8. Conclusions

There is a great deal of interest today in the special properties of nanoparticles and their potential applications. An overview of the potential functional applications, which include electronic, optical and magnetic applications, based on these special properties shows the broad range of possible applications. Most applications demand some control over particle size, width of the particle size distribution, crystallinity, stoichiometry, inter-particle connections and mixing or doping with other materials.

Gas-phase processes, although having some drawbacks, have the largest means of control and are therefore the method of choice here. Processes based on pure physical processes are used more often than chemical methods as they allow to obtain the highest purity. The evaporation-condensation method is chosen in this work as synthesis method. The Brownian coagulation inherent to this method makes it impossible to obtain narrow particle size distributions but allows to obtain a mixing of particles of different materials for the synthesis of nanocomposites.

The formation of nanoparticles by the evaporation-condensation technique was analyzed using a moment model describing nucleation, condensation and coagulation. Using the model, it was shown that the temperature in the evaporation furnace and the cooling rate determines the mean particle size and that an aerosoldynamical model allows the prediction of the temperatures needed for nanoparticle formation.

For describing more complicated systems having more degrees of freedom, such as distribution of particle sizes, different chemical compositions, distribution of charges and distribution of number of primary in an aggregate, a newly developed Direct Simulation Monte Carlo method was shown to be suitable. Two examples of practical relevance containing multi-dimensional particle dynamics were given: coagulation with chemical reaction in droplets (microreactors) and the coating of solid particles with nanoparticles.

Aggregation of mixtures of charged nanoparticles is useful for obtaining composites. This was studied by means of the Direct Simulation Monte Carlo method. The selectivity of mixing is characterized by means of the fraction of symmetric aggregates. It was shown that for each charge distribution different conditions lead to a highest possible selectivity of mixing.

Two chemical compounds were selected for further experimental study based on a literature review. One component is selected for its potential quantum confinement effects. PbS is a narrow-gap IV-VI semiconductor with a cubic rock salt structure and is an attractive candidate for the study of quantum confinement effects as its hole Bohr radius is 9 nm. This results in strong confinement effects. The confinement results in a blue shift in the optical absorption spectrum. The other material, SnO_x, is selected for its gas-sensing properties.

A size uniformity is important in order to study or make use of quantum size effects, as a distribution of particle sizes will decrease or smear out these effects. In view of the high specific surface and activity of SnO_x nanoparticles, considerable improvement in sensing properties is expected. In order to understand the relationship between gas-sensing behaviour and particle size better it is necessary to generate monodisperse nanoparticles, because a distribution of particle sizes will decrease the special properties and complicates the interpretation of the experimental results. At the moment no gas sensors based on almost equal-sized nanoparticles are available. For this goal, size fractionation by means of a Differential Mobility Analyzer was applied. This decreases the yield, but for functional applications the quality more than the quantity of the nanoparticles is decisive, as most of the functional applications need only thin films.

Several new instruments have been developed in this work which allow to obtain more control over processes which are essential for the synthesis method chosen here. They allow nanoparticle charging, nanoparticle size fractionation and nanoparticle deposition.

A newly developed nanoparticle charger, the twin Hewitt charger, allows a larger fraction of the nanoparticles to be charged, which is important to get a higher yield. A higher yield would result in a shorter deposition time, which is important because the deposition takes at present hours to days. The instrument has been extensively investigated experimentally in order to find the optimal charging conditions.

Differential Mobility Analysis at lower pressures than atmospheric pressure has been investigated theoretically. It was found that the lower particle limit is not changed fundamentally with lower pressures when keeping the mass flow rate constant, but that the higher particle limit is decreased due to decrease of the maximum allowed field strength. A Differential Mobility Analyzer design for low pressure has been presented. It is important to perform size fractionation at pressures lower than atmospheric pressure, as usually the system pressure is decreased in order to get a higher yield of smaller nanoparticles and to increase the cleanness of the system.

An Electrostatic Precipitator has been developed which allows the investigation of electrical properties of nanoparticle films under clean conditions direct after deposition. This is especially important for semiconductor nanoparticles, where the surface influences the optical and electronic properties.

Synthesis of the semiconductors PbS and SnO_x in form of almost equal-sized monocrystalline nanoparticles is showed to be possible with the help of carefully controlled techniques such as size fractionation and in-flight sintering. The processes of sintering of PbS has been investigated with help of a quantitative model which allowed the determination of sintering parameters, which are usually unknown for semiconducting materials. The process should be controlled such that sufficient sintering and crystallisation but no evaporation and re-condensation occurs, as this destroys the monodispersity.

As examples of the suitability of synthesized PbS and SnO_x nanoparticles for functional applications, the absorption spectrum of PbS and the gas-sensitivity of thin films composed of SnO_x nanoparticles were investigated. As a first indication of changing physical properties with particle size, the absorption spectrum of PbS particles is shown to shift toward the blue, indicative for quantum confinement effects. The measurements also indicate that the optical properties are strongly influenced by the handling of the nanoparticles directly after their formation, such as sintering and/or evaporation processes taking place in the sintering furnace. By choosing suitable initial mobility diameters, it should be possible to attain the commercially interesting bandgap range around 1.3 eV, used for very fast optical data transfer by means of GaAs solid state lasers. Further investigations should be accompanied by careful observations of the actual particle diameter and crystallinity after sintering at different temperatures, preferably by means of high-resolution transmission microscopy.

First measurements show that it is possible to create thin gas-sensitive layers composed of equal-sized SnO_2 nanoparticles. The experimental procedure allows to produce thin films, in which the properties of the nanoparticles can be independently controlled by size-fractionation and in-flight sintering and oxidation. Further properties of the films, such as the inter-particle contacts which are very important for the electrical conductivity, can be influenced by post-deposition annealing This opens a way to investigate fundamental gas-sensing properties of a better controlled microstructure than available at present.

Finally, the possibility of gas-phase synthesis of tailored composite nanoparticles by means of aggregation of oppositely charged, size-selected nanoparticles of two different materials is shown. Electrical effects are applied for size-selection, selective mixing, separating the composite nanoparticles from unaggregated nanoparticles and the deposition of nanoaggregates. Size, size dispersion and materials can be selected independently. As an illustration of the method, composite nanoparticles consisting of differently sized Ag and PbS nanoparticles have been obtained. The method allows the investigation of fundamental properties of composite nanoparticles both in-flight and after deposition in form of a thin film.

Concluding, the field of nanoparticles for functional applications is exciting and rapidly developing. Synthesis and handling methods are available, but the main challenge lies in obtaining a better control over the particle characteristics so that the desired properties of the functional applications can be attained.

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