Stochastic modeling of multidimensional particle characteristics with parametric copulas for the analysis of microstructural effects during separation processes of particle systems

SPP 2045 – Zentralprojekt Z2

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Objectives and work packages

The goal of the priority program "MehrDimPart" (SPP 2045) is to develop new separation processes for systems of particles with sizes below 10 µm. For that purpose the central project Z2 deals with the characterization of particle systems using multivariate distributions of particle descriptors (Objective 1), which we typically determine from tomographic image data, see Figures 1 (left) and 2 (bottom row).

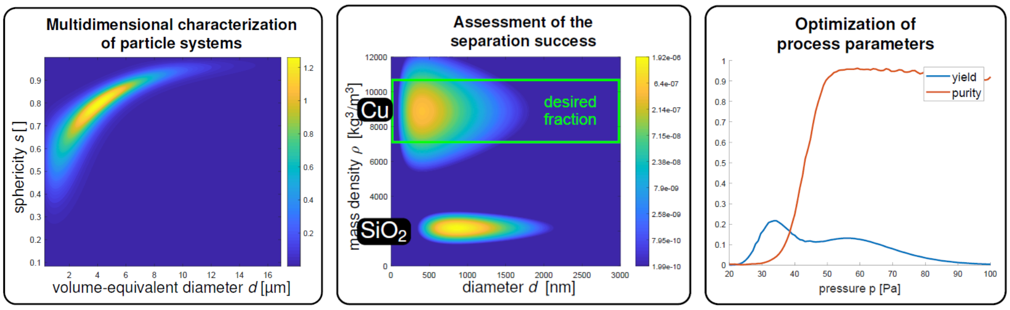

Figure 1: **Objectives of the central project Z2.** Left: Multidimensional characterization of particle systems. For example, using the bivariate distribution of the volume-equivalent diameter and the sphericity of a particle system. Middle: Assessment of the separation success. In the depicted example the bivariate probability distribution of a Cu/SiO_2 particle mixture is shown. By comparing such distributions before and after separation, we can compute quantities for assessing the separation success of the desired fraction. Right: Optimization of process parameters. If the dependency between process parameters and separation behaviour is known, process parameters can be optimized with respect to quantities which assess the separation success (e.g., purity and yield) [6].

In the first funding period of the SPP 2045 the central project Z2 developed segmentation procedures for reliably extracting individual particles from tomographic image data, followed by the stochastic modeling of their particle descriptors using multivariate probability distributions [1, 2, 4, 5]. The characterization with probability distributions reduces the complexity of large data sets to a few model parameters. In the second funding period of the SPP 2045 the developed methods will be applied to tomographic image data of particle systems measured before and after separation, which will enable the assessment of the separation success (Objective 2), see Figure 1 (middle). Furthermore, by comparing multivariate distributions of particle descriptors of the feed material and the product, we will analyse microstructural effects on the quality of separation processes. For example, we will compute multivariate partition functions which are generalizations of the commonly investigated univariate partition/Tromp curves. Once the influence of process parameters on the separation success is quantified, e.g., by mapping process parameters on the separation process' partition function, we can compute optimal process parameters (Objective 3), see Figure 1 (right) [6].

