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Application of laser diagnostics on particle technology processes

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Abstract
High porous metal nanopowders are useful filler materials in conductive polymer-matrix composites, because they combine electrical conductivity with improved thermomechanical properties of the composite. These ultrapure powders, which are produced in the Inert Gas Condensation (IGC) process, develop from coalescence growth and coagulation processes of metal vapor in a helium gas as a result of succeeding nucleation. The agglomeration of silver nanoparticles in such an aerosol has been monitored on-line as a function of background pressure and particle content in the aerosol. Various laser optical methods have been used to prove quantitative measurement of the particle velocity, as for example: Laser Doppler Anemometry (LDA) and Particle Image Velocimetry (PIV). The results are correlated with the data which has been obtained from microscopically observation and discussed by using models for aggregation due to particle diffusion in a laminar gas flow.

1. Introduction

Materials synthesis from a gas phase includes technological processes for functional and gradient materials (bulk and film), ceramic and metallic powders, semiconductor devices and components. Particularly high porous metal nanopowders, formed by gas synthesis methods, are useful as fillers in polymer matrix composites [2]. Such fillers improve the mechanical properties of the composite, especially under thermal cycling at a high level of the composite's electrical conductivity. These properties strongly depend on the powder morphology, which is mainly determined by the process of aggregation in an atmosphere of helium. The quality assurance for the resulting powders requires well-defined porosities and particle sizes.

It is also well known that in other fields of research, as for example 'crystal growth', 'aerosol synthesis technology' and 'combustion synthesis technology', buoyancy convection has a strong influence on the morphology of the resulting solid material. This paper describes and discusses the use of different laser visualisation and diagnostic methods for observing the process of particle aggregation under normal gravitation conditions.

2. Particle Processing Technologies

In a so called "bottom-up" approach advanced materials with tailored properties can be produced by synthesizing particles from the gas phase.

One of these techniques is the "Inert Gas Condensation (IGC)" technique, which is widely used in a laboratory scale (see figure 1) to produce metallic nanopowders of ultra-low ionic impurity content. Reliable processing data, so as suitable on-line diagnostic techniques are needed for a scaling up of this process to production rates of some kilograms per day, which are required for a profitable mass production.

It is necessary to distinguish between the different phases of this complex process, in order to get first experimental results, balance them one by one and finally optimise the complete process by improving the single process phases. The particle flow depends on the process parameters, which therefore have to be completely monitored and controlled. The following process parameters are essentially: the subatmospheric pressure in the downstream tube, the temperature of the 'boat' (an electric resistance furnace as vaporizer), the metal wire rate of feed into the 'boat' and the performance of the Roots vacuum booster, which induce the circulation of the helium gas at partial vacuum.
Regarding the production of particle nanopowders, the formation of primary particles and their precipitation is only of subordinate interest. Therefore the experimental examinations concentrate on the aggregation phase of the process.

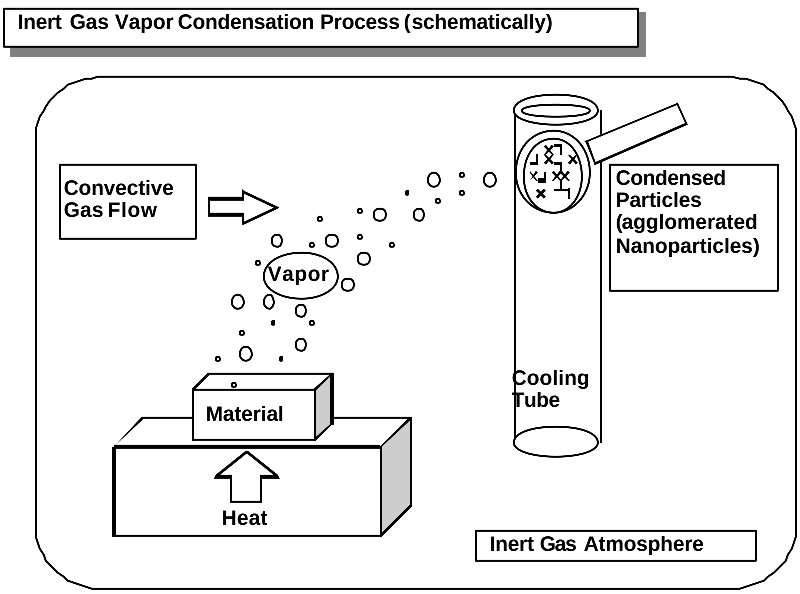


Figure 1: Principle sketch of the Inert Gas Condensation Process (IGC).

Flow measurement:

- Particle velocity
- Root Mean Square value
- Velocity distribution

Particle measurement:

- Particle size
- Particle size distribution
- Aggregation

Table 1: Requirements for on-line monitoring of metal nanopowders generation (aggregate size between 0.1

and 50µm).

Under normal gravity conditions the particle flow of the IGC process is influenced by thermal convection and by sedimentation effects of the aggregates. These effects lead to an undefined relationship between the attained morphology (porosity etc.) and the possible aggregation mechanisms. Regarding the flow, buoyancy effects affect the order of the turbulence and influence the fractal dimension of the aggregates. The turbulent flow also causes the aggregates to move in curved patterns or even along vortical paths within the gas and therefore leads to complications in observing the aggregates. Microgravity experiments allow the establishing of a steady state for particle coagulation with well-defined parameters. Such investigations will show the influence of thermally induced buoyancy convection on the spatial non-homogeneity of the particle size distribution and on the morphology of the particle aggregates. They will lead to a better understanding of the relationship between the parameter variation and the powder morphology.

