DEVELOPMENT AND QUALIFICATION OF AN AEROSOL GENERATOR FOR INVESTIGATIONS UNDER THERMAL-HYDRAULIC SEVERE ACCIDENT BOUNDARY CONDITIONS

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ABSTRACT

During a severe accident, the largest part of the radioactive inventory inside the containment consists of airborne particles of different species. They are generated in the reactor pressure vessel and then released into the containment. The major aim of this project is to investigate the behavior of aerosols under the thermal-hydraulic boundary conditions of severe accidents with a core melt-down. This paper deals mainly with the generation of representative non-soluble aerosols under those conditions as well as the design of a test facility.
The initial idea was to design a test facility in which thermal-hydraulic boundary conditions of temperatures up to 200°C, pressures up to eight bar gauge, relative humidity up to 100 % and condensation conditions are possible to be investigated. Furthermore, particles are limited to solids of tin dioxide and silver. For the test facility a particle concentration was defined from 0.1 g/m³ to 5 g/m³ with a specific size distribution (aerodynamic diameter) less than or equal to 10 µm.
These requirements were a main challenge for the aerosol generation units. Therefore, a suitable unit for each material was designed to generate reproducible particles constantly for more than eight hours. For the non-soluble particles, silver and tin dioxide, a mechanical dispersing unit was designed. In the design, the flow and the particle discharge were optimized to increase the efficiency of the generation unit.

KEYWORDS
aerosol behavior, aerosol generation, containment phenomena, severe accidents

1 INTRODUCTION AND MOTIVATION

During a severe accident, the largest part of the radioactive inventory inside the containment consists of airborne particles of different species. They are generated in the reactor pressure vessel and then released into the containment. Much research in recent years has focused on developing codes to simulate the behavior of the aerosols in the containment [1, 2]. The main problem is that for simulations of these phenomena, experiments with a sufficient measuring technique and a high resolution are needed. Therefore, the measurement and the characterization of aerosols is a challenging area, especially the measurement of the particle size distribution and the relative humidity. Normally commercial aerosol characterizing measurement devices measure ex situ and under environmental conditions. That is the reason why commercial devices are mostly designed for ambient pressures and temperatures. Therefore, the investigated atmosphere must be conditioned to lower pressures and temperatures. With a national funded project called IN-EX, the question of if it is possible to develop and qualify optical measurement

devices to characterize airborne particles and to determine the relative humidity under the thermal-hydraulic boundary conditions of a severe accident with a core melt-down will be answered [3]. In this context a new test facility is under construction. With this facility, the thermal-hydraulic boundary conditions of containments are imitated so that different aerosols can be investigated at pressures up to eight bar gauge and, at the same time, temperatures up to 200 °C.

Especially if a severe accident with a core melt down appears at the end of a fuel cycle, any species from the periodic table can be found in the reactor core, including most of their isotopes [1, 2]. A wide range of species are strongly interactive with each other because of their chemical and physical characteristics. Those characteristics can change with the thermal-hydraulic boundary conditions in the reactor core or in the containment. As a result, a full investigation of this chaotic process is to complex. Therefore, representative materials must be selected and investigated to understand the basic behavior of these particles in combination with changing thermal-hydraulic boundary conditions. In general, the basis for the selection of representative materials was the type of the chemical bonding, solubility, hygroscopicity and the density. In this paper only the non-soluble materials are investigated. These materials are silver (metal, non-soluble, high density) and tin dioxide (ceramic, non-soluble, medium density).

In the design phase of the test facility it became evident that the aerosol generation would be a major problem. The reason for this was the necessity to generate these reproducible particles constantly for several hours. Another requirement was the variation of the aerosol concentration in a range from 0.1 g/m^3 to 5 g/m^3. When developing the measurement system, it was important to know if the devices could measure accurately at low as well as high concentrations. Commercial aerosol generators do exist, however they cannot fulfil all of the necessary requirements. Therefore, the aerosol generation device was redesigned. This paper deals with the design and the first results of the generation of tin dioxide and silver aerosols.

2 AEROSOL GENERATION UNITS FOR TIN DIOXIDE AND SILVER

There are several different methods to generate an aerosol from solid materials. The generation is mainly based on dispersing a powder into a gas flow. Different commercial systems exist to disperse the powder. A common method is to disperse the aerosol with a brush. In general these systems consist of a storage of the bulk material, a feed piston, a rotating brush, a gas inlet and an aerosol outlet. One commercial system is the RBG1000 made by Palas GmbH. In figure 1 the functional principle of this aerosol generator is shown.

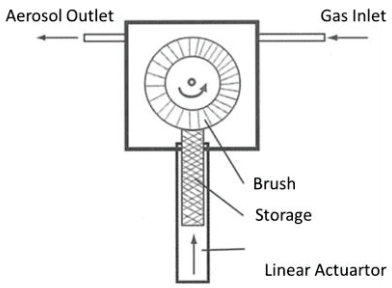

Figure 1. Functional Principle of the RBG1000 from Palas GmbH [4].

