Dry starch particle coating:optimization and coating quality measurement by CLSM

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Introduction

In the food industry, the coating technology is frequently used to separate and protect ingredients from their environment (water, acid, oxygen, other food ingredients). It could be used to stabilize ingredients during processing (heat, pressure, moisture), to impart controlled release (during processing, storage or consumption), to change physical characteristics of the original material by reducing the hygroscopicity, improve flowability, reducing dustiness or modifying density (Ivanova et al.2005, Arshady 1993).

Coating with polymer powders has been studied as an innovative technology and an alternative to the traditional coating with an aqueous or organic polymer dispersion, which comprise, respectively, a time and energy consuming process and a solvent recovery step. Compared to solvent and water based coating the dry coating method is favorable regarding environmental friendliness, safety and cost.

Dry powder coating consists of coating large particles (host) with fine powder (guest). The adhesion of these guest particles is accomplished using a mechano-chemical treatment or plasticizer. A plasticizer generally produces a decrease in glass transition temperature (Tg) of the polymer and promote coalescences between guest particles in order to obtain a continuous film (Wheatley 1997).

The plasticizer method is particularly recommended when particles soft, sensitive to heat and to deformation by severe mechanical forces, need to be coated (Obara et al. 1999, Naito 1993).
The present contribution wishes to provide preliminary results to apply dry powder coating in the food and pharmaceutical field. Specifically, the aim of this study is to obtain host particles coated with polysaccharides by the dry powder coating process in a pan coating reactor.

Materials & methods

A “Pan Coater” device (prototype designed and manufactured in our laboratory), 40cm in diameter and 25cm in width, was used. An air compressed spray nozzle, operating at 0,3 bars, was fixed at 20cm from the coater surface. Microcrystalline cellulose pellets (CELLETS®1000-1400µm, IPC Process-Center GmbH & Co. KG, Dresden, Germany) were used as host particles. They were coated with the following guest powder particles: starch octenyle succinate (E1451), tapioca dextrin (National Starch, Düsseldorf, Germany), malto-dextrin, aluminium starch octenyle succinate and octenyl succinate dextrin of waxy maize (Roquette, Lestrem, France). The following plasticizers were tested: triethyl citrate (TEC, Merck KGaA, Darmstadt, Germany), glycerol (Acros Organics, Geel, Belgium).

Experimental procedure: 400g of cellulose pellets were loaded in a pan coater reactor. The rotational speed of the reactor was fixed at 35 rpm. The plasticizer is charged in the reactor chamber

by an air compressed atomization nozzle at rate of 2 ml/min and the polymer powder is charged at a rate of 5 g/min. The total process time is 5min per each coating level.

Polymer analysis: The particle size of the polymer powders was determined by laser light diffraction (Mastersizer, Malvern, United Kingdom).

Coating variables: The coating efficiency and coating level were calculated by the following relations. In these, M_{cP} and M_{uP} are respectively the weight of coated and uncoated pellets, M_{Poly} is the amount of coating polymer and M_{Plas} is the amount of plasticizer.

$$\textbf{Coating process Efficiency (\%)} = \frac{M_{cP} - M_{uP}}{M_{Poly} + M_{Plas}} \qquad \textbf{Coating Level (\%)} = \frac{M_{cP} - M_{uP}}{M_{uP}}$$

Scanning electron microscopy: The coated particles were observed by a scanning electron microscope (SEM) (JSM-6400M, Jeol, Japan).

Confocal laser scanning microscopy: A Bio-Rad Radiance 2000 Confocal Laser Scanning Microscope (Bio-Rad, UK), combined with a Nikon Eclipse TE300 inverted fluorescence microscope (Bio-Rad, UK), were used in order to check the quality of the coating.
Images of 100 coated beads were recorded under the CLSM. These images were then analyzed using the ImageJ software (National Institutes of Health, USA).