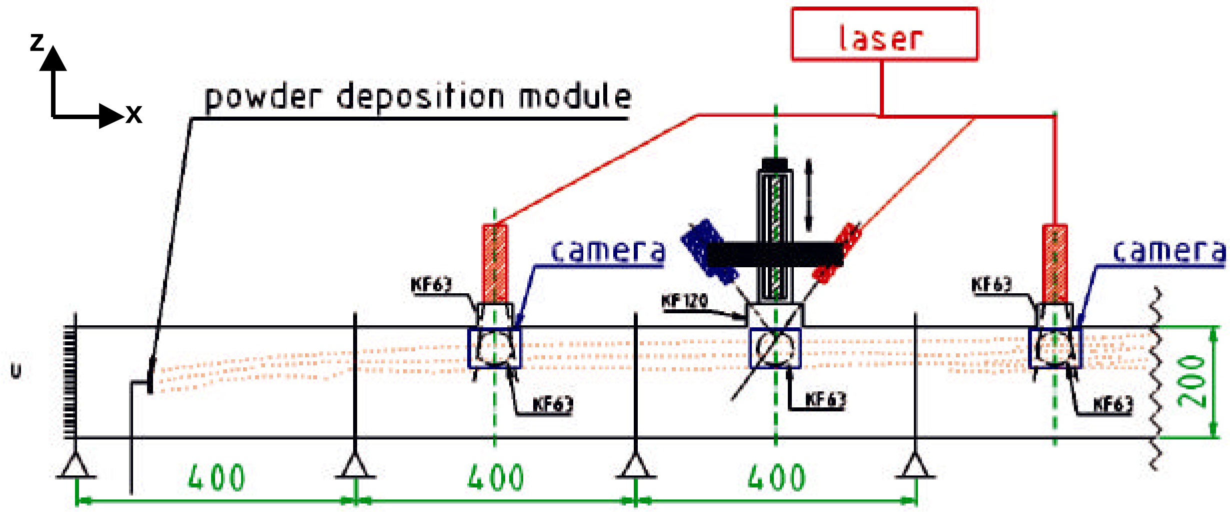


Figure 2: Principle sketch of the evaporator and the downstream tube.

3. Experimental set-up and measuring techniques

The experimental set-up was developed as an evaporator with a downstream tube (figure 2), for on-line monitoring of the metal nanopowders aggregation process in the helium atmosphere. The evaporator is the same kind which is used in real processes, whereas the downstream tube was especially designed to fit visualisation and measurement demands. The downstream tube consists of three identical sections which are provided with two pairs of observation flanges in rectangular arrangement. The different observation flanges along the reactors axis allow to observe the growing of the aggregates. The horizontal center line of the used flanges has a distance of x = 430 mm, x= 830 mm and x = 1230 mm, in the longitudinal axis, from the electric resistance furnace (generally called 'boat'). In vertical direction the flanges centre line coincides with the lateral axis of the tube. The 90° arrangement of the observation flanges was chosen to establish a laser light sheet optic, perpendicular to the observation camera, for classic flow visualisation and for 'Particle Image Velocimetry' (PIV).

4. Preliminary results

The process parameters, which are measured during on-line monitoring, are summarized in table 1. The most important parameters are the particle size as a function of time and place as well as the particle speed in the gas flow. A non-contact measuring method is recommended in this case. According to these requirements, it was decided to start with measurements of the velocity at three different downstream positions to obtain the local velocity development of the aggregating particles along the process reactor. Flow visualisation studies were done in addition, using laser light-sheet technique(figure 6 and 7) at all three observation flanges.

The coordinate system as shown in figure 2 is consequently used in all graphs and pictures: the longitudinal axis is called as x-axis, the horizontal axis is called as y-axis and the vertical axis is called as z-axis.