In the RBG1000, the bulk material gets moved to the rotating brush with a defined feed rate by a linear actuator. Above the brush, the gas flow is horizontally connected to the brush chamber. The function of the brush is to disperse and transport the bulk material into the gas flow. It is important that the gas

channel extends over the whole width of the brush with a rectangular cross section (mixing area). Furthermore, the top of the brush is in the flow channel so that the gas flow passes the top of the bristles of the brush. A nozzle follows the mixing area to create a circular cross section with an inner diameter of four mm.
A version of the RBG1000 is available to generate an aerosol to an overpressure of 3 bar. To check, if the system was suitable for the tests in general, an equivalent device without the resistance to pressure was analyzed. The main problems were the small storage tank for the bulk material and the deposition on the brush as well as the area where the particles and the gas flow (carrier gas) are mixed and the aerosol outlet. Although the resistance to pressure is limited to 3 bar gauge, the system could be safely operated up to a pressure resistance of eight bar gauge, if the whole system were to be set in a pressure vessel. Then the pressure difference between the pressure in the process and in the vessel could be compensated for or limited to 3 bar. Nevertheless, the main problem was the deposition in the aerosol outlet and the mixing area. To quantify the particle losses in the device, the output was measured with absolute filters. The result was that 45 % to 60 % of the material remained in the device. This is a problem because these particles block the flow. The result is that a consistent generation for a long time is not possible with this device. This was the main reason to design a new aerosol generator called “Powder Dispersing unit with a Brush” (PDB) which is based on the RBG1000. Figure 2 shows the deposition on the brush (left) and the mixing area and nozzle (right) at a generation of 5 g/m³ after 30 minutes.

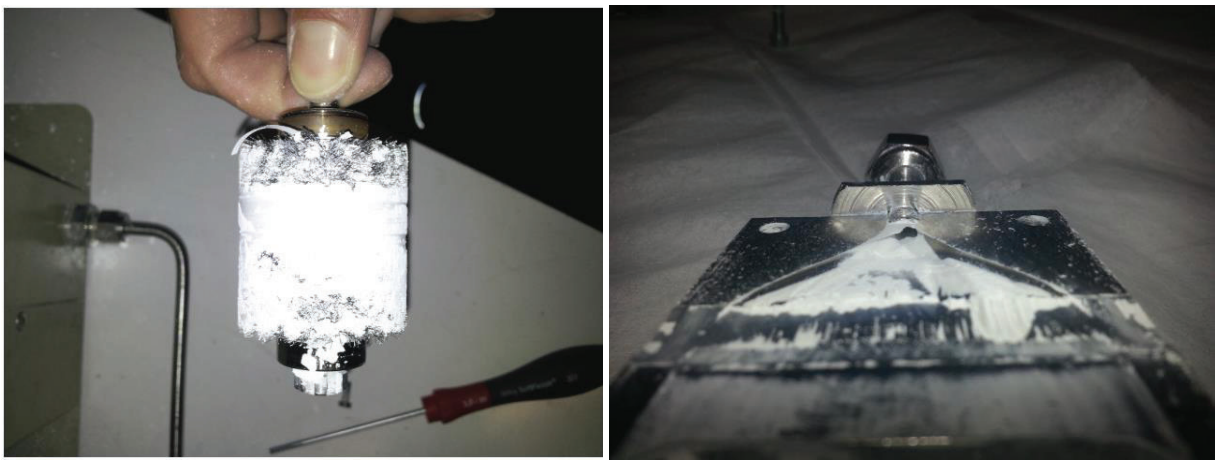

Figure 2. Tin Dioxide Deposition on the Brush (left) and in the Nozzle and the Mixing Area (right) of the RBG1000.

2.1 Technical Design and Important Changes of the PDB

Similar to the RBG1000, the PDB consists of two power trains. One pushes the bulk material towards the brush with a linear actuator and the other drives the brush with an electric engine. The power train of the brush and the brush chamber in which the brush disperses the material are decoupled from each other by a radial oil seal. With this seal, the particles, as well as the overpressure, cannot enter the environment or the drive housing of the power train. To pressurize the whole system, the feed piston, which is connected to the end of the linear actuator, is sealed with two O-ring seals parallel to each other. The basic design is shown in figure 3.

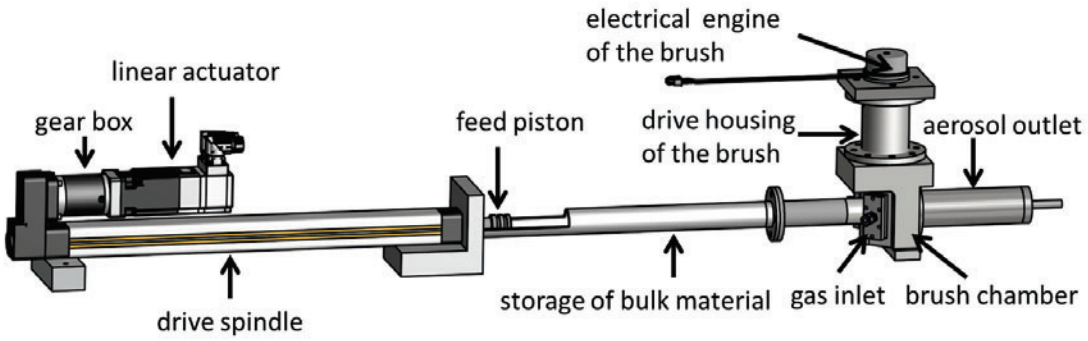

Figure 3. Overall Technical Design of the PDB.

The main requirement for the technically redesigned device was to produce as little deposition in the aerosol generator as possible. Therefore, the technical design of the PDB focuses on the flow of the gas and the aerosol. As a result, the injection of the gas flow was allocated from one horizontal flow over the brush to two vertical flows around the brush. The flows are configured in such a way that the gas flows through the whole length of the bristles. In figure 4 the functional principle of the PDB is shown.

