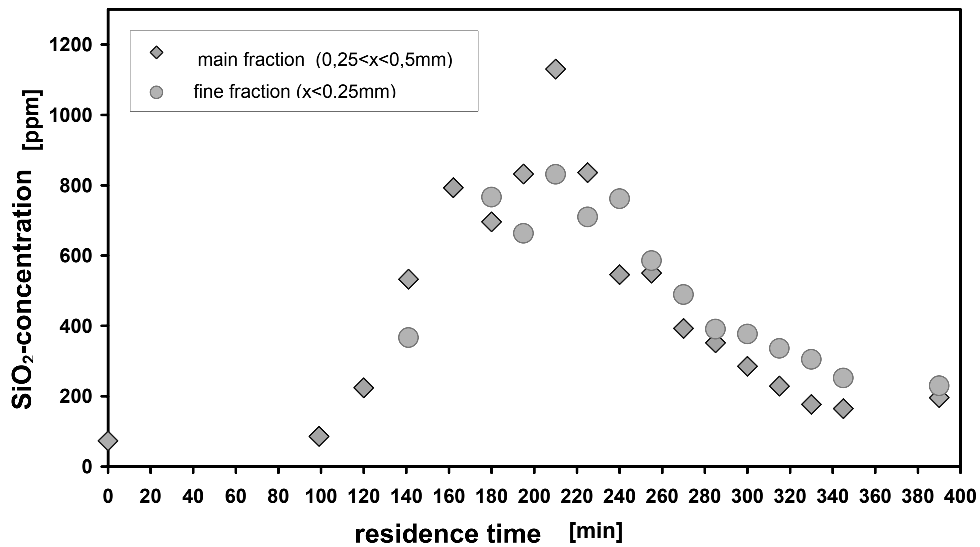

Figure 2. Measured SiO_2 concentration at the drum exit, as a function of test duration

Within the limits of measurement accuracy the shape of the curves does not exhibit any difference between main and fine fraction; the occurrence of bead size screening along the drum axis can therefore be excluded, at least in the horizontal drum.

It can further be recognised that the residence time distributions deviate from a Gaussian distribution. The SiO_2 concentration exhibits a comparatively fast rise to a pronounced maximum and a comparatively slow decay, resembling the shape of a logarithmic normal distribution. The reason for this phenomenon is axial dispersion due to drum rotation, which will be explained in more detail below.

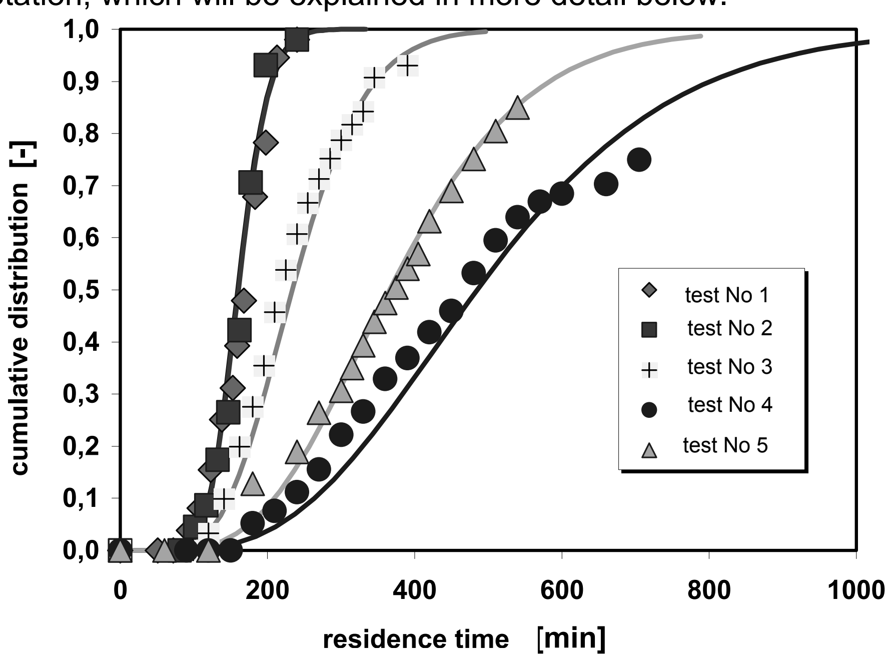

Figure 3. Measured and calculated residence time distribution sums
(cf. Sections 3 and 4)

Fig. 3 shows all the measured residence time curves as distribution sums. It was taken into account that the introduced tracer has not yet left the bead drum completely at the end of the test; this is revealed by the fact that the SiO_2 concentrations are different from zero at the end of the test, as shown in Fig. 2.