4.1 Flow visualization with laser light sheet technique an PIV generated light sheet images

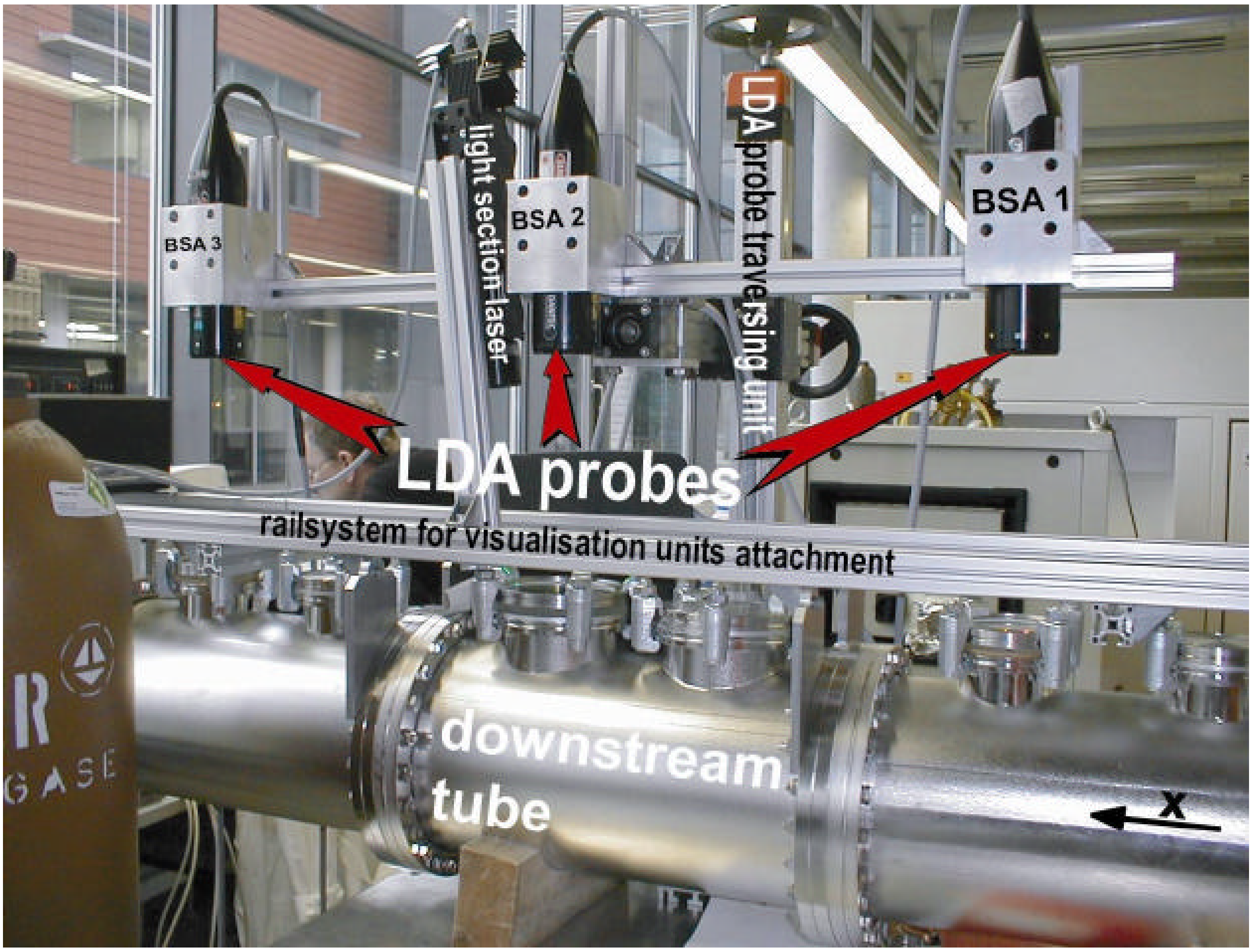


Figure 3: Photo of the LDV probe arrangement applied to the Inert Gas Condensation (IGC) process.

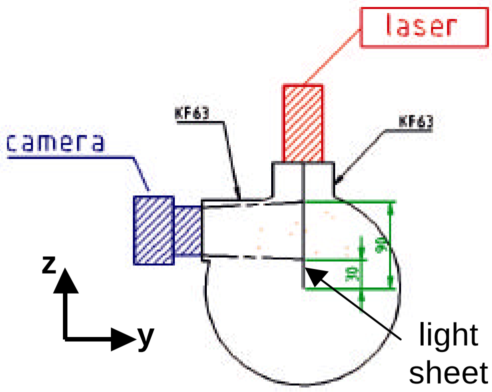


Figure 4: Sketch of the 90° visualisation arrangement (cross sectional view).

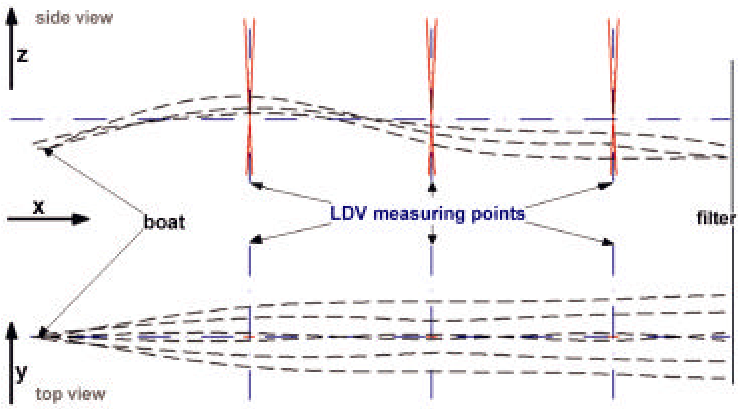


Figure 5: Principle sketch of the particle flow in relation to the LDV measuring plane.

A DANTEC FlowMap PIV system with a Kodak Megaplus ES 1.0 camera, was used for PIV measurements. The CCD sensor array of the camera has 1,008 (H) x 1,018 (V) light sensitive elements (pixel). These pixels are 9 µm square and have a centre to centre spacing of 9 µm.

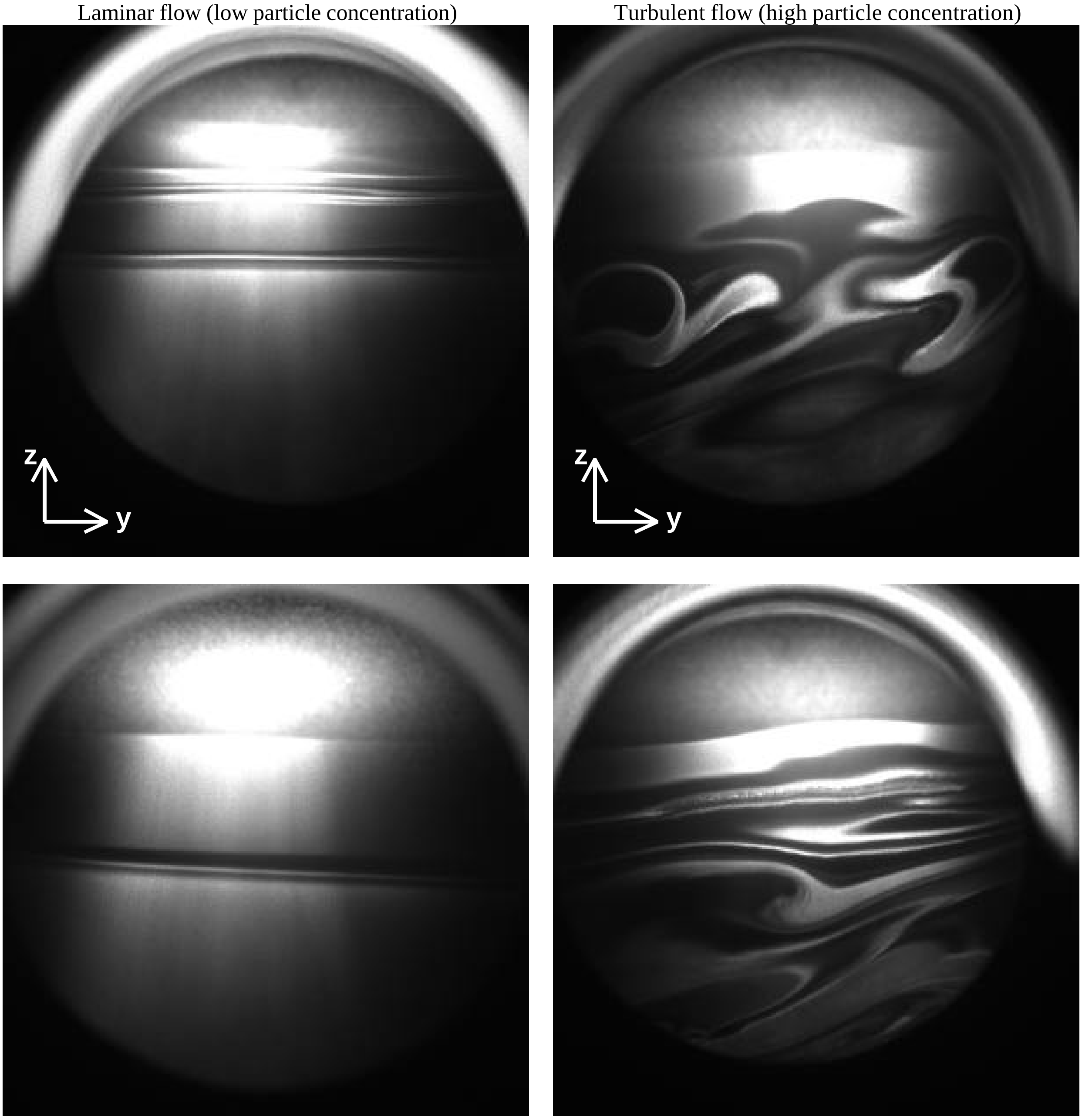
The PIV test measurements did not lead to satisfying results, due to problems with the resolving power of the CCD-camera lens. The resolution of the CCD-camera lens was not high enough to visualise the small aggregates (down to about 2 µm) inside the particle flow. But the PIV pictures were quite usable as light sheet flow visualization shots. Table 3 shows typical PIV pictures of laminar and turbulent flow in the downstream tube. The turbulent flow is mainly induced by the density of the metal vapour inside the tube, which is controlled by the metal

