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In-line particle sizing for real-time process control by fibre-optical spatial filtering technique (SFT)

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Abstract

Sizing of particles in industrial processes is of great technical interest and therefore different physical-based techniques have been developed. The objective of this study was to review the characteristics of modern sizing instruments based on a modified fibre-optical spatial filtering technique (SFT). Fibre-optical spatial filtering velocimetry was modified by fibre-optical spot scanning in order to determine simultaneously the size and the velocity of particles. Sizing in-line instruments of Parsum GmbH use these measuring principles and may be adapted to different process conditions. Particles with sizes of 50 µm to 6,000 µm and velocities up to 50 m/s may be measured by the probe system IPP 70. An overview is given to real-time sizing of particles in different technical applications: fluid-bed granulation, high shear wet granulation, Wurster coating, mixing, spray drying, crystallization and milling.

Keywords: spatial filtering technique, particle size, in-line measurement, real-time process control, process analytical technology

1. INTRODUCTION

Sizing of particles in industrial processes is of great technical interest and therefore different physical-based techniques have been developed [1]. Especially in-line measurement techniques may provide a better understanding of the phenomena in particle systems and also improve the control of processes. Advantages are the improvement of the process stability and of the product quality and otherwise the reduction of waste, store and costs. Another interesting parameter is the particle velocity in particle laden flows or in particulate materials. The simultaneous measurement of size and velocity of each particle allows the determination of the local particle mass flow rate at the measuring position.

Particle size measurement techniques may be divided into two categories, stream scanning and field scanning. Stream scanning means the examination of one particle at a time and field scanning the simultaneous examination of an assembly of particles. Both principles may be applied on particle sizing of industrial processes. Inserting a probe directly into a process line is called in-line particle sizing. Ideally, the whole process flow would be examined, but process streams often operate at high particle flow rates. It is therefore a common method to measure in a side-stream which can be isolated from the main process flow. Another option is the application of a dilution step between the particle stream and the measuring device.

Known in-line measuring devices based on field scanning use the laser diffraction, incoherent space frequency analysis and ultrasonic or light attenuation. Examples of in-line sizing devices based on stream scanning are the shadow Doppler velocimetry (SDV), the focused beam reflectance measurement (FBRM), light scattering, imaging technique using CCD cameras, the phase Doppler method (PDA) and the spatial filtering technique (SFT). Based on the spatial filtering technique, Petrak [2] has described a new optical probe for the simultaneous measurement of size and velocity at one particle. Fibre-optical spatial filtering velocimetry (SFV) was modified by fibre-optical spot scanning (FSS) in order to determine the particle size. Meanwhile better in-line measuring devices with different probes based on modified spatial filtering technique are available by PARSUM GmbH (Chemnitz, Germany) with the particle size range of 50 µm – 6,000 µm and the particle velocity range of 0.01 m/s – 50 m/s with a data rate up to 20,000 s^{-1} [3], [4].

The present work gives an overview of the new SFT-sizing in-line instruments and of real-time sizing of particles in different technical processes: fluid-bed granulation, high shear wet granulation, Wurster coating, mixing, spray drying, crystallization and milling.

2. MEASURING PRINCIPLE

The spatial filtering velocimetry (SFV) is a method of the velocity determination of an object by observing the object through a spatial filter in front of a receiver. An overview about fundamentals and applications of the SFV method was given by Aizu et al. [5]. The modified spatial filtering technique for the simultaneous measurement of particle size and velocity can be described as follows.

The fibre-optical spot scanning (FSS) is an addition to the SFV. The basic operation of the FSS is to observe the shadow image of a moving particle through a single optical fibre with a small diameter *b*. When the shadow image passes through the single optical fibre, an impulse is generated and the width of which depends on the particle size *x*, the particle velocity *v*, and the random spatial location of particle and fibre. The shadow image of the particle has the same size as the particle, if the light diffraction by the particle and the influence of a divergent angle of the illumination beam can be neglected. The composition of the total impulse width is shown in Figure 1. The total time Δt of the impulse is then:

$$\Delta t = (t_2-t_1) + (t_3-t_2) + (t_4-t_3) = (b/v) + (x-b)/v + (b/v) \qquad (1)$$

and the particle size *x* is equal

$$x = v \cdot \Delta t - b \, . \qquad (2)$$

The particle trajectory has a random position in relation to the spatial fixed single fibre. If the particle is a sphere with the radius *r*, the chord length *a(y)* of the shadow image of the particle is then

$$a(y) = 2\sqrt{r^2 - y^2} \qquad (3)$$

with *y* as the normal distance of the chord from a parallel axis through the centre.

The cord size distribution for a random cut across the particle shadow is given by the following equation

$$P(a_1, a_2) = \frac{\sqrt{4r^2 - a_1^2} - \sqrt{4r^2 - a_2^2}}{2r} \qquad (4)$$

where $P(a_1,a_2)$ is the probability of obtaining a chord size between a_1 and a_2. Langston et al. [6] have shown that particle size analysis from chord measurements can be evaluated using Bayes' theorem.