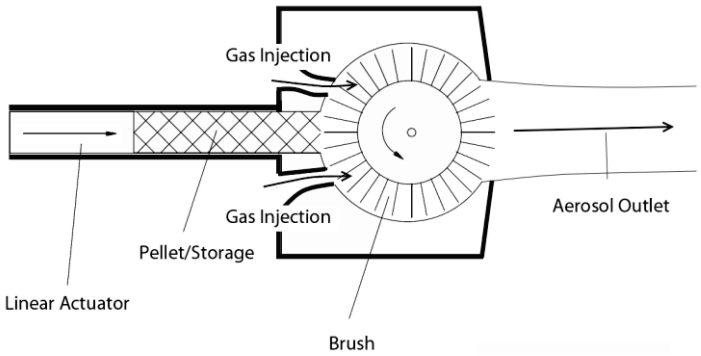

Figure 4. Functional Principle of the PDB.

To reduce the deposition in the PDB, the device was constructed without sharp boarders. Therefore, the aerosol outlet was designed as a nozzle with a small angle of the taper. The cross section of the brush with respect to the outlet of the brush chamber is a rectangle. To create a circular cross section, the inner surface of the nozzle was adjusted with a fluent transition from a rectangle to a circle. The circle was increased to an inner diameter of 10 mm. Figure 4 shows that the gas injections and the aerosol outlet merge seamlessly into each other. Additionally, the free space was reduced to a minimum around the brush. What this means specifically is that the distance is only 0.5 mm between the end of the bristles and the inner wall of the brush chamber. As described before, the gas inlet is on both sides next to the brush. Nozzles are used to increase the impact of the gas flow on the brush. They can be changed so that it is easier to clean the brush chamber and to allow for the possibility to investigate the impact of the gas velocity on the discharge of the dispersed particles. The main characteristic of the nozzles is their outlet area. It consists of a slot perpendicular to the middle of the bristles over the total length of the brush. The external shape of the outlet area is adjusted to the inner form of the brush chamber to prevent edges at the transitions. Behind the slot is a free space so that the gas gets distributed evenly as a gas blanket. Figure **5** shows a section through the gas inlet nozzle.

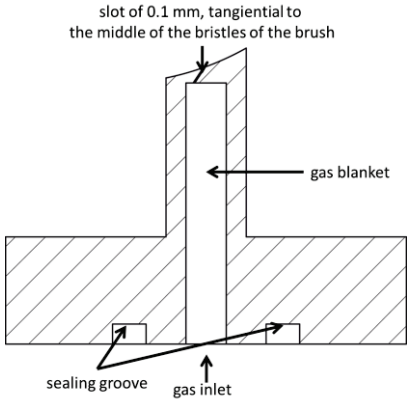

Figure 5. Section Through the Gas Inlet Nozzle.

2.2 Qualification of the PDB with Tin Dioxide

Tin dioxide was used for the qualification of the PDB. Tin dioxide is a ceramic and therefore dimensionally stable. Therefore, it is possible to press pellets from the bulk material. The main advantage to this is that the density of the pellets is distributed more homogeneous than in the bulk material itself. Experiments have shown that the pellets reach reproducible densities of about 3.500 kg/m³. If pellets are used, another advantage is the possibility to use the PDB vertically as well as horizontally. As a result, the application becomes more flexible.

In the qualification phase, the amount of deposition in the device, the impact of the rotation speed of the brush, different mass flows of the carrier gas, different nozzles for the gas injection and different feed velocities of the linear actuator were investigated. Additionally, the long-term stability of the output was considered. During the qualification phase two experimental setups were used. For analyzing the deposition in the PDB, an absolute filter was directly connected to the aerosol outlet of the PDB. The result was that only 15 % to 33 % of the material remained in the device. Thereby, the time of each measuring conducted to 50 minutes at a mass output of about 36 g/h. This examination was repeated five times. The experimental setup is shown in figure 6.

Figure 6. Experimental Setup of the PDB for the Qualification of the Deposition in the PDB.

For the other experiments, a Scanning Mobility Particle Sizer (SMPS) and an Aerodynamic Particle Sizer (APS) from TSI Inc. were used. With these systems, the particle size distribution was detected for each experiment and then compared. Figure 7 shows the experimental setup for the qualification phase of the PDB with the aerosol measuring devices APS and SMPS.

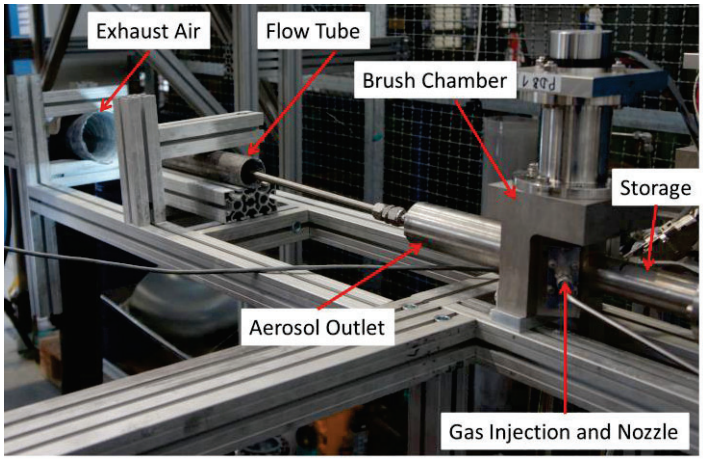

Figure 7. Experimental Setup of the PDB for the Qualification of Tin Dioxide.