•	Roots pump	20 Hz
•	Metal wire feed	20 1/min
•	Helium vacuum	20 mbar
•	Furnace fower	1.4 V, 400 A
•		

Table 2: Process parameter during visualization experiments.

wire rate of feed into the ‘boat’. For further PIV measurements it is planned to use a lens-system with a higher resolving power to be able to resolve aggregates of at least a size of 1 µm on the CCD sensor array of the PIV amera.

Laminar flow (low particle concentration) | Turbulent flow (high particle concentration)



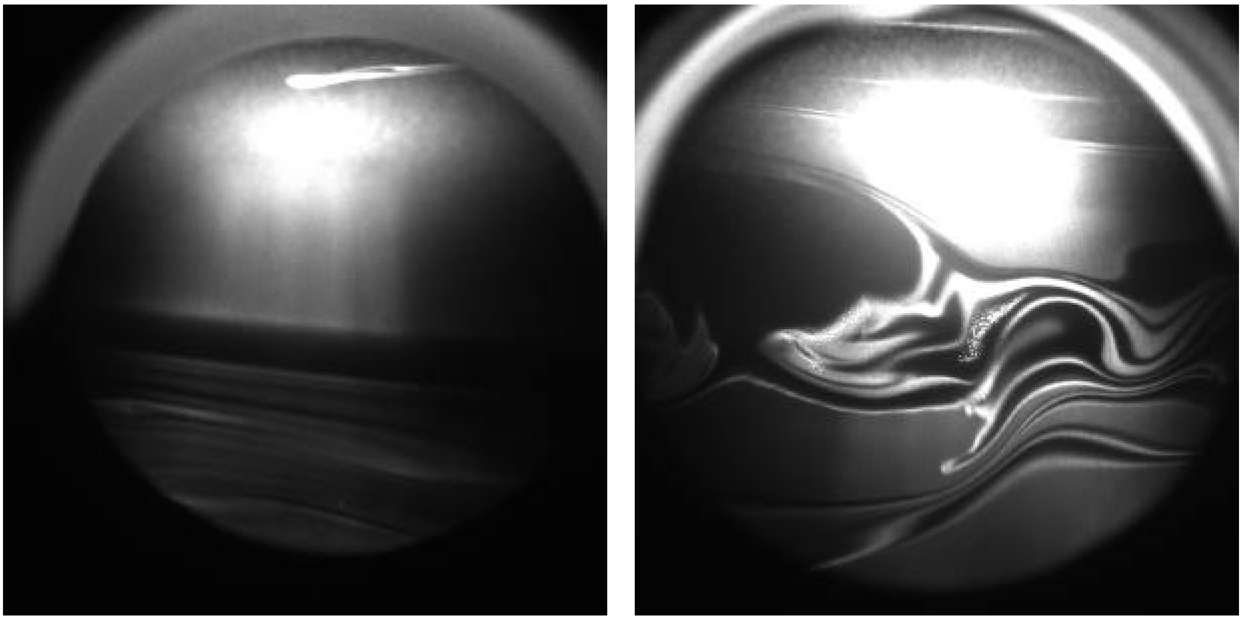


Table 3: PIV generated light sheet images taken at the second observation flange (x = 836 mm), with the light sheet established at the horizontal centre line of the downstream tube (image dimensions: x = 132,6 mm, z = 133,9 mm). The used process parameters are listed in table 2.

4.2 LDV measurements

Three different LDV (Laser-Doppler Velocimetry) optic probes, with a focal length of 400 mm (the technical data is listed in table 3), are installed at the flanges of the tube reactor: either at the horizontal or vertical observation flanges (figure 4). Different positions in radial direction of the tube reactor can be reached with a traversing unit. The LDV-probes are mounted (figure 3) on this unit.

The LDV measurement system consisted of three DANTEC BSAs and three 10 mW lasers and a Personal Computer for controlling. Each laser is connected to a probe, by a fiber-optical light guide. The dimensions of the downstream tube as well as the breadboard assembly, forced us to use a focal length of 400 mm for the LDA probes (table 4). The types of BSAs in use is shown in table 5.

The LDV measurements have shown that only a time-dependent LDV analysis is possible within the normal process parameters. It is more likely to get good conditions for steady state LDV measurements with laminar flow inside the downstream tube. Laminar flow can be achieved by a slow superimposed current and moderate heat of the ‘boat’ and thereby a low exhaustion of the gas flow with particle aggregates.

