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Characterisation of the work of adhesion of food grade coating materials on a maltodextrin model surface

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ABSTRACT

The aim of the present study was to evaluate two models, the Owens–Wendt–Rabel and Kaelble model (OWRK) and the Young-Dupré model (YD), for characterisation of the wetting and adhesion properties of cellulose- and shellac-based coating solutions on a maltodextrin model surface (MDGSS). In addition, for the OWRK model, the impact of a variation in the combination of reference liquids on the free surface energy (SE), the contact angle (CA) and the work of adhesion (W_A) was investigated.

Results of the present study demonstrate that the use of reference liquids covering a wide range of polar and disperse fraction of the SFT is recommended for the OWRK model. It was shown that at least the use of water and diiodomethane is needed for reliable values of the SE and the W_A. Additional reference liquids, e.g., ethylene glycol or formamide, are recommended to obtain robust results. With respect to the coating solutions, the calculated contact angle according to the OWRK model did not provide qualified values due to an inappropriate combination of polar and disperse fractions of the SFT and the SE of MDGSS for methylcellulose, Nutrateric® and shellac. Results for the W_{A-OWRK} and the W_{A-YD} provided reliable data, but differed for all coating solutions. Therefore it is recommended to perform both analyses in comparative studies on particle-liquid-interactions.

1. Introduction

Powders have a high importance in the food industry. A wide range of ingredients is processed in powderous form during the production process of the final food product. Among these ingredients are functional food ingredients added for nutritional purposes, which are often susceptible against adverse environmental conditions. In this context a coating can provide an efficient barrier e.g., against moisture sorption or oxygen permeation. A coating can also protect against undesired interactions between a functional ingredient and the food matrix itself. Therefore the coating protects against physiochemical stress during storage and facilitates the incorporation of powderous food ingredients (Ahn et al., 2008; Kuang et al., 2010; Milanovic et al., 2010; Mokarram et al., 2009; Dewettinck and Huyghebaert, 1999; Kayumba et al., 2007; Bifani et al., 2007; Ramírez et al., 2012; Mishra et al., 2010). In addition, there is a wide spectrum of functional coatings with different properties such as targeted release or taste masking. The functionality of targeted release of food grade coatings has already been investigated (Ravi et al., 2008; McClements and Li, 2010; Wei et al., 2008) and reviewed (Shukla and Tiwari, 2012; Lesmes and McClements, 2009).

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Nomenclature

CA_m	directly measured contact angle [°]
CMC	carboxy methylcellulose
CA_{OWRK}	contact angle calculated according to OWRK [°]
D	diiodomethane as a reference liquid
E	ethylene glycol as a reference liquid
F	formamide as a reference liquid
HPMC	Hydroxypropyl methylcellulose
MC	methylcellulose
MDGSS	maltodextrin model surface
N	Nutrateric®
OWRK	model according to Owens, Wendt, Rabel and Kaelble
SH	shellac
W	distilled water as a reference liquid
W_A	work of adhesion [mN/m]
W_{A-OWRK}	work of adhesion according to the model of OWRK ([mN/m])
W_{A-YD}	work of adhesion according to the model of Young and Dupré ([mN/m])
YD	model according to Young and Dupré
γ_S, SE	surface energy of a surface [mN/m]
γ_{LV}, SFT	surface tension of a liquid [mN/m]
γ_S^p, SE_p	polar fraction of γ_S [mN/m]
γ_S^d, SFT_d	disperse fraction of the γ_{LV} [mN/m]
γ_L^d, SE_d	disperse fraction of γ_{LV} [mN/m]
γ_L^p, SFT_p	polar fraction of γ_{LV} [mN/m]
θ, CA	contact angle [°]

Cellulose films are widely used in recent studies in order to investigate their functionality. Hydroxypropyl methylcellulose is known to form oil resistant but water soluble films (Baldwin et al., 2012), carboxymethylcellulose forms films that improve storage stability due to a reduction of water-vapour permeability (Bifani et al., 2007); (Cheng et al., 2008); (Hussain et al., 2010). Methylcellulose forms similar to hydroxymethylpropylcellulose an oil resistant film that can be used to reduce the uptake of oil in fried convenience products (Garcia, 2004); (Tavera-Quiroz et al., 2012). Shellac is a very common food grade coating for targeted release applications (Farag and Leopold, 2011; Limmatvapirat et al., 2008; Stummer et al., 2010) or for applications, which require improved storage stability, because of its barrier properties against water, gas and fat (Soradech et al., 2012).