For these experiments, an isokinetic sampling was needed, because only this sampling method guarantees the same size distribution of the aerosol in the sampling line as well as in the flow tube [5]. Therefore, the velocities had to be adjusted between the aerosol outlet and the sampling volume. Moreover, a flow tube with a larger diameter was attached to the aerosol outlet, as shown in figure 7. Due to the wider diameter, the velocity of the aerosol decreases. This deceleration is needed to increase the cross section of the sampling line. To measure a developed flow in the flow tube, a sample is taken after a run-in distance of about ten times of the diameter of the flow tube.

The impact of the rotation speed of the brush on the particle size distribution was also investigated. The revolutions per minute of the brush were varied between 500 rpm and 2250 rpm. Figure 8 shows the changing particle size distributions at the different revolutions per minute with a constant feed rate of 10 microns per second and nozzles of 1 mm width.

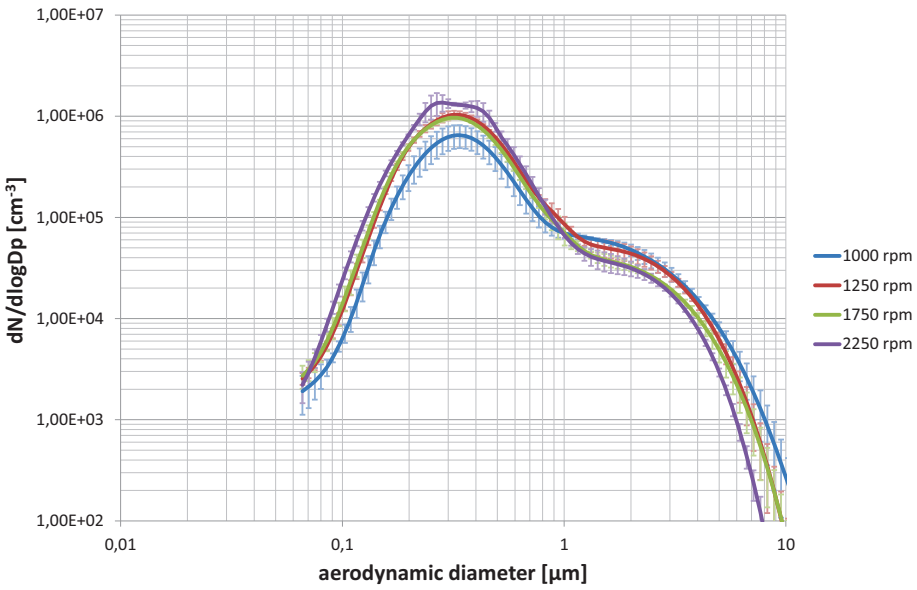

Figure 8. Particle Size Distribution at Different Rotation Speeds of the Brush.

The study shows that the particle count increases significantly with the increasing rotational speed of the brush up to 1250 rpm. In the case of a higher rotation speed, the particle count only increases slightly. Additionally, it is of special interest that there were a greater number of particles with an aerodynamic diameter above one micron observed at a lower rotation speed of the brush than at higher speeds. This phenomenon makes sense because at a lower rotation speed and the same feed rate, the brush has to disperse the same mass as at higher rotation speeds. Therefore, the brush has to carry off more particles in one revolution than at higher rotation speeds. The consequence is less dispersion which causes larger particles.

The impact of the feed velocity was also investigated with relation to the mass output and its related particle size distribution. To accomplish this, the feed rate was varied from 5 microns per second to 40 microns per second. With these feed rates, a theoretical mass output can be produced between 36 and 290 g/h. Furthermore, the particle concentration depends on the gas flow. So, a theoretical particle concentration can be reached between 1 and 8 g/m³ at a volume flow of 6 m³/h. The real output is decreased by the deposition in the device and the tubing. The deposition in the tubes can be neglected for the qualification phase because the tubes used are very short. As described before, the deposition amounts to between 15 % and 33 %. The particle size distribution is shown in figure 9 for a variation of the feed rate and a constant volume flow of 6 m³/h.

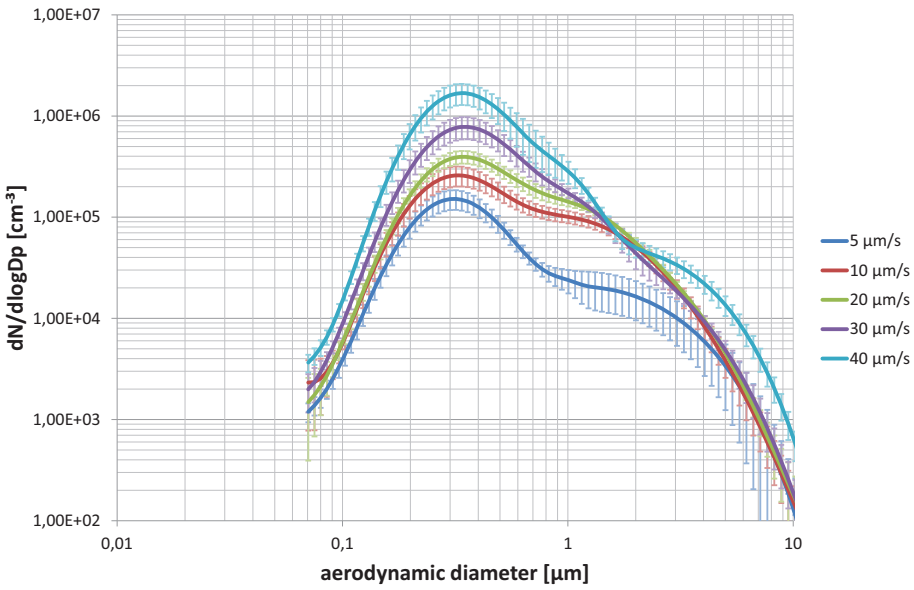

Figure 9. Particle Size Distribution at Different Feed Rates.