The velocity of the particle flow was measured inside the downstream tube. The velocities have been measured at different locations in the process as well as the velocity distribution, using the parameters in table 4. Representative preliminary results are summarized in the diagrams in figure 6 and 7.

focal length	400 mm
measuring volume:	
length	4.069 mm
diameter	0.190 mm
number of fringes	28
interference distance	6.69 µm

Table 4: LDV-System parameter.

BSA 1	BSA enhanced	57N21	Master	x = 431 mm
BSA 2		57N10		x = 836 mm
BSA 3	BSA enhanced	57N20		x = 1234 mm

Table 5: The DANTEC BSAs used for the LDV measurements.

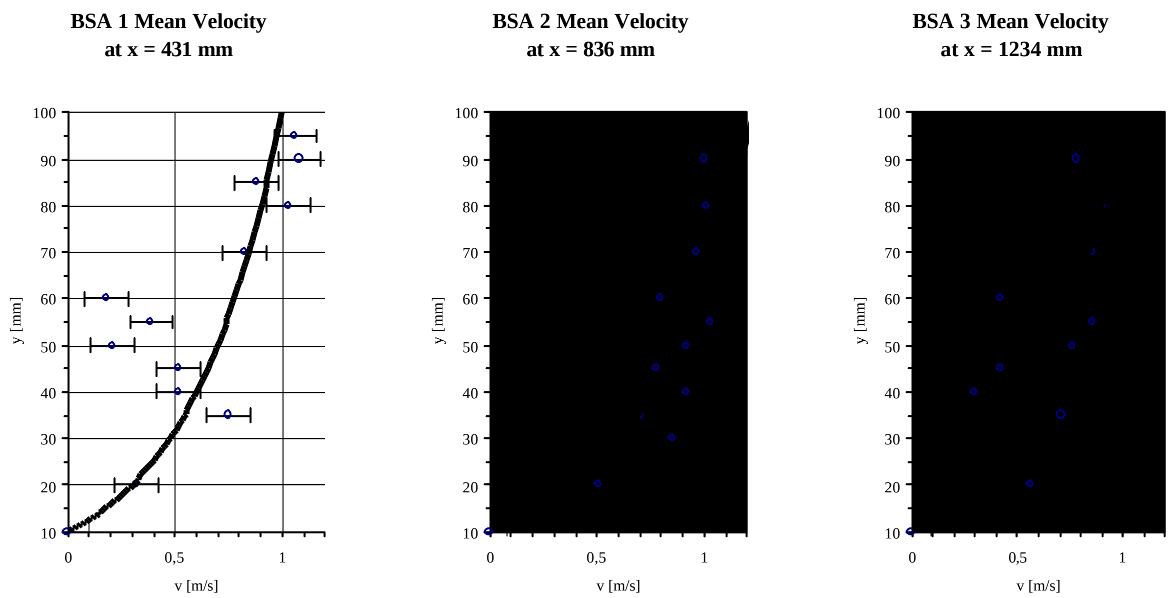


Figure 6: Velocity distribution at the three different observation points in dependence on the relative altitude (z = 10 to 100 mm, y = middle of the tube, Roots pump with 20 Hz).

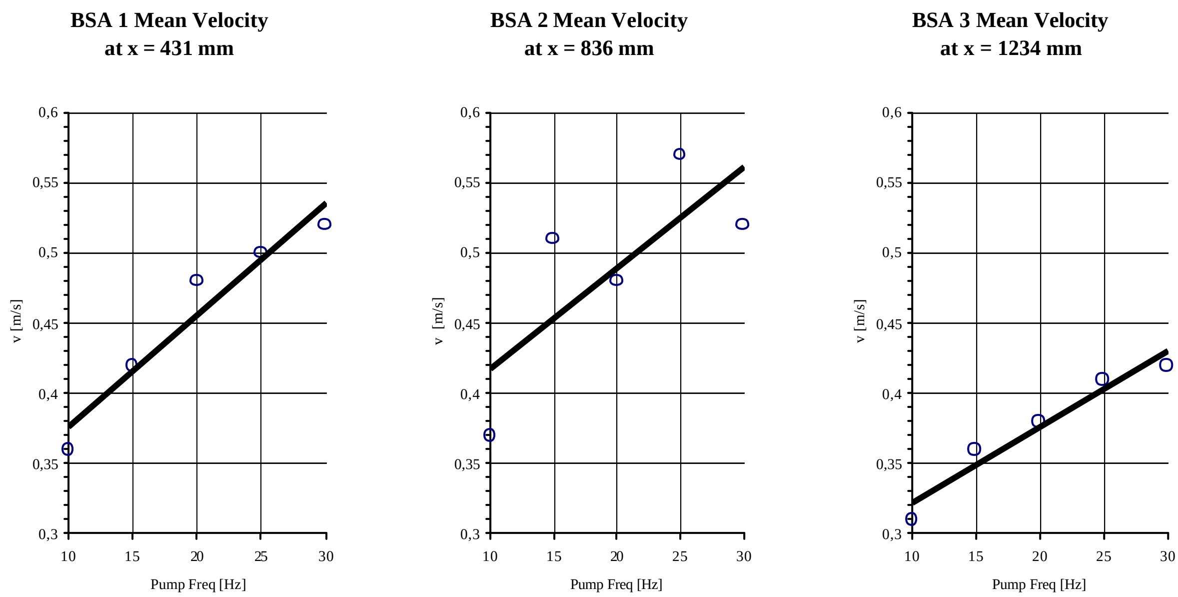


Figure 7: Flow rate in dependence on the power of the roots pump at a relative altitude of z = 55mm (y = middle of the tube).

Figure 10 shows a representative measurement of the pipe flow profile in the lower half section of the downstream tube. The LDV measurement volumes were placed at the three measuring points at x= 431 mm, x= 836 mm and x = 1234 mm (distance to the boat), varying the relative height in a range between z = 10 to z = 100 mm.