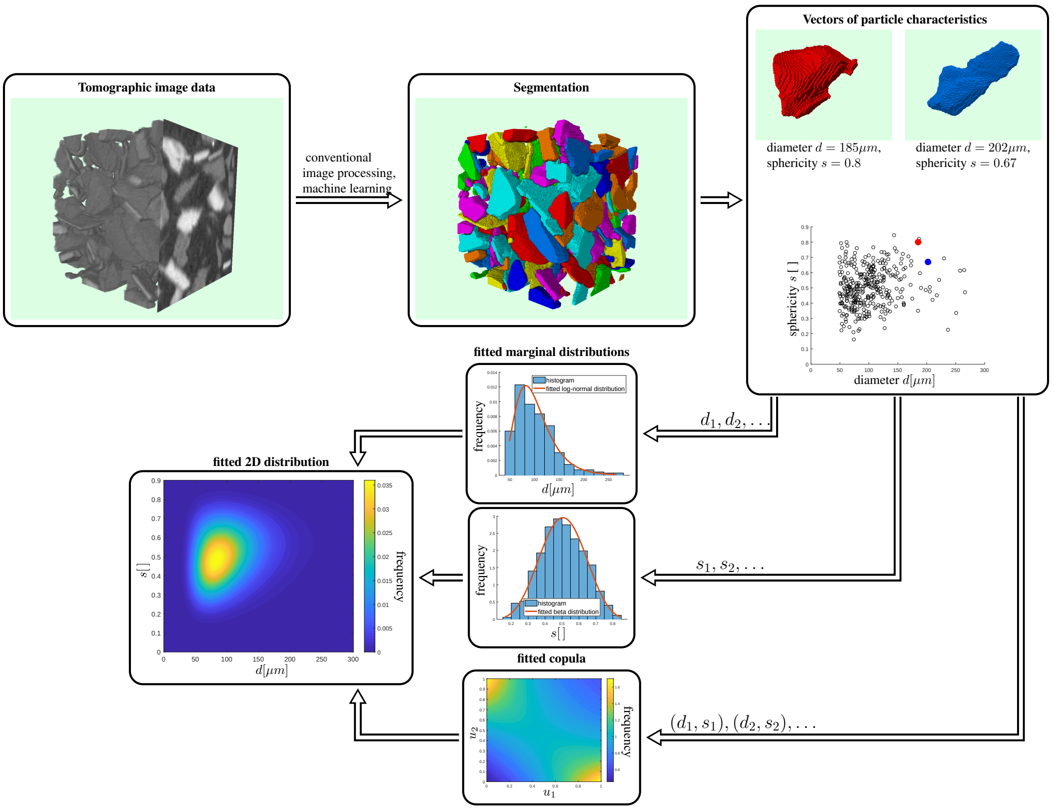

Figure 2: **Workflow for modeling the multivariate distribution of particle descriptors.** First, tomographic image data is segmented using image processing and methods from machine learning. Then, individual particles and their vectors of particle descriptors are extracted. For modeling the multivariate distribution of vectors of particle descriptors, we first fit the marginal distributions. Then, by fitting a so-called copula, the multivariate distribution is obtained [1, 4, 5].

The work packages of the central project Z2 are structured as follows:

- **Analysis of methods for quantitative evaluation of the separation success**
  This includes the characterization (Objective 1) of particle systems and the evaluation of results (Objective 2) achieved by separation experiments performed by project partners within SPP2045.

- **Computation of multivariate distributions of particle descriptors from 1D measurements**
  This enables us to leverage univariate measurement techniques, which are available in many laboratories, for deriving multivariate distributions of particle descriptors (Objective 1).

- **Data-driven calibration of stochastic 3D particle models**
  Using segmented tomographic image data, we will derive stochastic 3D particle models which can generate virtual but realistic particles (Objective 1).

- **Generation of virtual particle systems for the analysis of separation processes**
  The particle systems generated by the stochastic particle models will be used for numerical simulations performed by project partners, for investigating the influence of microstructural effects on the separation results (Objectives 2 and 3).

- **Stereological calibration of 3D particle models using 2D data**
  The particle systems generated by the stochastic particle models will be used for calibrating a stereological predictor which can characterize properties of a particle system from 2D image data, like for example, scanning electron microscopy data (Objective 1).

Methods

For the analysis of a system of particles from tomographic image data a particle-wise segmentation is necessary, see Figure 2 (top row). To achieve this, we deploy both conventional image processing algorithms (e.g., watershed transform) and methods from machine learning (e.g., convolutional neural networks such as the U-net), such that it is possible to extract each particle for further analysis [2, 5]. Then, the segmented particle system can be efficiently characterized, e.g., using multivariate probability distributions (see Figure 2, bottom row, and Figure 3) as well as stochastic 3D particle models, see Figure 4. In particular, the following characterization and modeling techniques are considered.

Multivariate characterization of size-shape descriptors. Due to the segmentation of tomographic image data it is possible to determine a vector of descriptors like volume, surface area, sphericity, convexity and elongation for each particle, which describe its size and shape. By doing so, for the entirety of the particle system one receives a large sample of such vectors which makes it possible to determine a multidimensional probability distribution of descriptors for the system of particles. Parametric copula distributions are a viable option for modeling such distributions, since they can incorporate correlations and dependencies between descriptors [4]. In Figure 2 (bottom row), a copula with five parameters is used two describe the multidimensional distribution of such vectors of descriptors.