A comparison between the different measurement series shows that the mean residence time decreases, as expected, when the throughput increases (compare runs 2 - 4) or the inclination of the drum decreases (runs 3 and 5), while the number of revolutions has no influence on the mean residence time (runs 1 and 2).

It can further be seen that the width of residence time distribution increases as residence time increases. This effect is due to axial remixing and dispersion, respectively, superimposed onto the flow through the drum, which depends on the throughput. Due to rotation an axial transport takes place in the drum, whereby - in analogy to diffusion in gases - dispersion in the flow direction and counterwise are equally probable. Moreover, the dispersion rate is independent of orientation and only depends on the number of revolutions. The higher the latter, the higher the dispersion rate, and vice versa. As residence time increases axial mixing is also enhanced, and this effect is reflected by a broader distribution, which increasingly deviates from the Gaussian distribution.

Finally it appears worth mentioning that the drum with an ascent towards the exit does not produce a broader distribution than the horizontal drum at comparable mean residence time.

3.2 Modeling residence time distribution

3.2.1 Simple geometric model

A simple residence time distribution model is based on simple geometric assumptions, determining the mean residence time τ by equation (1)

$$\tau = \frac{\text{(volume of carbon black bed x apparent density)}}{\text{mass throughput of carbon black}} \qquad (1)$$

where the volume of the carbon black bed is determined by the drum geometry and the assumption of a static bed.

This model obviously has the following shortcomings:

1)
In reality, drum rotation at constant weir height results in a clear increase of carbon black bed mass, as has been shown experimentally with a beadforming disk. Figure 4 shows the results of these investigations, representing the increase of carbon black mass due

to rotation in the beadforming disk for different weir heights and bed levels, respectively. It has e.g. been found that a weir height of 100 mm and a number of revolutions of 28 rpm, as in run 1, give rise to a 40 % increase of carbon black bed mass. When this increase of carbon black mass is neglected a serious error results in the mean residence time (cf. Equ. 1).

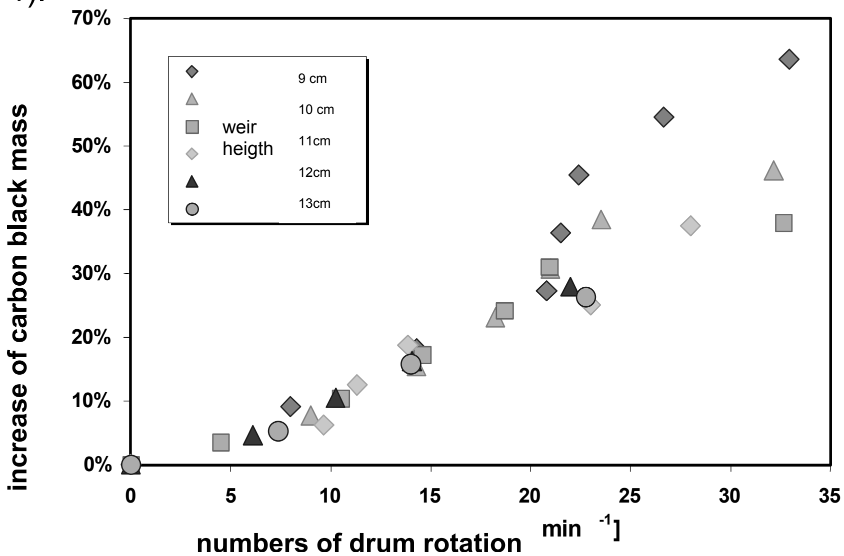

Figure 4. Increase of carbon black bed mass in rotating drum (D = 0.52 m)

2)
The model only yields a mean residence time since plug flow in the bead drum is assumed. Actually, however, axial remixing (see 3.1) gives rise to a residence time distribution in the drum.

