

# Stable Particle Films for Catalysis and Gas Sensing

## Mechanical Properties of Aggregated Nanoparticle Films

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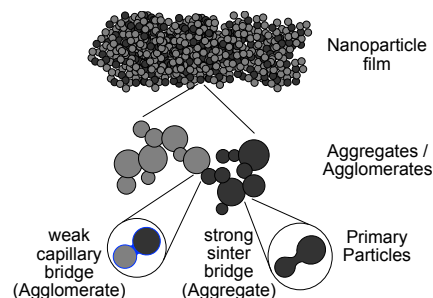
### In Short

- Nanoparticle Film Compaction
- Aggregated Nanoparticles
- Elastic Film Deformation

Aggregated nanoparticles films show promising results in terms of efficiency, effectiveness, sensitivity and selectivity in many applications, including catalysis and gas sensors. In these fields of application it is crucial to provide specific structural properties of the used particle films, such as specific surface area, porosity, pore size distribution and percolation threshold. Fluid molecules have to enter the film through pores to adsorb at the surface. Therefore, the pore structure has to allow molecules to first enter the film through large pores and subsequently diffuse through smaller pores into all regions of the film to reach the surface of the particles. Furthermore, in the case of gas sensors the reaction of molecules on the particle surface leads to free electrons that have to travel to the electrodes attached to the particle film. Hence, percolating (conducting) paths will increase the sensitivity and response time of the sensors.

Nanoparticle films synthesized by flame spray pyrolysis are capable of providing these requirements in a one step deposition process. However, the resulting particle films often suffer from low stability against mechanical stress. It has been shown that low-pressure compaction using a two-roll laminator can significantly improve the mechanical resistance of the layers [1]. With increasing lamination pressure (in the range from 0.75 to 2.50 MPa) the Young's Modulus measured by AFM-Colloidal Probe measurements increased. Thorough characterization has shown that the increased mechanical stability results from restructuring events within the particle film. This can be seen in a decrease in porosity and a shift in the pore size distribution to smaller pores. However, the restructuring in dependency of particle properties such as primary particle size distribution and aggregate size distribution remains unclear and is very difficult to evaluate experimentally.

The modelling of such aggregated nanoparticle films requires knowledge about the components of the films, such as primary particles that form aggregates *via* sinter bridges and aggregates that are

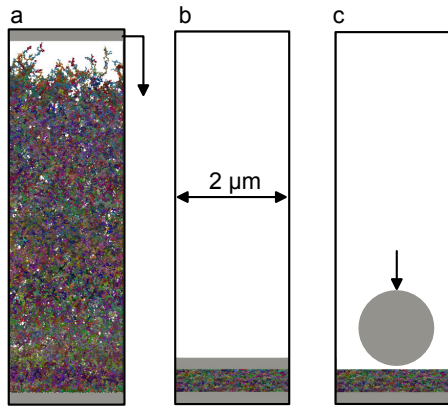


**Figure 1:** The building blocks of nanoparticle films. Primary particles are connected via strong sinter bridges (aggregates) and weak capillary bridges. This results in a highly complex network.

bond with other aggregates *via* capillary bridges (agglomerates, 1). From experiments [2] and atomistic simulations [3] we could learn a lot about the capillary bridges. In a humid environment the primary particles are covered with a water layer. Compared to the size of the primary particles (approximately 10 nm in diameter) the thickness of the water layer cannot be neglected. On this small scale it is necessary to consider the molecular structure of water, which determines interactions even at low humidity.

The experimental characterization of the sinter bridges within aggregates however, is highly challenging. We hypothesize that the tensile/shear strength and Young's modulus of these sinter bridges are responsible for restructuring and mechanical resistance during compaction. They determine the porosity with respect to applied compaction pressure, the pore size distribution and the percolation threshold. Hence, it is an important parameter that has to be understood in order to design mechanically stable nanoparticle films.

In this project we aim to investigate the influence of aggregate sinter bridges and their material parameters (Young's Modulus, tensile/shear strength) on the compaction of nanoparticle films. To achieve this, we apply discrete element method (DEM) simulations on aggregated nanoparticle films. DEM simulations consider primary particles as spherical particles that can interact with other particles by normal force, rolling and sliding friction. In previous projects we have extended the open source DEM software LIGGGHTS [4] by a long-range coarse-grained potential that is capable of representing the capillary forces between aggregates [6]. Furthermore, it is possible to implement elastic sinter bridges characterized by a tensile/shear strength and a Young's modulus to model sinter bridges between particles



**Figure 2:** The setup of the simulations. (a),(b) The nanoparticle film (600,000 particles) is compacted by the upper wall until the targeted pressure is reached. (c) Subsequently, the top wall is replaced by a colloid, which elastically deforms the film.

within an aggregate.

The DEM is applied on aggregated nanoparticle films as generated by a simple deposition algorithm as proposed by [5]. These films include experimentally obtained polydisperse primary particle and aggregate size distributions and match the porosity of particle films by flame spray pyrolysis and therefore, will serve as initial film configuration (fig 2 (a)). The film compaction will be carried out by the application of two walls as shown in fig 2 (a),(b). The upper walls compacts the layer at a constant velocity, while the pressure exhibited by the particle film on the walls is calculated. This enables the validation of the pore structure and the porosity with respect to the compaction pressure to experimentally obtained. Furthermore, the simulations can give more detailed information on the breaking behavior that cannot be characterized in the experiments.

This approach opens the door to investigate structural properties more detailed than experimentally possible. Therefore, we aim to derive a set of sinter bridge parameters (Young's Modulus and tensile/shear strength) that matches experimentally obtained data [1].

Based on these parameters and the resulting particle films, the elastic deformation of the films will be investigated by means of the Colloidal Probe Technique (CPT) (fig. 2 (c)). The films elastic response on the penetration of the colloid will be simulated, which allows the calculation of a films Young's Modulus. This Young's Modulus is an indicator for the films resistance against mechanical stress and therefore, mechanical stability. The flame spray pyrolysis allows to synthesize aggregated nanoparticle films in a wide range of different primary particle and aggregate size distribution. With the models, methods and knowledge gained from this project, we can relate mechanical and electrical properties including elastic

film deformation and electric conductivity to structural and particle properties, such as particle size distribution, aggregate size distribution, pore size distribution and percolation threshold. This helps to derive the most important structural and particle properties, specifically for each of the wide range of application. From this, we can derive design parameters for the synthesis of mechanically stable aggregated nanoparticle films and help to develop highly sophisticated products, tailored for their specific application.

## WWW

<http://www.uni-bremen.de/>

## More Information

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## Project Partners

Fraunhofer Institut IFAM, Bremen

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