Figure 9 shows a clear increase in the number of particles in correlation with an increasing feed rate. Especially at particle sizes up to one micron, the particle count increases and shows a comparable behavior for the different feed rates. The difference in particle count for feed rates between 10 and 30 microns per second decreases for diameters between 0.7 and 1.5 microns. At diameters larger than 1.5 microns, a significant difference in their particle count is not apparent. For a feed rate of five microns per second the particle count decreases between diameters of 0.35 microns and 2.5 microns. Above 2.5 microns the size distribution approaches that of the feed rates of 10 to 30 microns per second. For a feed rate of 40 microns per second, the size distribution is comparable to feed rates between 10 and 30 microns

per second up to two microns. However, above 2 microns the particle count increases and shows a parallel course to the other feed rates. Nonetheless, what is striking is that the peak of the size distribution is nearly constant at a particle diameter of about 0.35 microns.
Another influence on the aerosol generation is the mass or volume flow. It is important because of the particle concentration in the aerosol and its probability of agglomeration. At increasing particle concentrations in combination with a smaller volume flow rate, the possibility of an interaction increases between two or more particles. This leads to a shorter free length of the generated primary particles and causes the formation of larger particles. Figure 10 shows the different particle size distributions for a constant feed rate of ten microns per second while the volume flow rates were varied.

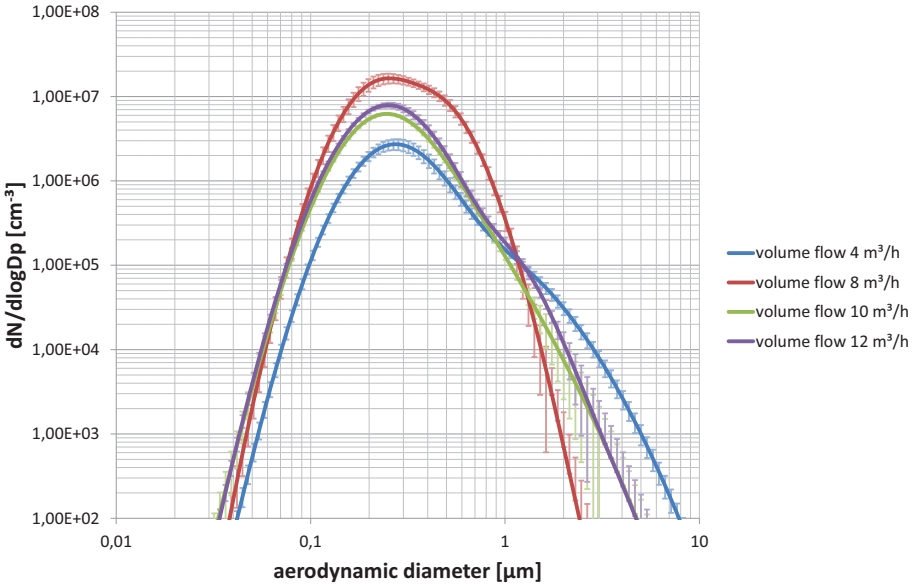

Figure 10. Particle Size Distribution at Different Total Volume Flows.

It is important that at the two gas injection nozzles the flow rate is equally distributed. The study shows a typical single mode particle size distribution for a total volume flow rate of 8 m³/h. The particle size distribution covers aerodynamic diameters between 0.04 microns and 2.5 microns. The peak is between 0.3 microns 0.5 microns. The particle count decreases rapidly after the peak indicating a high number of especially small particles. Particles cannot be reliably detected above 2.5 microns. The distributions of the total flow rates for 10 m³/h and 12 m³/h are comparable to each other. They show only a few differences in the range between 0.8 and 3 microns and the number of particles in their peaks. Up to 0.1 microns, the distributions for 8 m³/h, 10 m³/h and 12 m³/h are similar to each other. After 0.1 microns, the gradient is lower for the higher volume flow rates so their peaks are much lower than at 8 m³/h. Additionally, the peak of the graph for 8 m³/h is wider compared to the higher volume flow rates. At about 1.3 microns the three graphs cross each other. This is a point of inflection for the volume flow rates of 10 m³/h and 12 m³/h. A reason for this can be the higher velocity at the outlet of the injection nozzles leading to more turbulence in the device. Therefore, a higher transverse exchange results in the device leading to a higher interaction between the particles. Finally, this causes a higher agglomeration to occur and larger particles are measured. At a total volume flow rate of 4 m³/h, the graph for the particle size distribution has nearly the same characteristics as the flow rates of 10 m³/h and 12 m³/h. Compared to the higher flow rates, the gradient to the peak is the same but shifted to larger particles. The same applies to the peak, although it is also lower. However, at a diameter of about 1.3 microns, there is again an even more distinctive point of inflection. This leads to an increased and larger spectrum of the particle size. Due to the fact that the

volume flow rate is lower, a lower velocity arises behind the gas injection nozzle leading to a worse dispersion quality. Because of the slower velocity, the particles stay in the device longer. So, the opportunity to interact increases and the particles can agglomerate leading to larger particles.