3.2.2 Cascade model

Non-ideal flow reactors can be described mathematically using the so-called cascade model (tanks-in-series [3], see equations 2 and 3). This model introduces the serial arrangement of N ideal mixing vessels, the output function of a mixing vessel being simultaneously the input function of the following mixing vessel. The resulting residence time curve for the entire system thus depends only on the model parameter N (number of mixing vessels). N = 1 corresponds to the case of the ideal mixing vessel (complete mixing), while N → ∞ describes the ideal flow tube (plug flow, no mixing) (Fig. 5).

$$E(\theta) = N^N.\theta^{N-1}.\exp[-N.\theta] \frac{1}{(N - 1)} \quad (2)$$

$$F(\theta) = \int_{t=0}^{t=\infty} E(\Theta)d(\Theta) \quad (3)$$

In this context, Θ is the normalised residence time ($\Theta = t/\tau$), $E(\Theta)$ is the residence time distribution density and $F(\Theta)$ represents the residence time distribution sum.

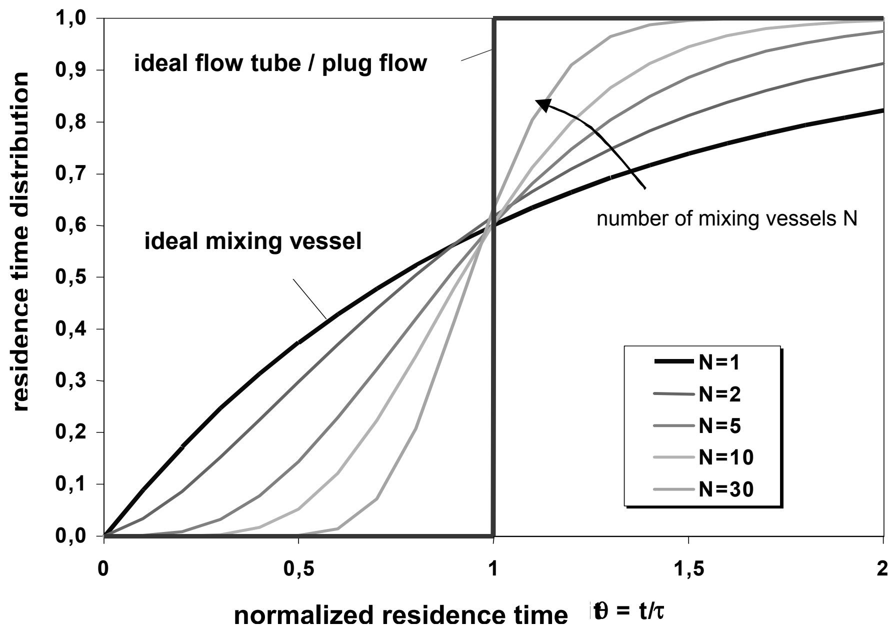

Figure 5. Residence time distribution sum according to the cascade model

Thus, the experimental data (Fig. 3) were re-evaluated using the following procedure:
I)
Calculation of the mean residence time according to Equ. 1, introducing the real change of apparent density along the drum;
II)
Correcting the mean value by introduction of the rpm-dependent carbon black bed mass;
III)
Generation of a residence time distribution around the corrected mean value using the cascade model.

It turned out that appropriate adaptation of the only model parameter "Number of mixing vessels N" brings model and experiment into very good agreement (see Fig. 3).

The number of mixing vessels N, necessary to describe the residence time distribution depends on the mean residence time (Fig. 6). At comparatively short mean residence times in the drum (τ < 180 min) where no noticeable axial remixing occurs, N reaches large values. The system then approaches the case of a plug-flow. At long residence times, however, where remixing dominates, N becomes very small and the system approaches the case of the ideal mixing vessel. This good match of model and experiment allows the conclusion that the model describes essential physical effects.