Unless it is very difficult to get reproductive conditions with the breadboard assembly there are always slight differences within the preset process parameters. Therefore the measurement results shown in figure 10 and 11 are not totally comparable (like at x = 55 mm and $f_{Roots\ pump}$ = 20 Hz), but they have both been taken at the process parameters shown in table 2.

Figure 7 consists of graphs of the flow rate according to the pumping level of the Roots vacuum booster, measured at z = 55mm at all three measuring points. In most cases we measured flow velocities between 0.3 and 1 m/s.

Unfortunately the above mentioned high focal length of the LDV probes cause a relatively large measuring volume. It is out of question that for further investigations the measuring volume has to get smaller for the optimal detection of the aggregates, because it is very likely that aggregates of a size of about 1 to 10 µm are to small for a measuring volume with a length of 4069 µm and a diameter of about 190 µm.

The conclusions which can be drawn from the results shown in figure 10 are, that we have a typical pipe flow inside the downstream tube which is manly influenced by the buoyancy convection of the particle aggregates. From the diagrams in figure 7 the influence of the power of the Roots pump can easily be drawn.

4.4 Particle size detection

Further investigations were done with a microscopic optic, which can observe a two-dimensional sheet of the particle flow (160 x 160 μm^2). This observation technique, as described in [1], gives information about the aggregate size in a range of $0.5\mu m \leq d \leq 50\mu m$. Furthermore, it is possible to calculate the density distribution of the vapour as well as the condition of the particle aggregation (figure 8). It was applied and optimised by the Astrophysical Institute at the University of Jena [1].

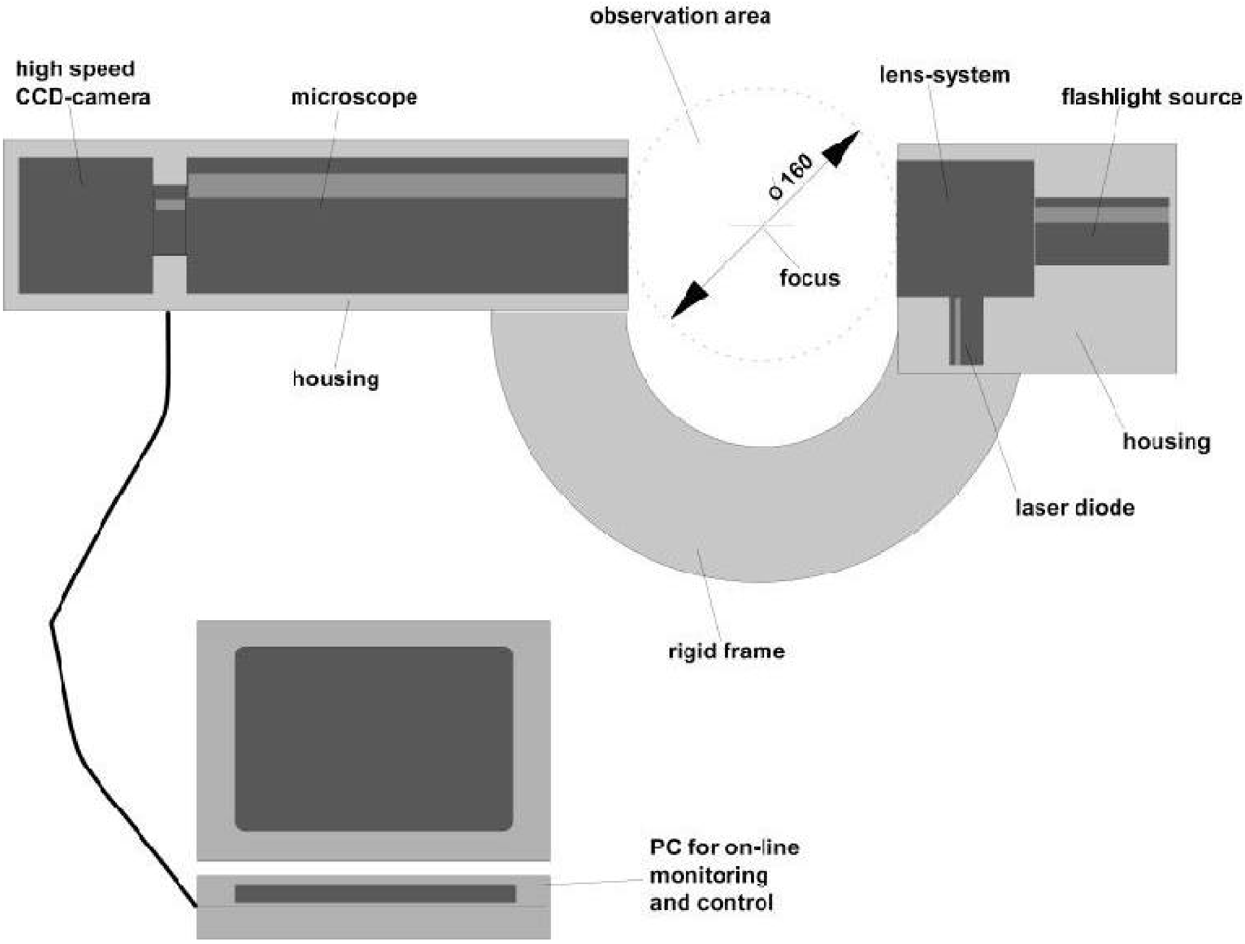


Figure 8: Principle sketch of the PATRICIA System [1].

A sample snapshot of the PATRICIA (Particle Tracking by microscopical imaging and correlation for insitu analysis) system is shown in figure 9. We were able to use the prototype of this particle flow analyser for the first investigations of the particle flow inside the downstream tube [3].