To allow for the possibility to investigate its impact, the gas flow has to be regulated with mass flow controllers at each nozzle. Due to the fact that there are two nozzles, their impact is analyzed by using different gas injection rates of volume flows through each nozzle. This is shown in figure 11 for a total volume flow of 8 m³/h and a feed rate of 10 microns per second.

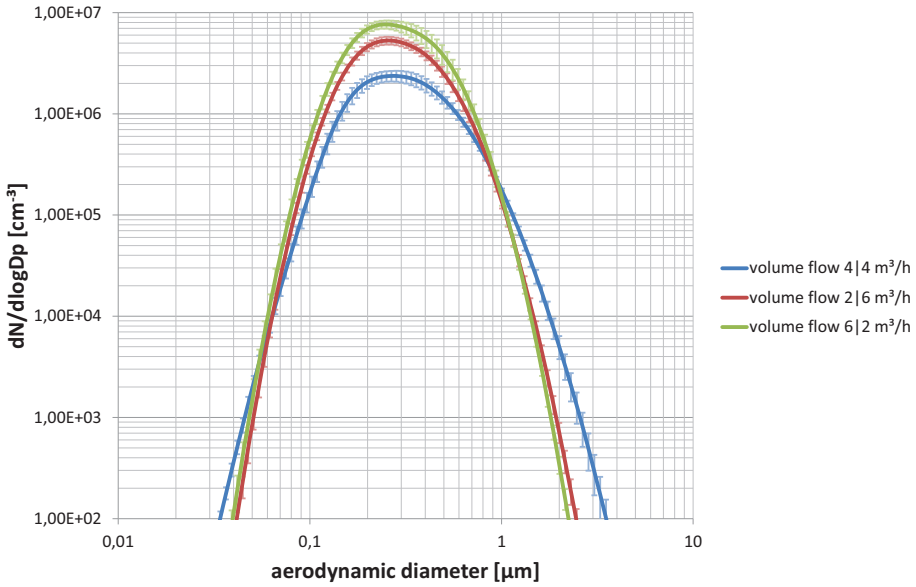

Figure 11. Particle Size Distribution for a Total Volume Flow of 8 m³/h at Different Gas Injection Rates.

Figure 11 shows three particle size distributions. The blue line is the presented particle size distribution with the same flow rate through each gas injection nozzle (compare figure 10). The red line represents the higher flow rate of 6 m³/h on the side where the directions of the gas injection and the rotation of the brush are opposed. The experiment for the green and the red lines are nearly identical. The difference is the distribution of the flow rates at the gas injection nozzles. The green line represents the higher flow rate of 6 m³/h on the side where the directions the gas injection and of the rotation of the brush are the same. Between both lines, there are negligible differences. However, the differences between an equally distributed and a non-equally distributed flow rate at the gas injection nozzles are interesting. The equal distributed gas injection has a lower peak and wider size spectrum. This is caused by the fact that at the nozzle with the higher flow rate, the impact on the brush and the dispersed particles is higher. The consequence of a higher flow rate is that the particles are better dispersed and the deposition in the brush gets reduced on one side.

To investigate the impact of these nozzles, nozzles with two different slot widths were manufactured. One has a width of 0.1 mm and the other of 1 mm. So, with those nozzles, different impulses and velocities can be set. For manufacturing reasons a slot thinner than 0.1 mm is impossible for the given configuration (see figure 5). At a volume flow of 6 m³/h, velocities in the nozzle can be established, from a purely mathematical point of view, of about 24 m/s (slot width 1 mm) and 245 m/s (slot width 0.1 mm). Additionally, by changing the nozzle size for the gas injection, the deposition on the brush and in its

chamber can be analyzed. The deposition rate directly influences the particle size distribution. Especially if the deposition in the brush is higher, its ability to disperse the particles is worse. Figure 12 shows the impact of different nozzle widths at a constant volume flow rate of 8 m³/h.

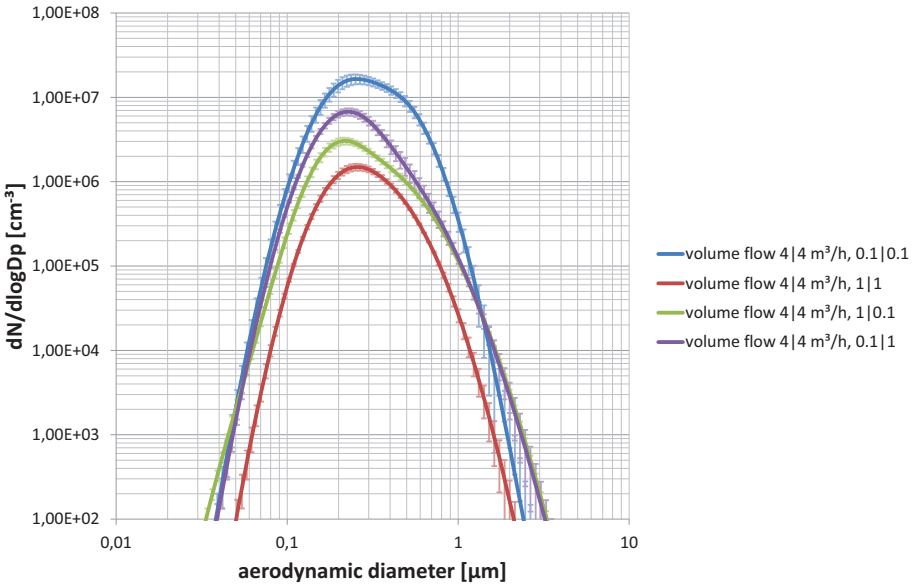

Figure 12. Particle Size Distribution for a Total Volume Flow of 8 m³/h with Different Gas Injection Nozzles.