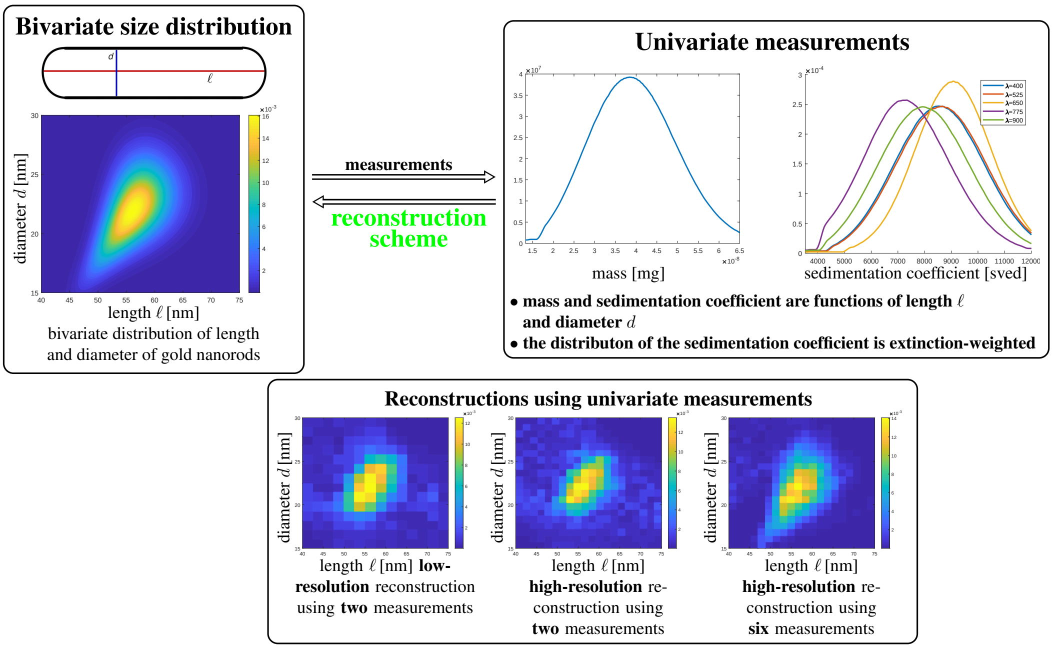

Figure 3: **Reconstruction of bivariate probability densities from univariate measurements.** Top: Bivariate probability density of length and diameter distribution of gold nanorods with hemispherical endcaps (left). Univariate probability densities of mass and sedimentation coefficient can be measured by an aerosol particle mass analyzer and by multiwavelength analytical ultracentrifugation, respectively (right). Bottom: Reconstructions of the bivariate probability density using univariate measurements [3].

Alternatively, if no tomographic image data is available, multivariate probability densities of particle descriptors can still be reconstructed from measured univariate probability densities. For example,

Figure 3 depicts the reconstruction results for the bivariate probability density of length and diameter of gold nanorods from univariate distributions of their mass and sedimentation coefficient [3].

Stochastic 3D particle models. Instead of modeling the distribution of descriptors which are aggregated quantities for the characterization of particles, we can perform a more holistic characterization by fitting stochastic geometry models to particles observed in tomographic image data [7]. More precisely, from a fitted stochastic 3D particle model, we can generate arbitrarily many virtual particles which are statistically similar to the corresponding particle system to which it was calibrated to, see Figure 4. Since the developed methods are scale-invariant, they can be applied on tomographic 3D image data of a wide variety of particle systems. There is also a wide range of stochastic geometry models for the generation of virtual particles [7]. By utilizing the spherical harmonics representation of particle surfaces, stochastic geometry models can be defined that can generate star-shaped (a generalization of convexity) particles, see Figure 4 (bottom row). For the generation of faceted particles or the inner grain architecture of polycrystalline particles, random mosaics (tessellations) are viable stochastic geometry models, see Figure 4 (top row).