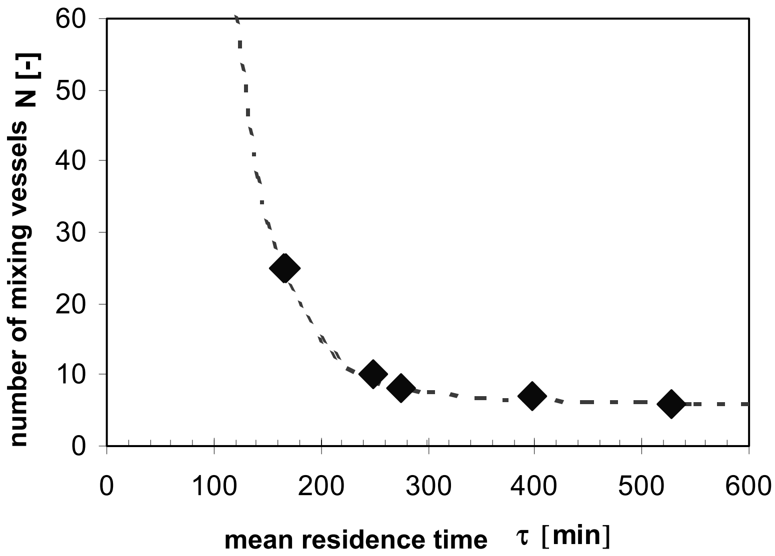

Figure 6. Model parameter "Number N of mixing vessels" depending on the mean residence time τ of carbon black in the bead drum

4. Simulation of the influence of product recycling on the residence time distribution

Finally, the residence time distribution model was used to simulate the influence of continuous product recycling, a measure frequently used to stabilise the beading process.

Fig. 7 shows the result of such a simulation in case that the quantity of recycled product amounts to 20 % and 50 % of the throughput, respectively. The starting level used was a mean residence time of 240 min without product recycling (Figure 7, solid curve). The distribution widths were determined using Fig. 6.

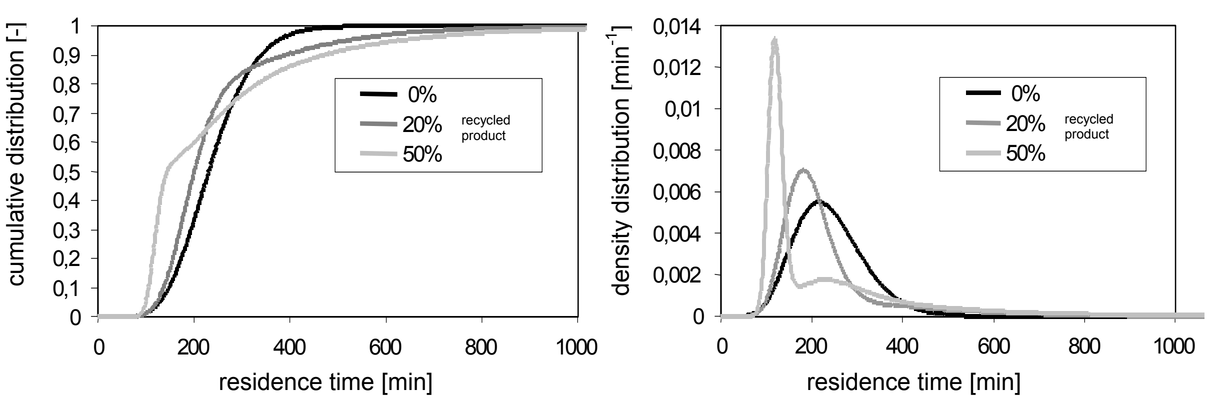

Figure 7. Change of residence time distribution depending on the quantity of recycled product

When the bead-forming drum is operated with product recycling the throughput increases and the residence time distribution correspondingly shifts to shorter times. At the same time, however, an increase of distribution width can be observed. The monomodal residence time distribution without product recycling (0 %) is then converted into a bimodal distribution and the distribution sum arrives at a value of 1 not until long time. At 50 % product recycling e.g. 50 % of the carbon black have left the drum after less than 145 min, while appr. 10 % of the carbon black stay in the bead-forming drum for more than 500 min.

5. Summary

Systematic investigations into dry granulation of industrial carbon black have been carried out, with special regard to the residence time characteristics, using a novel experimental method.

A model for the determination of residence time distribution of carbon black in the bead-forming drum has been developed and very good agreement of model and experiment has been demonstrated.

Simulations based on the model demonstrate that product recycling during bead formation results in a considerably broader residence time distribution.

Literature

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