Figure 12 shows four particle size distributions. In the experiments represented by the red (1 mm) and blue (0.1 mm) lines, the nozzles with the same width were used at both gas injection locations. For the purple and green lines both widths of the nozzles were used. With the purple and green lines, the impact of their location was analyzed. This is comparable to the study of the different gas injection rates. It is shown in figure 12 that the width of the nozzles' slots has an impact to the deposition and the particle size distribution. Although the characteristics of the blue and red lines are similar to each other, the main differences are the height of the peak and the smaller particle size spectrum. The differences in the spectrum lead to a higher deposition in the device. This is caused by a lower impact to the brush because of the wider nozzle cross section. It is important that their maxima are located nearly at the same diameter. The two experiments with different nozzle widths are similar to each other and are in the range between the other experiments performed with similar nozzles. A difference is only seen near the maxima. The peak is higher for the experiment with the smaller slot on the side where the injection flow is in the same direction as the rotating direction of the brush (purple line). Here the high velocity gas from the nozzle does not decelerate the flow of particles because they are both in the same direction. The result is a better dispersing property of the device.

2.3 Feasibility Study for a Silver Generation

In contrast to tin dioxide, silver as a metal is too ductile to press pure pellets. A pure silver/gas aerosol is desirable to provide the flexibility to generate a multi-component aerosol with different concentrations and amounts of different source materials. In a first test, compound pellets consisting of silver and tin dioxide were manufactured and investigated. Those pellets can carry up to 25 w/o silver. Acquiring a homogenous mixture out of the two bulk materials is an extensive process. This is due to the fact that the

two materials have a large difference in their density. To reach a homogeneous mixture both bulk materials were mixed up in a tumble mixer over a long period of time and pressed directly to pellets to avoid the separation by gravity. After the manufacturing process of the pellets, they were dispersed in the PDB. Figure 13 shows a scanning electron microscope image of the dispersed composite pellet of tin dioxide and silver containing an amount of 5 w/$_o$ silver.

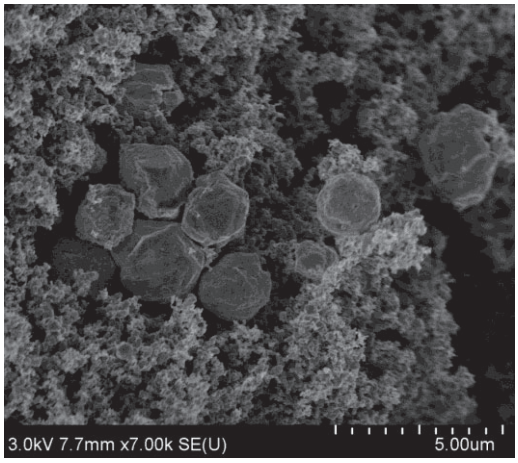

Figure 13. SEM Image of a Dispersed Compound Pellet of Tin Dioxide and Silver (5 w/$_o$).

The study has shown that, in principle, the generation of a composite pellet consisting of silver and tin dioxide is possible. The problem is that irreversible interactions were detected between the primary particles of silver and tin dioxide. They led to deformations of the primary particles especially in the more malleable silver. Another problem is the small amount of silver, with a maximum of 25 w/$_o$. At higher amounts of silver, the pellet cannot be manufactured because the ductility of the silver reduces the stability of the pellet.

As described before, pure airborne particles of silver are used to increase the flexibility of the aerosol generation as well as the usage in the different experiments. Figure 14 shows the experimental setup of the PDB for the qualification of silver.

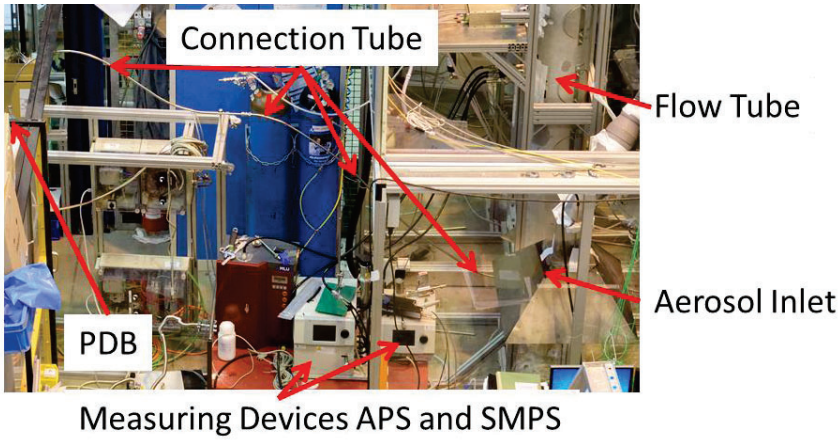

Figure 14. Experimental Setup of the PDB for the Qualification of Silver.

As shown in figure 14, the PDB was set up in the vertical direction. The experimental setup was different to the qualification phase for tin dioxide because the actual task for this experiment was to measure the same aerosol with different types of aerosol measurement devices. The setup consists of the vertical PDB. The aerosol is injected into a flow tube with a diameter of 300 mm in which the aerosol is analyzed. Both the flow tube and the PDB were connected by a tube. Due to height differences between the flow tube and the PDB, the connection tube had to be deflected smoothly so that the aerosol flow was only minimally affected.