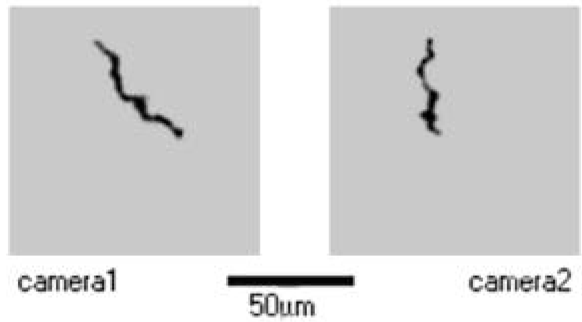


Figure 9: Snapshot of an aggregate in the CODAG experiment (front and side view). [1]

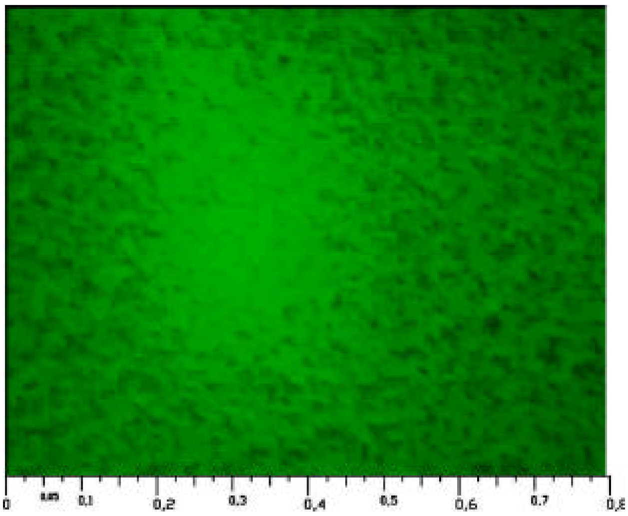


Figure 10: Snapshot of Particles between 2 and 10 µm on a glass substrate.

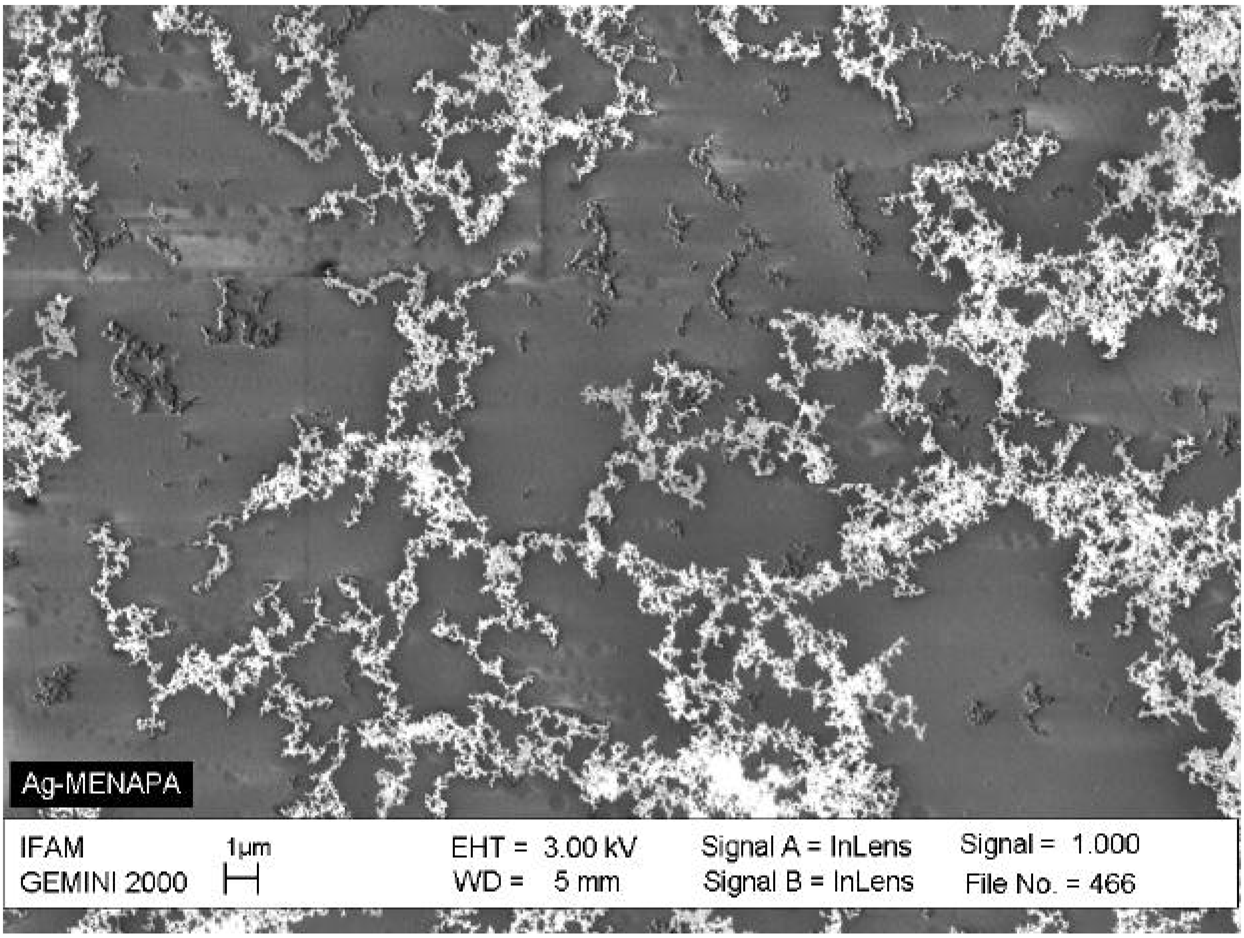


Figure 11: SEM picture of the particle layer on the glass substrate.

The PATRICIA system was also applied at the endflanges of the tube reactor, hence the illumination and the microscope unit had to be attached to the tube in a larger distance of 330 mm as these were calibrated for a focal length of 160 mm. Due to this larger distance we were not able to use the maximum potential of this measuring system.

In the end the particle agglomerates were not detected online during the aggregation process, but they have been detected on a glass substrate which was attached inside the tube, at a distance of 80 mm form the microscopic lens, in the focal plane. On this substrate, which is shown in figure 14, we were able to visualise particle aggregates of a size between 2 and 10 µm. The SEM image in figure 11 is taken from the same glass substrate sample and shows a closer look on the aggregates morphology. Further experiments with an optimised focal length and a better integration of the microscope are projected, which should lead to a practicable on line particle diagnostic.