With respect to the quality of a coating, the interaction of the powder particle (the solid phase) and the coating solution (the liquid phase) is of utmost importance (Vanderroost et al., 2011). The interaction includes spreading of the coating solution on the surface (wettability) as well as its adhesion on the surface. As reviewed by Kwok and Neumann, the contact angle measurement is a reliable method for investigations on the wettability of surfaces during coating processes (Kwok and Neumann, 1999). Direct contact angle measurements on the particle surface is not possible, because of the small size of about 100 μm–1 mm. Therefore it is important to create a model surface which is appropriate for direct contact angle measurements. Meraz-Torres et al. (2011) e.g., investigated droplet recoiling of water on maltodextrin tablets. Werner et al. (2009) used anhydrous milk fat as model surface, since it represents the surface of spray-dried dairy-type particles. However, most investigations during the last years were performed with food grade coating solutions on model surfaces like glass, polytetrafluoroethylene (PTFE) or plastic films (Farris et al., 2011). For coating applications in the food industry, these surfaces are not appropriate to evaluate the interaction of particles and food grade coating solutions. Reviewing the available literature no studies on the adhesion of coatings on food grade surfaces could be identified.

The aim of the present study was to evaluate two models for the characterisation of the wetting and adhesion properties of cellulose- and shellac-based coating solutions on maltodextrin model surfaces. The model surface (MDGSS) was prepared by drying of a maltodextrin solution on an object plate to ensure that the maltodextrin is in a glassy state similar to its physical state in spray-dried powders. In the first model, according to Owens–Wendt–Rabel and Kaelble (OWRK), the free surface energy (SE) with its polar and disperse fractions of MDGSS was investigated by measuring the contact angle of reference liquids with known surface tension and known polar and disperse fractions. After the characterisation of MDGSS the surface tension (SFT) of different coating solutions with its polar and disperse fractions was evaluated by the pendant drop method. From these data the work of adhesion (W_{A-OWRK}) and the contact angle according to OWRK (CA_{OWRK}) can be calculated. The second model was the Young-Dupré model (YD) for which the contact angle of the coating solution samples has to be measured directly on the maltodextrin model surface (CA_m). For the calculation of the W_{A-YD} the SFT of the coating solutions needed to be examined by pendant drop measurements at the air–water-interface and the CA_m needed to be detected. In addition, the OWRK model was further investigated by a variation in the combination of reference liquids in order to determine the most appropriate combination of reference liquids for the characterisation of MDGSS.

2. Experimental method

2.1. *Materials*

Hydroxypropyl methylcellulose (HPMC), carboxymethylcellulose (CMC), methylcellulose (MC, all Harke FoodTech GmbH, Mühlheim, Germany) Nutrateric® (N, consisting of SURELEASE® ethyl cellulose dispersion and NS ENTERIC® additive; Colorcon Limited, Dartford Kent, United Kingdom) and shellac (SH; Stroever Schellack Bremen, Germany) were used as coating solutions. All coating materials were kindly provided as samples by the companies mentioned above. The coating solutions were prepared with distilled water adjusting the viscosity to four different levels of viscosity: very low (vl; 2.7–2.8 mPa s), low (l; 7.5–9.1 mPa s), medium (m; 24.9–28.1 mPa s) and high (h; 84.5–87.6 mPa s). SH was only prepared in the viscosity levels vl, l and m, because the liquid sample provided by the supplier showed a viscosity in the range of the medium viscosity level. Concentrations of the coating materials in the coating solutions can be derived from Table 1. The viscosity of all coating solutions was determined with a rotational viscometer (Brookfield DV-II; Brookfield GmbH, Lorch, Germany) in combination with an UL-adapter kit. All samples were measured at a temperature of 20 °C ± 1 °C.

Maltodextrin (M3) with a dextrose equivalent (DE) of 18 was a kind gift of Ingredion Germany GmbH, Hamburg, Germany. Maltodextrin model surfaces were prepared in glassy state