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Comparative analysis of the coating thickness on single particles using X-ray micro-computed tomography and confocal laser-scanning microscopy

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ABSTRACT

The quality of coated products depends strongly on the structure of the coating layer. Confocal laser-scanning microscopy (CLSM) and X-ray micro-computed tomography (μ-CT) are used as non-destructive techniques for the quantification of coating microstructures, especially coating layer thicknesses. Combined with image processing, it is possible to evaluate and correlate chord lengths and coating thicknesses. In this study two different kinds of core particles were coated with crystalline and film-forming coating materials by fluidized bed spray granulation. The thicknesses of the coating layers were investigated using both measuring methods. Motivated by observed differences in the obtained CLSM results for the layer thickness, a new evaluation procedure is developed and discussed.

1. Introduction

Coating of ingredients is used in many applications in chemical, food and pharmaceutical industry. The coating layer adds certain functionality such as taste and odour masking, protection from environmental conditions or the controlled or sustained release of active ingredients to the particle [1]. The functionality that can be achieved depends strongly on the quality of the coating, e.g., layer thickness, coating uniformity or coating porosity. This means, that a precise characterisation or identification of the quality and micro-structure of the coating layer is required to gain a better understanding of the dependency of functionality of coated particles on the layer properties and the process conditions [2].

Techniques for characterisation of coating layers are, among others, X-ray μ-computed tomography (μ-CT) and scanning electron microscopy (SEM) [3]. SEM information is, however, restricted to the sample surface. To obtain information on the inner structure of a particle, e.g., layer thickness, the sample needs to be treated mechanically, for example to be cut, rendering the sample unusable for further analysis. The μ-CT provides volumetric information on the internal structure like coating thickness and micro-porosity, but has a long measurement and preparation time. Farber et al. [4] were among the first who used X-ray tomography for the determination of the porosity and pore size distribution of granules. Perfetti et al. [5] performed further investigations of coating layers. Especially porosities and layer thicknesses have been determined. By principle, μ-CT is not restricted to the investigation of coating layers but can also be applied to reveal the inner morphology of single particles and agglomerates [6]. Hancock et al. [7] and Zeitler et al. [8] provided overviews of dosage forms with special attention to in-vitro characteristics.

It is also reported that confocal laser scanning microscopy (CLSM) can be used for investigation and quantitative characterisation of the morphology of coated particles. CLSM is commonly used in biological and pharmaceutical applications. At the beginning of the 1990s, the CLSM supported a breakthrough in the study of food microstructures [9]. Proteins and lipids could be visualised and changes in food products could be observed dynamically.

Depypere et al. [10], Dewettinck et al. [11] and Ruotsalainen et al. [12] provide an overview on the capabilities of the CLSM to analyse coated particles and microcapsules. It is reported that CLSM is an appropriate technique for the quantification of thin coating layers (<5 μm). Furthermore, defects at the film-core interfaces and on the surfaces of coated particles can be characterised. Lamprecht et al. [13] performed experiments with microcapsules that consisted of an oil phase enclosed by gelatine and Arabic gum. It was reported that CLSM allowed measurements through the wall material and that a quantitative analysis of the structures not only on the surface but also in the interior of the materials is possible. Using suitable fluorescent dyes, the polymer distribution throughout the capsule could be visualised. Furthermore, liquid

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marbles have been observed and studied. Nguyen et al. [14] evaluated the structure of hydrophobic powder shells using CLSM. Not only biological substrates can be investigated with this system, also crystalline solids like marble were investigated with respect to their porosities and voids using special preparations [15].

In 2009, confocal microscopy was presented by Laksmana et al. [16] as a non-destructive method for the determination of coating thickness. The authors analysed the layer thickness distribution ($s \approx 30$ μm) on small single particles, as well as layer porosity and pore size distribution of granules coated with HPMC. Depypere et al. [10] investigated microencapsulated glass particles ($d \approx 200$ μm), which were coated with very thin sodium caseinate and gelatine layers ($s < 5$ μm) also by CLSM. The resulting images were analysed by image processing with regard to the layer thickness distribution on single particles. It is notable that a variety of mainly organic and only a few inorganic substrates were investigated with a CLSM measuring system.

The present investigation aims at contributing to a better understanding of the potential of CLSM for characterisation of the internal structure of coating layers by comparison of this method with high-precision μ-CT measurements. Inorganic, crystalline and organic film-forming coating materials will be used. Furthermore, a novel method will be developed for correlating obtained measurement results and coating layer thickness of particles. All particles investigated in this study are produced in a spray fluidized bed. It will be shown and discussed how CLSM measurements differ from μ-CT measurements, with respect to measuring time, maximum resolution and evaluation procedure and received information. It will also be shown that, contrary to previous publications, under certain conditions CLSM measurements do not yield the coating thickness but only surface projections. Finally, it will be shown how such information can be used to obtain the desired coating thickness.