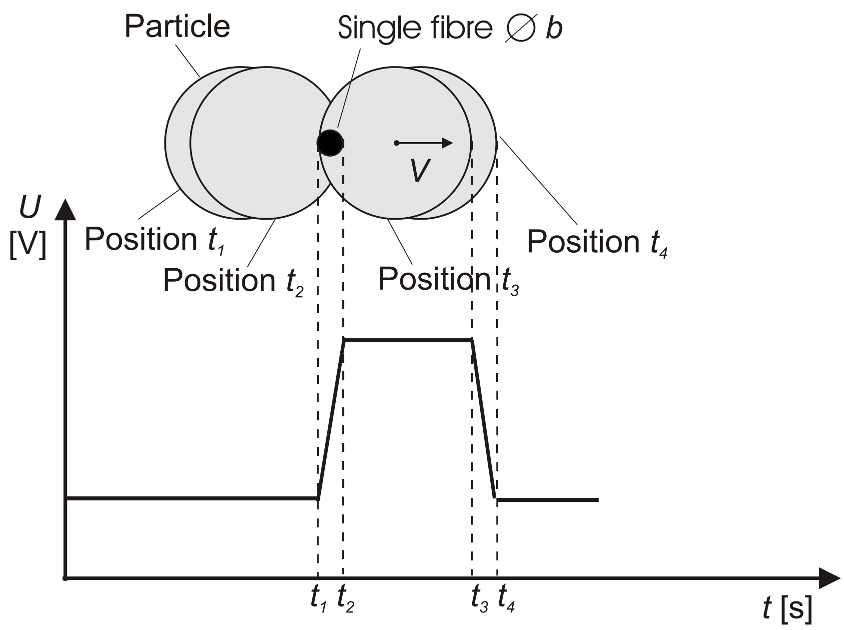

Fig. 1: Impulse generation by fibre-optical spot scanning

The unknown particle velocity v can be determined by the SFV method. As mentioned above, the basic operation of SFV is to observe the image of a moving object through a spatial filter placed in front of a photo-detector. The output signal of the photo-detector contains a frequency f_o related to the object velocity v:

$$v = f_o \cdot s/M \,, \tag{5}$$

where s is the interval of the spatial filter and M is the magnification of the imaging system. Various types of spatial filtering velocity-meters are mainly characterised by the physical type of the spatial filter: transmission grating type, detector type, optical fibre type, and special grating type. The optical fibre type was investigated by Hayashi et al. [7].

The probe line IPP 70 from Parsum with the modified spatial filtering technique uses a fibre-optical configuration as shown in Figure 2. The single fibre for spot scanning and a differential-type fibre-optical spatial filter are arrayed together. Optical fibres of diameter b are linearly arrayed at intervals of $s/2$. The shadow of a moving particle is formed at the entrance faces of every optical spatial filter with the interval s. The output light from every filter is collected, and an output signal is obtained by taking the difference between them to eliminate the pedestal frequency around zero spatial frequency.

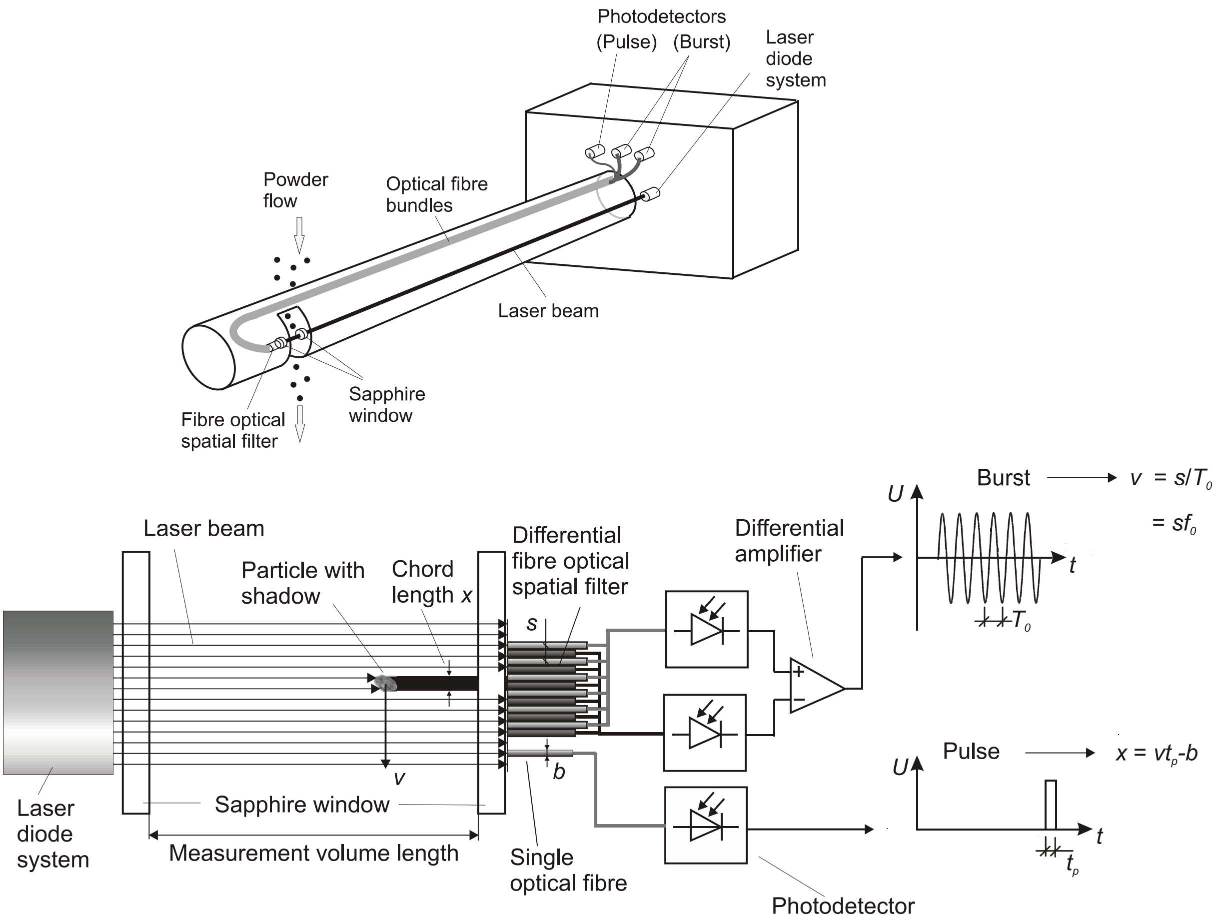

Fig. 2: Schematic probe diagram of the modified spatial filtering technique

The power spectrum $H(\mu,\nu)$ of the spatial filter function represents the selective characteristic in the spatial frequency domain. It can be obtained by Fourier transforming after weighting ±1 at the end faces of uniform optical fibres:

$$\left|H(\mu,\nu)\right|^2=\pi^2 b^4 (\sin\pi s\mu/2)^2\left(\frac{\sin N\pi s\mu}{\sin\pi s\mu}\right)^2\left(\frac{J_1(\pi b\sqrt{\mu^2+\nu^2})}{\pi b\sqrt{\mu^2+\nu^2}}\right)^2, \qquad (6)$$

where μ and ν are the spatial frequencies in the x and y direction, respectively, J_1 is the Bessel function of the first order, and $2N$ is the total number of the fibres with the diameter b. According to Eq. (6), the optical fibre array selects a narrowband spatial frequency to $\mu = 1/s$ and acts as a filtering function on the input function $F_p(\mu,\nu)$ in the space domain. The spatial power spectrum $G_p(\mu,\nu)$ is given by:

$$G_p(\mu,\nu)=F_p(\mu,\nu)\left|H(\mu,\nu)^2\right|, \qquad (7)$$

where $F_p(\mu, \nu)$ stands for the Fourier transform of the image intensity *i (x, y).* The temporal power spectrum $G_p(f)$ is given by integrating Eq. (7) with respect to the spatial frequency ν:

$$G_p(f) = \frac{1}{v} \int_{-\infty}^{\infty} F_p\left(\frac{f}{v}, \nu\right) \left|H\left(\frac{f}{v}, \nu\right)\right|^2 d\nu, \tag{8}$$

where a relation of $\mu = f/v$ has been used. The temporal power spectrum $G_p(f)$ may have a peak at $f = f_o = v/s$. By measuring the frequency f_o, the particle velocity v can be determined from Eq. (5).

The spatial filter should be constructed to have the appropriate filtering characteristic by choosing the following parameters: transmittance, interval of filter elements, number of filter elements. A systematic deviation smaller than 1 % of the central frequency f_o is almost negligible for the number of filter elements $N > 5$. The optical fibres have an insufficiently long aperture size in the direction perpendicular to the spatial filter axis. Therefore, the velocity signal has the best signal-to-noise ratio if the spatial filter axis and the particle trajectory have the same direction. The proposed method is more qualified for an almost parallel particle flow with a main flow direction. Various signal-analysing techniques applied in laser Doppler anemometry can also be used in spatial filtering velocimetry: spectrum analysis, correlation, frequency counting, and frequency tracking.

3. CHARACTERISTICS OF SFT INSTRUMENTS

Sizing instruments of Parsum GmbH are different probes which contain the modified fibre-optical spatial filtering technique. A Parsum Probe IPP 70 is shown in Figure 3. The Probe IPP 70 is constructed from stainless steel and has sapphire windows. This probe system is suitable for size measurements in the range 50 µm – 6,000 µm with an uncertainty of 1 % and for velocity measurements between 0.01 m/s and 50 m/s with an uncertainty of 0.5 %. The probes have a standard tube length of 280 mm and a diameter of 25 mm. The tube length can be extended to 4 m with a diameter of 50 mm.

The temperature at the measuring point reaches from -20°C to 100°C. With relation to the measurement principle, alignment and field calibration are not required. With relation to the process environment, a verification kit allows a control of the whole measurement system.

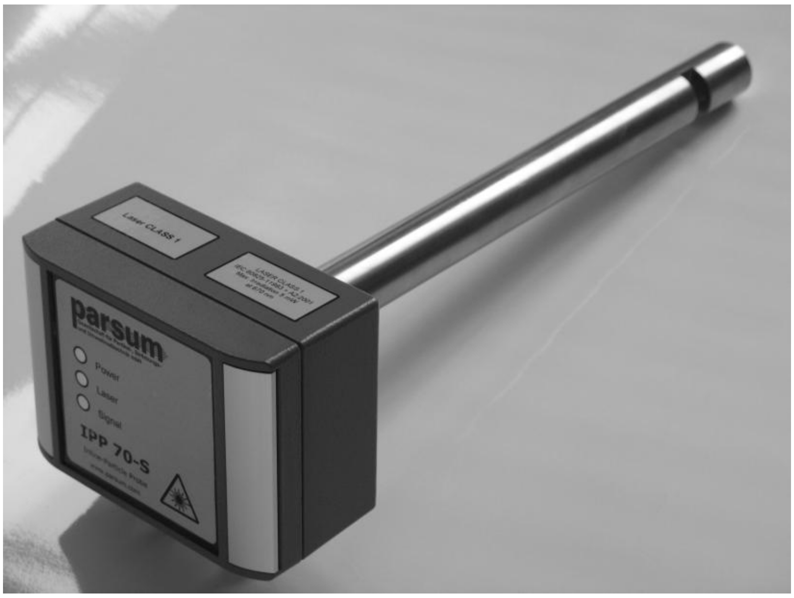

Fig. 3: Fibreoptical probe IPP 70 made by Parsum

The probe can be installed directly in a process line to provide real-time analysis. A range of accessories enables the application to particle systems with high concentration. By feeding compressed air or other gases to the measuring volume, the optical windows are kept clear using different flushing cells. Additionally, an in-line disperser can be used to dilute and disperse the sample flow if particle loading is too high. The IPP 70 with barrier housing and special air valves can be used for EX environments. In the basic configuration the probe is connected to a measurement computer at process level. Up to 4 single probes may be connected to a measurement computer simultaneously. An automatic feedback to a central control system is possible by 4 mA…20 mA Interface and by TCP-Server technology or by OPC-Server.