Nevertheless the experiments and the diagnostic tools are relevant for other related phenomena occurring e.g. in ‘flame synthesis’, ‘combustion synthesis’ and ‘laser ablation technique’.

5. Outlook on the research under microgravity conditions

In the IGC-Process sedimentation of the aggregates (particularly at low pressure of the carrier gas) and thermal convection have an effect on the particle flow under 1-g conditions. This leads to an incomprehensible causal connection between the measured morphology of the nanopowder-aggregates (porosity etc.) and the possible aggregation mechanisms (diffusive restricted or ballistic aggregation). With experiments under microgravity useful information can be obtained about particle aggregation behaviour occurring in all above mentioned processes. Effects of thermal convection and sedimentation of the aggregates can be avoided.

Microgravity allows to set stationary conditions for coagulation under well defined parameters and to determine the influence of buoyancy induced flow and non-homogeneity of the particle density on the morphology of the particle aggregates (e.g. the form and porosity). This enables the assignment between parameter modification and powder morphology, and will point out new ways to control the formation of aggregates from sinter active nanoparticles in a carrier gas.

In a next step experiments under microgravity conditions shall be realised during parabolic flights. Unless in parabolic flights the quality of microgravity is not homogenous within all three acceleration axes, the complete laboratory experiment can be operated in horizontal or vertical line-up.

It is planned in a further step, to integrate the experimental set-up into the ‘European Drawer Rack’ (EDR) on the European module (Columbus) of the International Space Station (ISS) to extend the observation time from some seconds up to hours. Spatial restrictions in the EDR (figure 12) allow a maximum size of a 480 mm reactor. Hence, a miniaturized diagnostic system should be developed in time .

6. Conclusion

The various laser optical methods, which have been used to prove the quantitative measurement of the aggregates velocity and size, have shown some basic problems within the visualisation and measurement of nanoparticle aggregates within a range of few micrometers. The Laser light sheet visualisation shows different flow phases within the downstream tube. Thermally driven turbulent flow occurs under regular process parameters, while under laboratory conditions we could also generate a laminar particle flow for the LDV and particle size measurement purposes. The same flow patterns have been visualised with the PIV system. Just the PIV generated images have a higher resolution. Quantitative results from the PIV system were not achievable due to the low resolving power of the CCD–camera lens. The LDV measurements have shown that a time dependent flow always occurs, even at laminar flow. Though for better LDV measurement results it is necessary to have a smaller measuring volume. Particle aggregates between 2 and 10 µm were detected (but not in motion) with the PATRICIA system from Jena University. The Particles could only be visualised on a glass substrate which was placed in the focal plane of the microscope lens inside the tube. The aggregates size could be validate with SEM images. Further investigations will concentrate on the improvement of the PIV and the PATRICIA system.

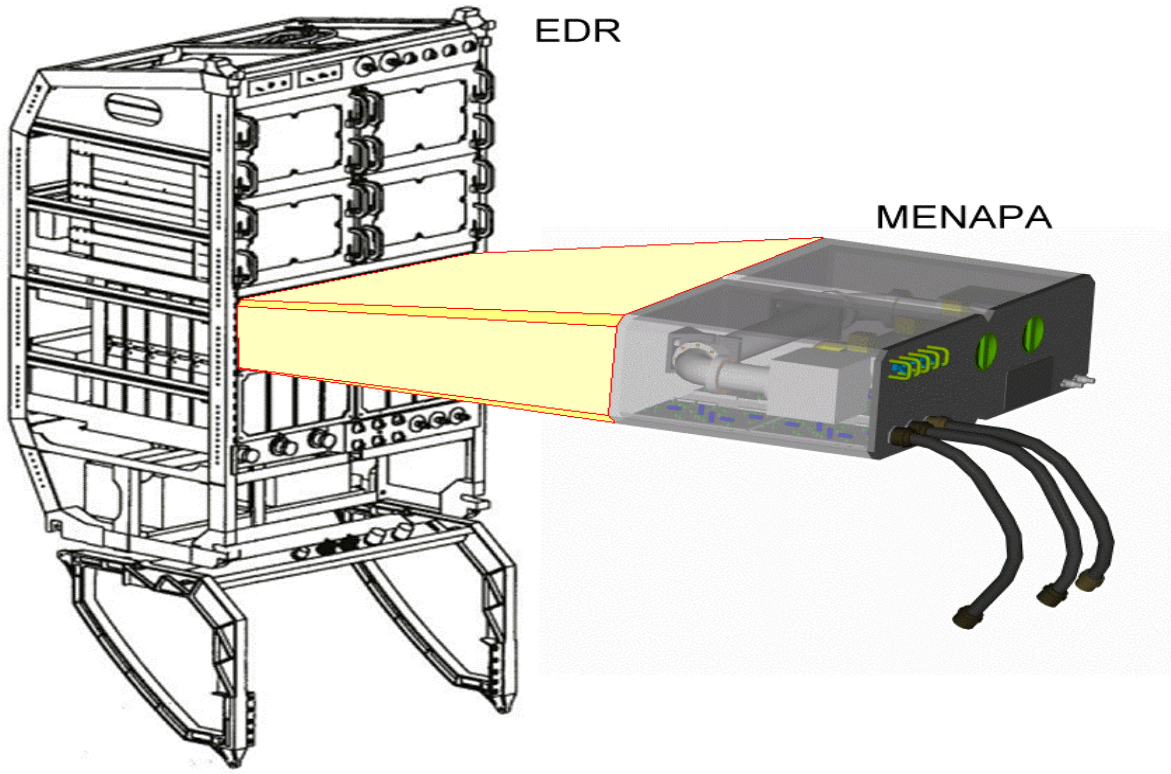


Figure 12: Integration of the modified tube reactor into the European Drawer Rack (EDR).

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7. References

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