2. Material and methods

2.1. Particle coating process

All particles investigated in this study were coated in a spray fluidized bed. The coating experiments were carried out batch-wise in a laboratory scale fluid bed granulator with 150 mm inner diameter of the cylindrical fluidization chamber (GLATT GmbH, Weimar, Germany). The coating solutions were produced with demineralised water, which was previously dyed with a fluorescent material to ensure the visualisation of the coating layer in the later CLSM measurement. All process parameters such as airflow rate, spraying rate and gas inlet temperature were kept constant during the experiments. The coating solution was applied in a top-spray configuration using a two-fluid nozzle (Schlick GmbH). The atomisation pressure was set to 1 bar. Depending on the process conditions the coating solution forms either a compact or porous layer on the particle. The general idea of this process is shown in Fig. 1.

Table 1 summarises the material properties of core particles and coating solutions of all performed experiments. Porous γ-alumina beads with mean radius of 900.0 μm and 315.0 μm were used as core particles in experiments A and B, respectively, having a very high sphericity (>0.9). Non-porous glass particles were used as core particles in experiments C and D. The mean radiuses were 305.0 μm and 137.5 μm, with sphericities of more than 0.9. The coating material used in experiment A was sodium benzoate (NaB). This salt has an antibacterial effect and is used as a preservative in the food industry. It possesses good solubility in water, so that a mass fraction of approximately 30% can be achieved under room conditions (20 °C). The sodium benzoate solution was produced with demineralised water. The coating solutions of the experiments B and C consisted of Shellac (SSB 57 Luna FL, BASF, Germany), which was dissolved in water and 2 Ma-% ammonia to increase the solubility. Shellac is a natural, film-forming material extracted from plant louses (*Tachardia lacca*). This material is widely used in food industry and to some extent in pharmaceuticals, health supplements and nutraceuticals. Further applications of Shellac are enteric coating and retard coating for food [17]. In experiment D the sodium benzoate solution was mixed with the polymer hydroxyl–propyl–methyl–cellulose (HPMC, trade name Pharmacoat 606, from Shin-Etsu, Japan). HPMC is used as a binder in pharmaceutical and food industry. In combination with NaB solution, the binder reduces the porosity in the coating layer significantly [18].

To all coating solutions the fluorescent dye uranine (TIFOO, Germany) was added. This dye is a common food colourant and not harmful to health in small amounts. It is used to visualise the coating layer with CLSM as the measuring method. Non-coloured and non-auto fluorescent constituents, e.g., the core particles, will be invisible to CLSM laser light and appear black.

The process parameters of all experiments are also given in Table 1. The differences in the gas inlet temperatures, spraying rates as well as spraying times where chosen deliberately to create different layer morphologies, based on previous preliminary experiments. The reduced fluidization flow rate for experiment D is due to the smaller mean diameter of the core particles.

2.2. X-ray μ-computed tomography

In this study, randomly picked particles from each experiment were scanned utilising a customised X-ray μ-computed tomographic device (ProCon X-Ray GmbH, Germany). The μ-CT gives volumetric information about the internal structure of a scanned object. Contrast within the overall image depends on differences in both, the density of structures in the sample and the thickness of those structures. The greater the difference in density or thickness of two adjacent structures, the

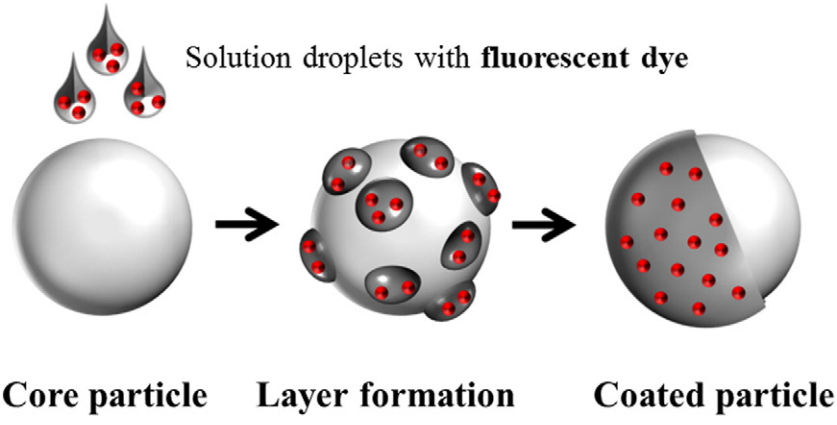

Fig. 1. General coating scheme [20].