The data rate is available up to 20,000 particles per second. Parameters derived from the chord length distributions e. g. the size values $x_{10,3}$, $x_{50,3}$ and $x_{90,3}$ can be correlated against other particle size parameters. Additional pharmaceutical options support applications in the pharmaceutical industry.

4. APPLICATIONS

The advantages of the SFT instruments are low hardware requirements, real-time measurement, variable process interface, user friendly handling, long time stability, variable buffer size and robust design at reasonable cost. The SFT instruments may be used for different processes: agglomeration in fluidized bed processes, mixing and coating, high shear wet and dry granulation, spray drying, sieving, grinding and dosing, transportation and filling, spraying. Basic principles of the at-line, on-line and in-line sampling technique with the probe system are illustrated in Figure 4.

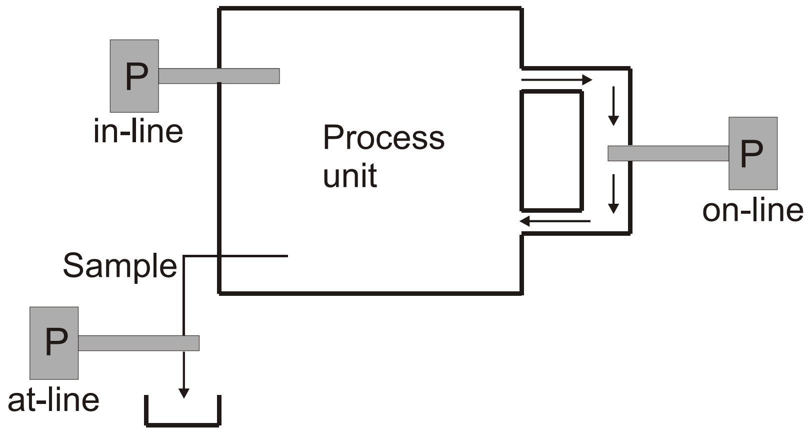

Fig. 4: Illustration of three kinds of measurement with the probe system (P)

In in-line measurement, particle size analysis is carried out during the process without any special sampling procedure. The in-line application of the Parsum Probe IPP 70 in the process unit enables monitoring of the particle size distribution during the process time t_M with the following results: increase of process transparency, short response time in the event of process disturbances, continuous control of the product quality, full feedback control for automated solutions, no need of samples and laboratory analysis.

4.1 GRANULATION
4.1.1 FLUIDIZED BED GRANULATION

The fluidized bed spray granulation plays an important role in many industrial processes [8]. It is used for the production of granular solids with application as particle catalysts or absorbents, waste water treatment granules, salts, pharmaceuticals and so on. The actual particle size distribution of the granules is one of the critical parameters determining the product quality relating to drying behaviour, solubility and agglomerate formation. Figure 5 shows the result of the in-line measurement with a Parsum probe in batch fluidized bed granulation of pharmaceuticals [9]. The granulation of 5 kg lactose with active pharmaceutical ingredients was performed in a laboratory granulator Glatt WSG 5 (Glatt GmbH, Binzen, Germany). The granule build-up can be followed with high time resolution and in real-time. Comparison of the $x_{50,3}$ - values between at-line laser diffraction measurements with a HELOS System (Sympatec GmbH, Clausthal-Zellerfeld, Germany) and the in-line measurements with Parsum probe shows a satisfying correspondence. The particle growth is strongly influenced by the spray rate and inlet air flow.