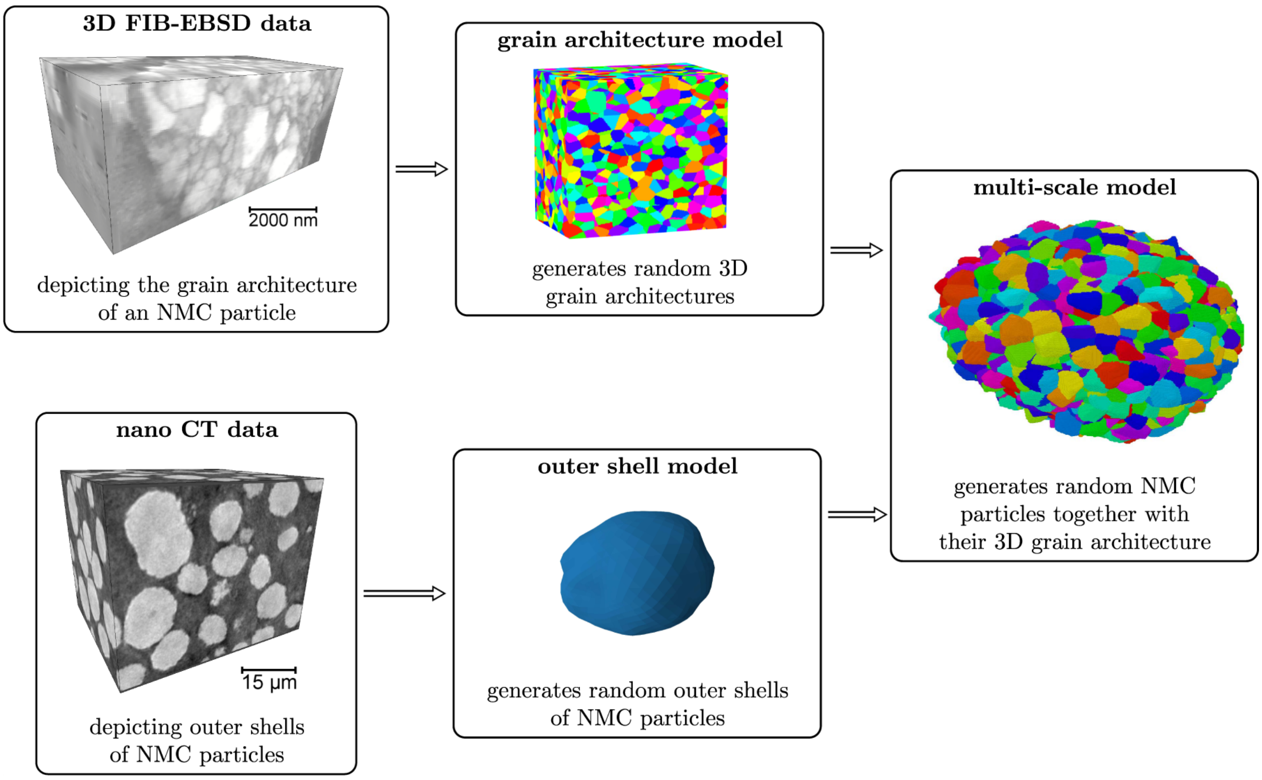

Figure 4: **Modeling the outer shape and grain architecture of cathode particles.** The grain architecture of a particle was imaged by combining a focused ion beam with electron backscatter diffraction. A stochastic grain architecture model was fitted using random Laguerre tessellations. Nano-computed tomography was used to calibrate a stochastic model for the outer shell of particles. By combining both the grain architecture and the outer shell model we obtain a multi-scale model from which numerous particles can be generated [7].

Generating virtual particle systems. Since parametric models are considered, such a stochastic 3D particle model is described by just a few parameters which in turn fully characterize the corresponding measured particle system. Moreover, by systematic variation of model parameters further virtual particle systems can be generated which deviate statistically from measurements. These particles will be used for numerical simulations performed by project partners, for investigating the influence of microstructural effects on the separation results. Moreover, a stereological predictor will be calibrated to the generated particles such that the parameters of the corresponding stochastic geometry model can be determined from 2D images, like for example, scanning electon microscopy data. Thus, a stereological predictor serves as yet another tool for an in-depth characterization of particle systems.

References

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