Nevertheless, in a first feasibility study, without any measuring devices, the setup was equivalent to the setup shown in figure 7. In both experiments, the bulk material was placed into the storage from the brush chamber. It had a density of about 6.000 kg/m³ to 6.500 kg/m³ in the storage. The bulk material was not compacted, because the silver powder flows easily and could have been discharged out of the storage. Additionally, the ductility of silver leads to modifications of the primary particles, as seen in the compound manufacturing process, and at higher pressures to galling. Due to the high density of the material, the feed rate of the linear actuator is very slow. In the experimental setup, shown in figure 14, the feed rate amounts to 1 micron per second and led to an aerosol concentration of 1 g/m³. The experiment started with a zero metering of the ambient air as the brush was rotating. After this the gas injection in the PDB was started with a volume flow of 6 m³/h on each side. A high concentration was directly seen, although the feed piston was not moving. After a short time the concentration decreased rapidly. Then the feed rate was set to 1 micron per second and the concentration was nearly constant at a very low level comparable to the ambient air. The concentration continuously increased in the next time period of about 1.5 hours. After this the concentration and the particle size distribution were constant for another 2.5 hours. Afterwards the experiment was stopped. Figure 15 shows the particle size distribution of silver at a feed rate of 1 micron per second and a brush rotation speed of 1.800 rpm for the constant generation period of 2.5 hours.

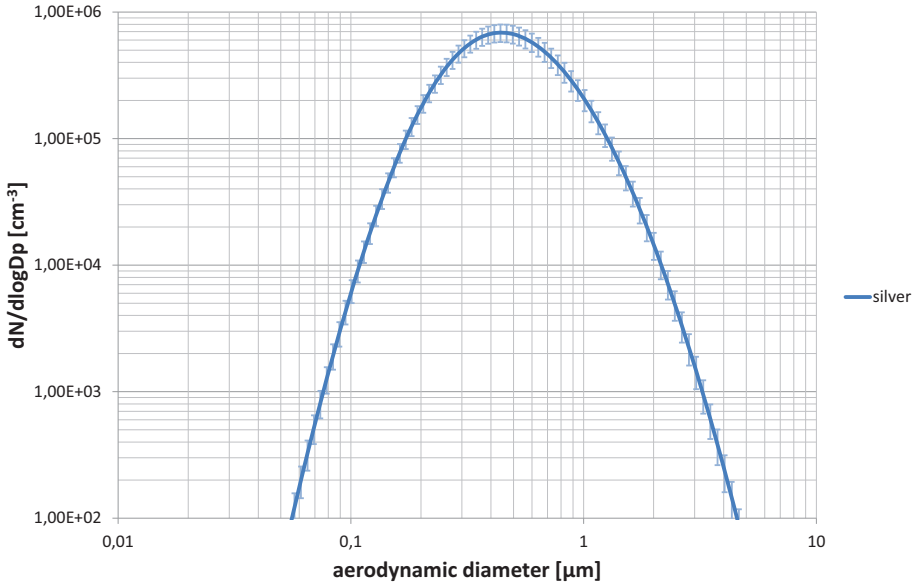

Figure 15. Particle Size Distribution for a concentration of 1 g/m³.

The reason for a direct high concentration is the high flowability of the silver bulk material. At the time when the gas injection started, an amount of the gas flowed in the direction of the storage of the bulk

material. Particles were fluidized and were transported to the aerosol outlet. So, the top part of the storage was directly discharged at the beginning of the gas injection. Additionally, the velocity of the feed piston was very slow, so that it took about 1.5 hours to balance the aerosol output and the tracking of the bulk material. A possibility to reduce the impact on the bulk material is to soften the gas injection.

3 CONCLUSIONS

As shown before, the new design of the dispersing unit can constantly produce a consistent aerosol over the course of several hours. The changes in the design by reducing the free space around the brush, the new arrangement of the gas injection and the small nozzle slots are responsible for the low deposition in the device. A high velocity behind the nozzles is especially important to increase the impact to the material from the bristles of the brush. With this high impact, the brush gets cleaned during the operation leading to a high reproducibility of aerosol generation. It is an advantage of the PDB that pure, compound pellets and a pure bulk material can be dispersed. With these properties, ductile materials, e.g. silver, can be dispersed with the system. Moreover, compound pellets, consisting of different bulk materials and degrees of hardness, can be dispersed with the PDB. With a precisely adjustable linear actuator, very low and even very high aerosol concentrations can be produced. The different studies have shown that the characteristics of the aerosol can be changed easily by a variation of the brush rotation speed, the volume flow rate and the feed rate. This makes the PDB very flexible so that the system can easily handle different demands of several experiments. The optimum operation conditions are: a rotation speed not lesser than 1250 rpm, a total volume flow of 8 m^3/h or higher, the volume flow shall be evenly divided over the two gas injection nozzles and the use of the small gas injection nozzles on both sides.
For the sake of completeness, it should be noted that the resistance to pressure of the PDB was only tested with a simple pressure test. The system withstood a test pressure of 13.2 bar gauge over 30 minutes without a significant pressure loss. As of yet, a qualification has not been done to analyze the particle size distribution at higher pressures or temperatures above ambient conditions. For those analyses, the full test facility is needed.

4 NOMENCLATURE

APS	Aerodynamic Particle Sizer
PDB	Powder Dispersing unit with a Brush
SMPS	Scanning Mobility Particle Sizer

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