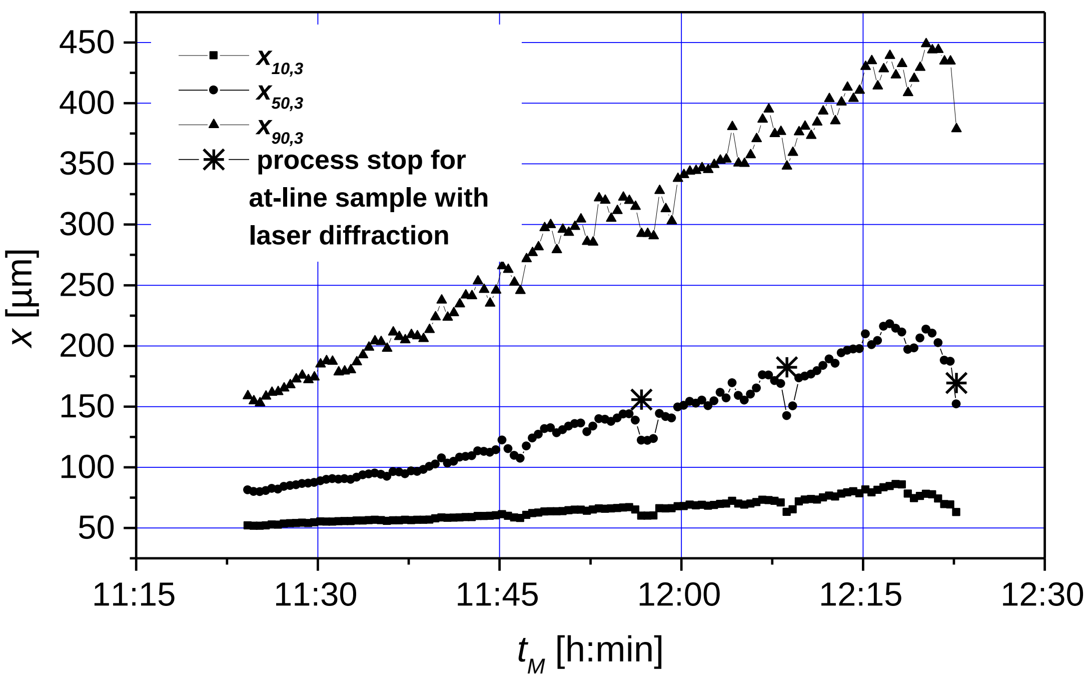

Fig. 5: Particle size values $x_{10,3}$, $x_{50,3}$ and $x_{90,3}$ in dependence on process time t_M in a batch fluidized bed granulation of 5 kg lactose with active pharmaceutical ingredients

Using at-line measurements with Parsum Probe IPP 70 for experimental studies of fluid bed granulation, predictive models for the final particle size distribution have been constructed [10], [11]. The granulations were performed in a granulator Glatt WSG 5 with variation of air humidity, granulation liquid feed rate and granulation liquid feed pulsing. Three different measuring methods were used for the determination of particle size distribution: sieve analysis, laser light diffraction and Parsum Probe IPP 70. The IPP 70 results for the median granule size model gave the best goodness of fit (R^2) and goodness of prediction (Q^2). Only the IPP 70 results gave acceptable R^2 and Q^2 values for the relative size distribution width $(x_{90,3}-x_{10,3})/x_{50,3}$ of the predictive size distribution. The IPP 70 measurements show that liquid feed pulsing is an efficient way to modify the particle size of the final granules.

The feasibility of the Parsum Probe IPP 70 as process analytical technology tool for the in-line monitoring of the particle size distribution during top spray fluidized bed granulation was examined by Burggraeve et al. [12]. The results indicate the ability to predict end granule properties based on in-line SFT data that can be beneficial in both development and routine production. Huang et al. [13] could show that the combined use of process analyzing by Parsum probe and multivariate tools improves the process understanding and the end product quality.

4.1.2 HIGH SHEAR WET GRANULATION

High shear wet granulation is a granulation process using high speed mixing blades. Granules were produced by addition of binder solution to powder with active pharmaceutical ingredient and excipients. Overgranulation may generate low porosity granules and affect the mechanical properties of the tablets. End-point determination based on particle size distribution was found to be the best. Figure 6 demonstrates the development of the granulation process in a Pilot Processor System P/VAC 10-6 (DIOSNA Dierks & Söhne GmbH, Osnabrück, Germany) with 15 kg mixture of lactose and microcrystalline cellulose.

The different process steps dry mixing, spraying and granulation are clearly characterized by the different size values $x_{10,3}$, $x_{50,3}$ and $x_{90,3}$.

4.2 PELLET COATING

Goal of coating of particles is to monitor layer thickness and to detect agglomerates. Two examples show the probe results of Wurster coating [14]. A typical pellet coating process with the development of a 50 µm thick layer is given in Figure 7. The generation of agglomerates is characterized by the development of a bimodal distribution with a second maximum, which is increased with the process time t_M.

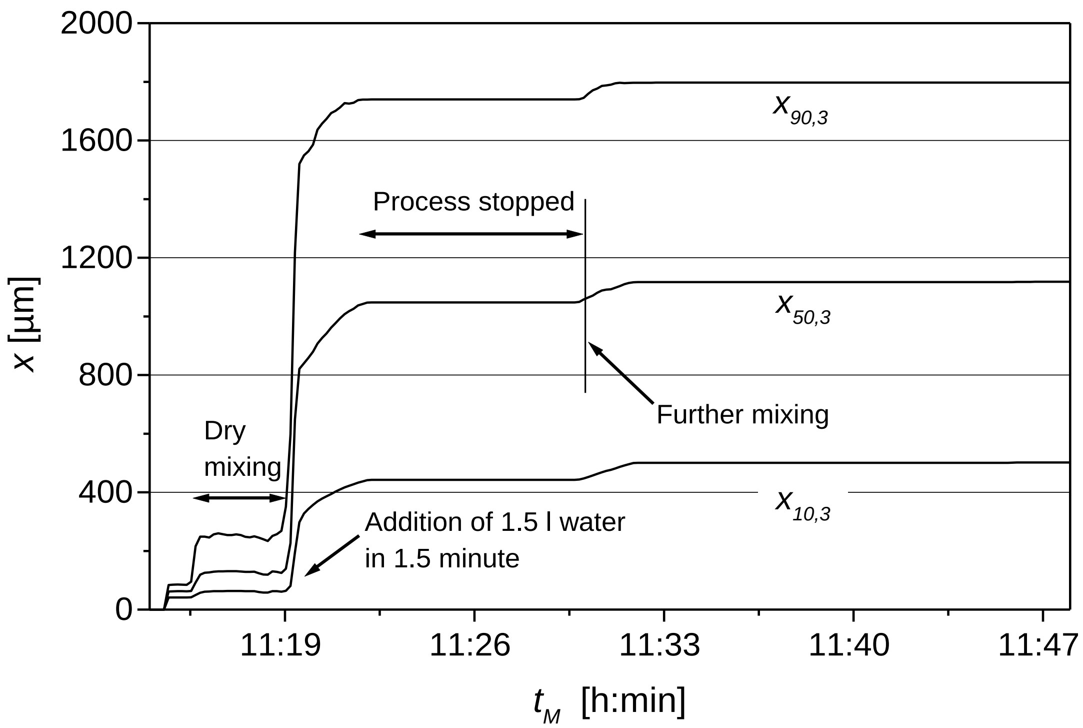

Fig. 6: Particle size values $x_{10,3}$, $x_{50,3}$ and $x_{90,3}$ in dependence on process time t_M in a high shear wet granulation of 15 kg mixture lactose with cellulose