Results and Discussion

Coating process efficiency results for aluminium starch octenyle succinate, tapioca Dextrin, and octenyl succinate dextrin of waxy maize were found to be between 85 and 95%, as reported in Fig.1A .

Low values of coating process efficiency and presence of aggregates of guest particles have been noticed in experiments conducted with malto-Dextrin and starch octenyle succinate. The particles of the latter mentioned materials are bigger than 50µm.
The capillary forces play an important role in this coating process and probably they are not strong enough to permit the adhesion of guest particles on host surface when their size exceeds 50µm.

Contrarily, a coating process done with guest particles smaller than 50µm have shown high values of coating process efficiency (Fig.1B) and uniform thickness onto the host surface (Fig.2).

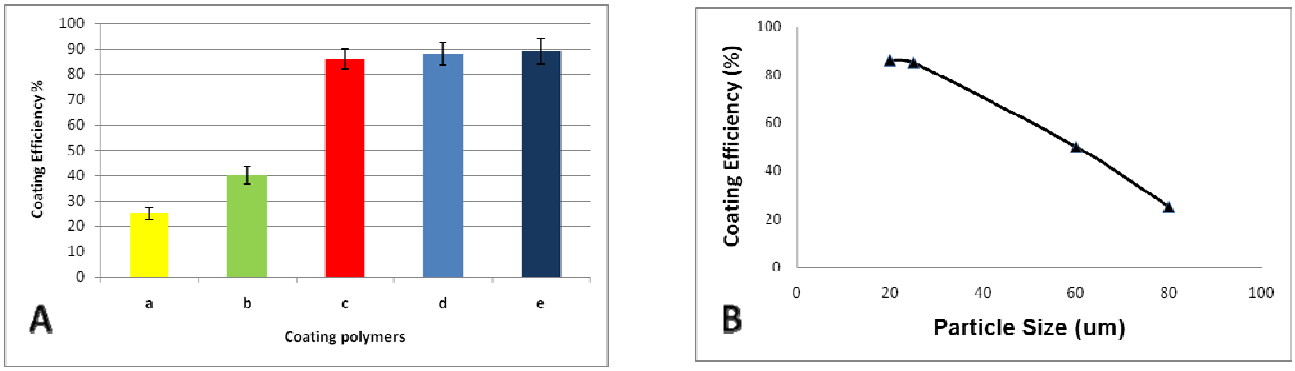

Figure 1: Coating efficiency (A) and relationship between coating efficiency and powder particle size (B) of malto-dextrin (a), starch octenyle succinate (b), tapioca dextrin (c), aluminium starch octenyle succinate (d) and octenyl succinate dextrin of waxy maize (e).

Triethyl citrate and glycerol have been selected as plasticizer/binder due to their high viscosity and good starch plasticization properties (Bonacucina et al.2006).
For both plasticizers high values of coating process efficiency have been recorded. In Fig.2A and Fig.2C differences in homogeneity of the coating shell formed by triethyl citrate and glycerol are shown.

Furthermore, as shown in Fig.2B, no occurrence of guest particles' coalescence onto the pellets' surface have been found when triethyl citrate is used as a plasticizer
In this case, the triethyl citrate behaves like a binder during the process and not as a plasticizer in its literal meaning. On the contrary, using glycerol as plasticizer, the onset of coalescence between guest particles, as shown in Fig.2D, could be noticed.

A scale-up of the dry powder coating process has been done in a pilot drum coater (LAB15, NICOMAC). The coating process efficiency was 90%, which suggests the feasibility of the process at an industrial level.