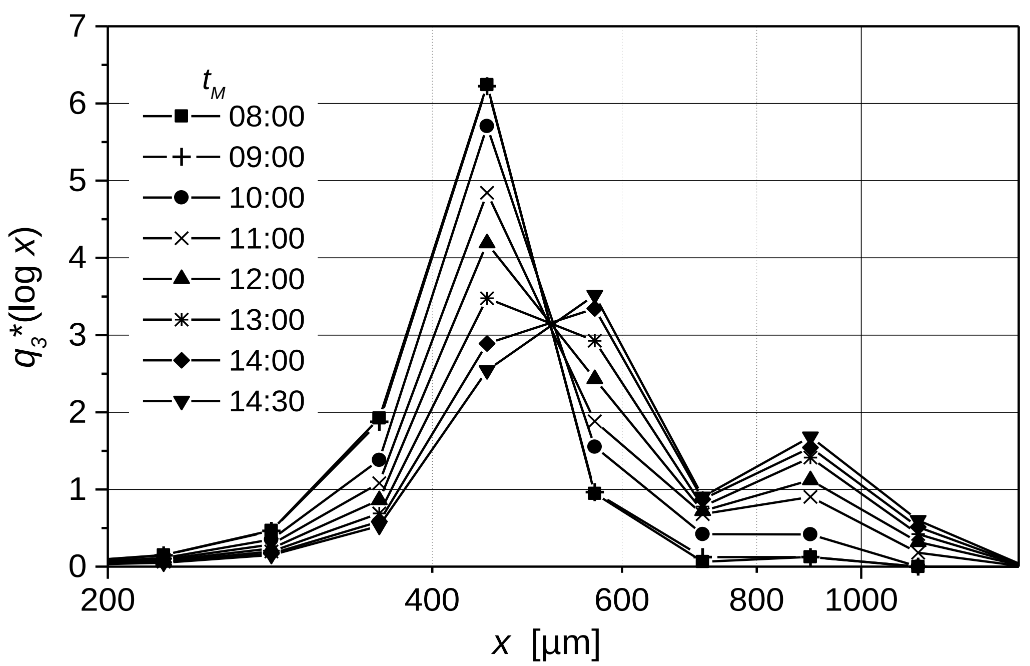

Fig. 7: Development of the density distribution by volume q_3*(log x) of a typical pellet coating process in dependence on process time t_M

A small increase of the agglomeration may be monitored by taking the difference between a reference size distribution at the process start with size distributions of later time. The probe was installed in a Glatt GPCG 2 fluidized bed system with a 6''-Wurster. The batch material was 1.5 kg cellets 200-355 with a 6 % pharmacoat -606-solution. The results for three spray rates were given in Figure 8. An increase of the spray rate is connected with an increase of the agglomeration.

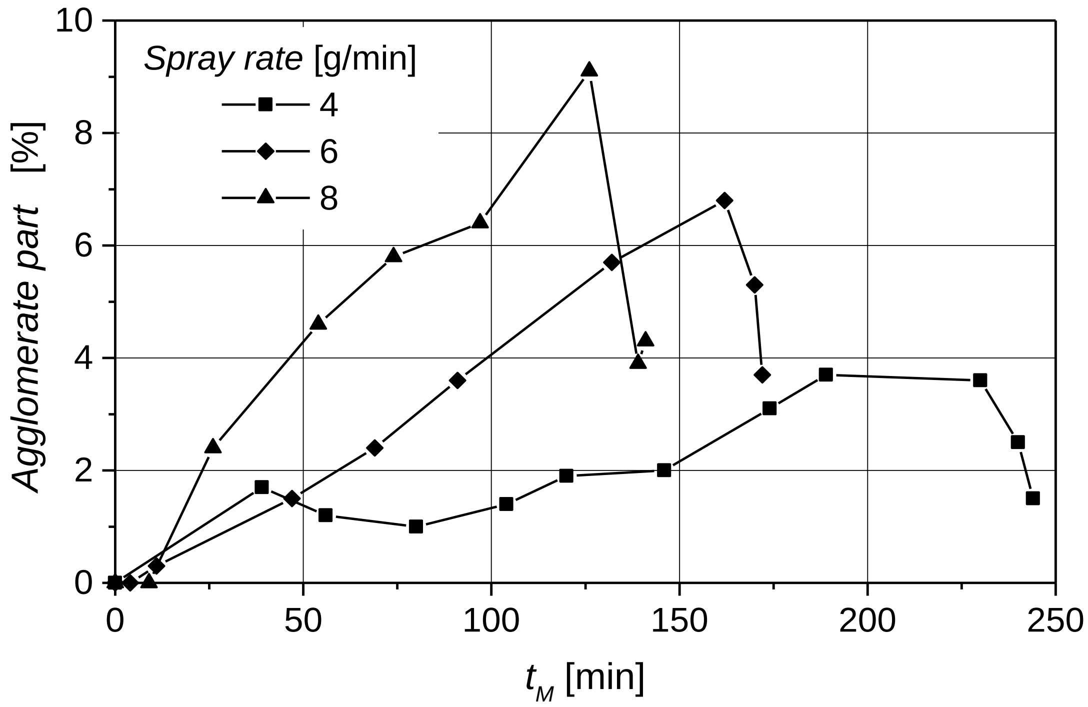

Fig. 8: Agglomeration in dependence on the spray rate for a coating process in a Wurster granulator

New probe results for a coating process were given by Fischer et al. [15]. The Parsum Probe IPP 70 was used to monitor the evaluation of the particle size distribution during fluidized coating experiments. The measured chord length distributions were translated into the corresponding particle size distributions by a geometric transformation approach. Alumina spheres served as nucleus material and were coated with sodium benzoate solution from a top spray nozzle. In comparison in-line data and off-line reference data bear a sufficient agreement for applications in granulation processes.

4.3 FURTHER PROCESSES

Figures 9-11 show further technical applications of the modified spatial filtering technique. The influence of the revolution number of a wheel atomizer in a spray drying process is demonstrated in Figure 10. After drying the droplets by hot gases, the solid particles were measured by the probe at the exit of the spray drying tower.

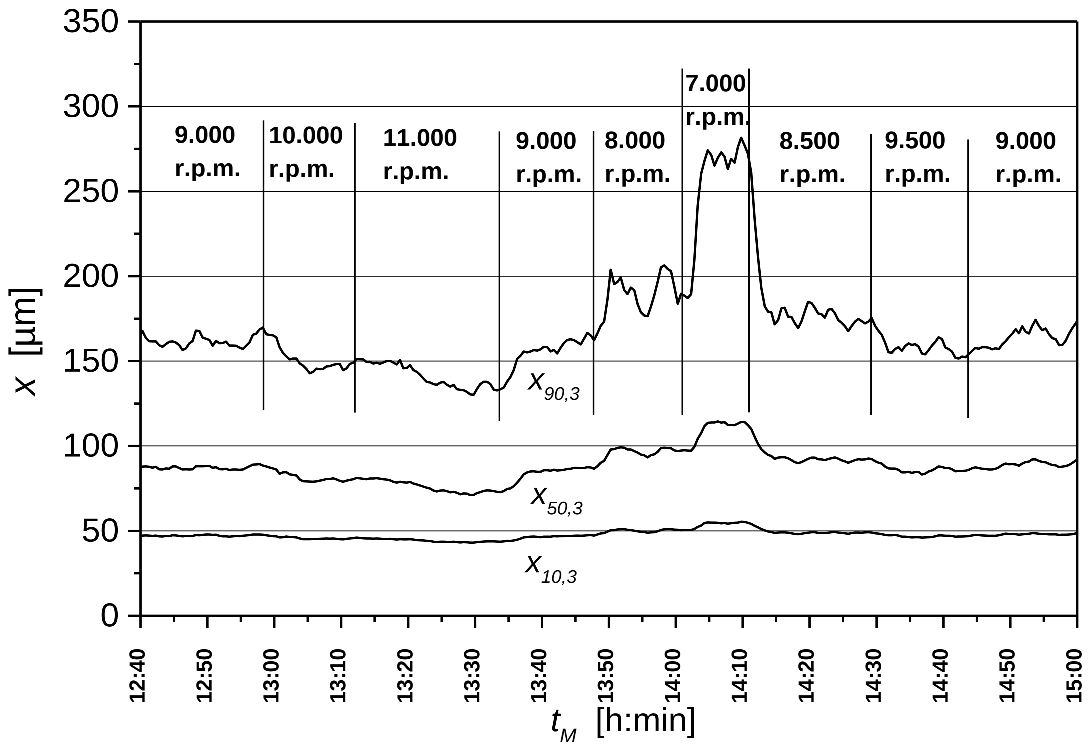

Fig. 9: Particle size values $x_{10,3}$, $x_{50,3}$ *and* $x_{90,3}$ in dependence on process time t_M for spray drying

Another example is the size and velocity measurement of droplets by the modified spatial filtering technique. Water droplets were produced by a spray nozzle of a fire sprinkler system. The measurement position on the axis of the spray cone was in a distance of 650 mm from the nozzle exit. The influence of the water pressure *p* on the median droplet size and median velocity is shown in Figure 10.

An example of a crystallization process is given in Figure 11. The generation of the ammonium sulphate crystals can be monitored clearly by the probe measurements.