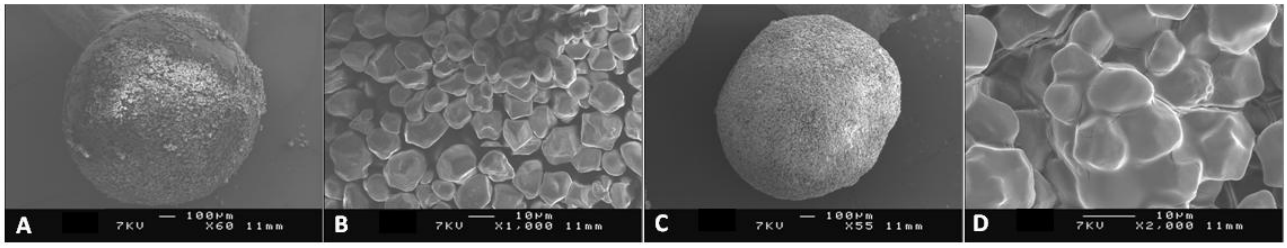

Figure 2: SEM image analysis of microcrystalline cellulose particles coated with octenyl succinate dextrin of waxy maize with triethyl citrate (A,B) and with glycerol (C,D) as plasticizer.

The application of Confocal Laser Scanning Microscopy (CLSM) in dry powder coating technology has been studied. Rhodamine, as a chromophore compound, was mixed with the liquid plasticizer, in order to achieve 1mg of chromophore on 4g of host particles.

The novelty about CLSM employment in the dry coating technique results from on the one hand the possibility to detect the plasticizer matrix inside the coating layer, and from on the other hand the possibility to measure the coating thickness and quality after image analysis.

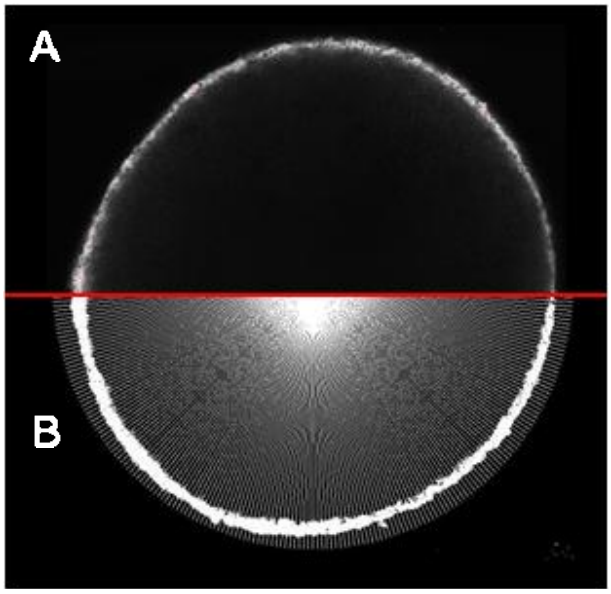

Figure 3: CLSM image (panel A) and processed image of coated pellet (panel B)

The dry coated particles have been processed by an image analysis protocol (Depypere 2005). In order to obtain an entire particle image, the protocol has been slightly adapted. This was needed because the coated particles were too large to fit in the field of microscope (Fig.3).
Coating thickness average, d, and heterogeneity, σ (defined by standard deviation of d) have been done on 100 coated particles. Afterwards the coating quality, defined as $(d-\sigma)/d$, has been determined (Table 1).

Coating level increase from 6 to 18% increase *d* from 19 to 25μm. As coating efficiency remain constant, it has been assumed that coating level increase more than coating thickness, due to high degrees of particles packaging. This may be confirmed by a coating quality increase.

Coating Level	d	σ	Coating quality
6%	19 μm	10 μm	45%
12%	22 μm	11 μm	53%
18%	25 μm	10 μm	60%

Table 1: Results of CLSM image analysis.

CONCLUSIONS

The results have shown the feasibility of a dry coating process in the "pan coater" device both in lab-scale and in pilot-scale conditions. The feasibility of modified starches to be used in a dry powder coating process with small losses of coating material, was shown.

Polysaccharides host particles, with size under 50μm, have resulted in a good uniform coating. This coating process is a highly efficient process (a short processing time of 25min causing low energy consumption) compared to a standard coating process.
The use of Confocal Laser Scanning Microscopy as a new characterization technique for dry powder coated particles has been demonstrated.

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