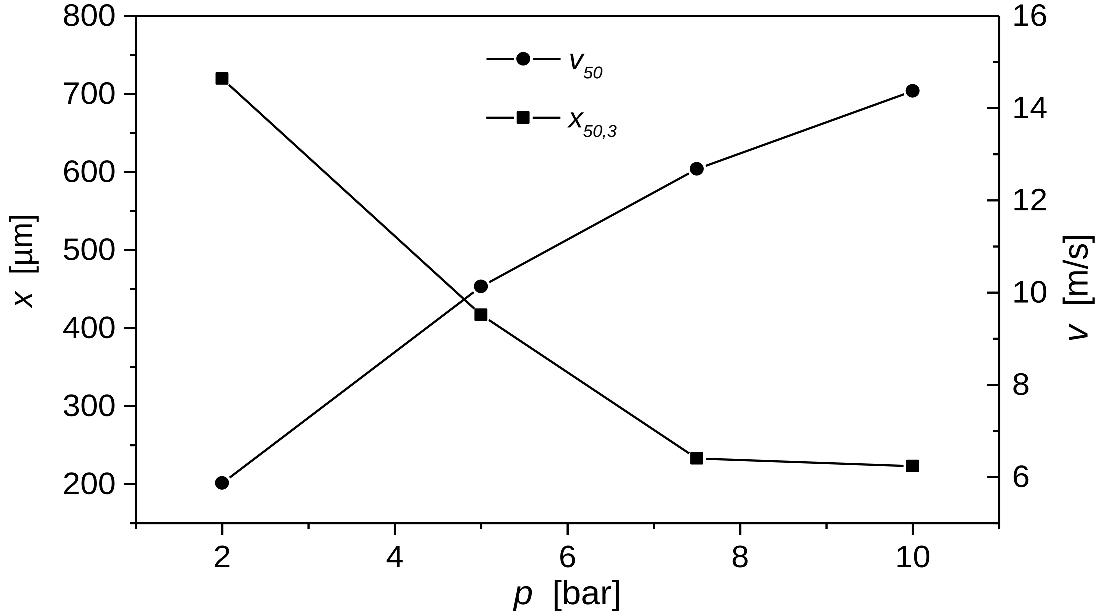

Fig. 10: Dependence of median droplet size $x_{50,3}$ and median droplet velocity v_{50} on the nozzle pressure p of a fire sprinkler system

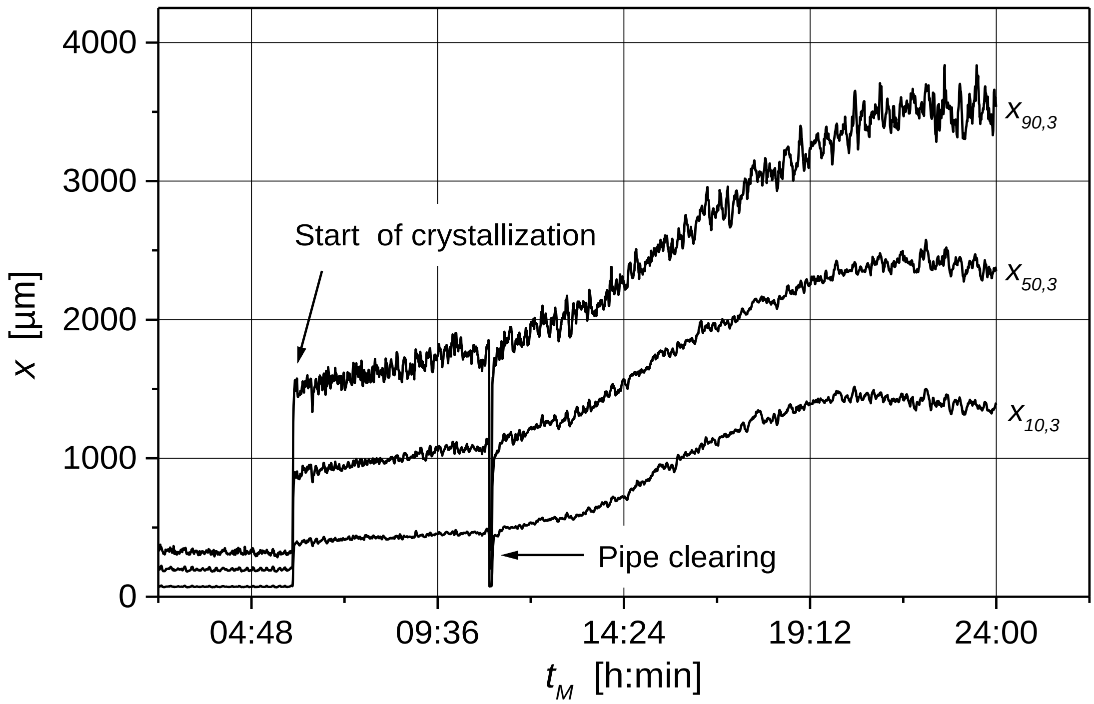

Fig. 11: Monitoring of a crystallization process of ammonium sulphate solution

Grinding of grain (whole meal) by a roller mill could be measured by a Probe IPP 70. Decrease of the mill slit reduces the product size.

5. CONCLUSIONS

The fibre-optical probe system IPP 70 based on a modified fibre-optical spatial filtering technique is suitable for the local measurement of particle size and particle velocity. Owing to the random cut across a particle, the measuring result is a chord size distribution. Particles with sizes of 50 µm to 6,000 µm and velocities up to 50 m/s can be measured by the probe system IPP 70. The sizing in-line instrument can be adapted to different process conditions by using flushing cells and an in-line disperser unit. Experimental results to real-time sizing of particles in different technical applications show the applicability on fluid-bed granulation, high shear wet granulation, Wurster coating, mixing, spray drying, crystallization and milling. The development of particle growth can be monitored and critical control parameter can be defined. The measurements promote the generation of mathematical relationships for the process development.

Nomenclature

a	chord length of a circle area
b	diameter of an optical fibre
f	frequency
f_0	signal frequency
F_p	filter function
$G_p(f)$	temporal power spectrum
$G_p(\mu, v)$	spatial power spectrum
$H(\mu, v)$	power spectrum of the filter function
$i(x, y)$	image intensity
J_1	Bessel function of first order
M	image magnification
N	number of fibres
p	pressure

P	probability
$q_3*(log\ x)$	density distribution by volume in a representation with a logarithmic abscissa
$Q_3(x)$	cumulative distribution by volume
Q^2	goodness of prediction
r	circle radius
R^2	goodness of fit
s	interval of a spatial filter
t	time
t_M	process time
v	particle velocity
x	particle size, chord length, coordinate
$x_{50,3}$	median particle size value with $Q_3(x_{50,3}) = 0.5$
$x_{10,3}$	particle size value with $Q_3(x_{10,3}) = 0.1$
$x_{90,3}$	particle size value with $Q_3(x_{90,3}) = 0.9$
y	normal distance, coordinate
μ	spatial frequency in x direction
v	spatial frequency in y direction

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