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Influence of liquid formulation and impact conditions on the coating of hydrophobic surfaces

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ABSTRACT

This study deals with elementary mechanisms involved in droplet–particle collisions during wet coating operations. The study focuses on rebound and splashing phenomena which may occur during the impact of liquid binder droplets on hydrophobic (non-wetting) surfaces. Generally, these result in a decrease in coating efficiency. Impacts of single droplets on hydrophobic surfaces were investigated using a high speed camera and image analysis. Aqueous solutions of a commonly used polymeric binder (CarboxyMethylCellulose sodium salt, CMC) with varying concentrations were prepared and droplet impacts on hydrophobic surfaces analyzed. A drop-on-demand technology was used to generate micron sized droplets with different velocities. Results showed that use of CMC drags along considerable changes in droplet behavior after impact. CMC increased viscoelasticity of the liquid binder which caused a substantial inhibition of splashing and rebound phenomena. Increasing the solution concentration decreased the maximum extent of spreading, recoil velocity, as well as the maximum height of rebound. The impact velocity was found to promote both spreading and receding of droplets. The effect of droplet size was also investigated and discussed through comparison between millimeter and sub-millimeter sized droplets. Plotting the maximum spreading diameter against the Weber number for different fluid viscosities showed the influence of inertia and viscous dissipation on the maximum spreading extent. Finally, an impact regime diagram based on *Oh* and *We* numbers was established based on collected data.

1. Introduction

The powder coating process has received considerable attention in many fields such as the pharmaceutical, food, agrochemical, dyestuffs, minerals and fertilizer industries [1–3]. Two types of equipment can be used to perform the coating operation: systems using mechanical agitation such as drums, pans and impeller mixers and those that use pneumatic solid mixing like the fluidized-bed, spouted-bed or Wurster apparatus [4–11]. In the case of wet coating process, a coating agent is dissolved or dispersed in a solvent and sprayed onto the powder. After collision with particles, the sprayed droplets spread on the particle surface. The subsequent drying leads to solvent evaporation and deposition of a solute layer on the particles [3,9,12–16]. Wetting is among the most important steps during the coating process. Efficient coating operation requires successful droplet deposition on particles, sufficient spreading of the coating liquid on their surface and sufficient coverage of the particles by droplets [9,14,17]. This makes the coating of hydrophobic surfaces (on which the contact angle of a water droplet is higher than 90°) a challenging task because of low liquid spreading and adhesion [18]. Other challenges are due to *splashing* of liquid droplets over the hydrophobic surfaces, or, to droplet *rebound* phenomenon which can particularly affect the quality of film deposition [9,13,19–22]. As an example, Laksmana's study [9] showed droplet rebound to have a significant effect on coating porosity, which is the presence of voids within the coating structure. In addition, several works highlighted the important effect of surface wettability [2,22–24], adhesion strength and fluid formulation on coating efficiency and quality [12,13,25,26]. Examples of hydrophobic cores used in food processing industry are nuts and peanuts. Some workers showed that adding surfactants was necessary to improve the coating efficiency of these hydrophobic cores by a whey protein oxygen-barrier layer [11,22,27]. In addition, many tablets have a hydrophobic surface because of the active pharmaceutical ingredient, or the excipients incorporated inside the tablet. Examples of these hydrophobic components are: lipophilic excipients such as waxes and oils, hydrophobic polymers such as polyamide 12, polyvinyl acetate and polyester resins and other excipients such as stearic acid, fumed silica, talc, magnesium stearate, and tributyl citrate [10,28–32]. This shows that extensive research is required for a better understanding of elementary mechanisms involved in "droplet–particle" collisions [33,34]. The study of droplet impact phenomena is therefore of undeniable importance, especially in the case of non-wetting surfaces. These impact phenomena have received much attention since the first work by Worthington [35]. Most of the reported studies have focused on the impact of single liquid droplets on dry plane surfaces [18,20,32,

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36–42]. However, some studies investigated droplet impact onto the surface of a static powder bed [43–46] and reported the role of surface wettability in drop impact mechanisms.

Literature data concerning droplet impact onto a plane hydrophobic surface highlights the important effect of inertia on the different droplet impact outcomes. Upon impact onto a hydrophobic surface, the drop starts spreading under inertia effect. At the same time, a new surface area is created and droplet's kinetic energy undergoes progressive conversion to surface energy. This kinetic energy is also dissipated by viscous forces. In the case of high inertia impacts, the effects of capillary forces and viscous dissipation are not sufficient to avoid "*splashing*" so the drop disintegrates into smaller droplets [4]. In the case of moderate inertia, when kinetic energy is completely converted to surface energy, the droplet stops spreading indicating its maximum diameter. Then, the droplet starts to recoil under the action of capillary forces. In the case of pinned contact line, the droplet attains equilibrium but if the retraction is high, the contact line moves and the drop bounces off the surface [4,32]. Dimensional analysis provides a list of relevant parameters useful in discriminating these drop impact outcomes. For example, Weber number which compares impact inertia to surface energy is given by:

$$We = \frac{\rho D {V_i}^2}{\gamma}. \tag{1}$$

where ρ is fluid density, D is droplet initial diameter, V_i is impact velocity and γ is fluid surface tension. According to previous studies, droplet rebound can take place when Weber is higher than unity and above a certain limit [18]. In addition, a very high *We* can lead to droplet disintegration [32,42]. Droplet impact and spreading can also be described by the Reynolds number defined as:

$$Re = \frac{\rho D V_i}{\mu} \tag{2}$$

where μ is fluid viscosity. This number compares droplet inertia to viscous dissipation which means that droplet impact of highly viscous droplets (resulting in low Reynolds number) can lead to low droplet spreading extent. In addition, Ohnesorge number expressed as follows reveals the importance of viscous forces with respect to the aerodynamic and capillary forces.

$$\mathrm{Oh} = \frac{\mu}{\sqrt{\rho \gamma D}} \tag{3}$$

Oh can be described to scale the resisting force to the recoiling motion. It has been reported that increasing *Oh* number slows down recoiling behavior and can even lead to rebound and splashing inhibition [40,41]. Gravity related effects are described through the Froude number which compares droplet inertia to gravity and the Bond number which is the ratio of gravity to surface tension. The Froude and Bond numbers are described by the Eqs. (4) and (5) respectively:

$$Fr = \frac{V_i^2}{g \cdot D} \tag{4}$$

$$Bo = \frac{\rho g D^2}{\gamma} \tag{5}$$

where g corresponds to gravity.

The role of viscous dissipation has been extensively studied and proven to play an important role in splashing and rebound inhibition [4,20,47]. However, a high viscosity has its drawbacks; such fluids would be too difficult to pump and atomize through spraying nozzles [4]. In addition, a higher viscosity reduces the maximum spreading diameter of the drop and consequently increases film coating thickness; which reduces the ability to form a thin, uniform and smooth coating [48,49]. The use of a shear thinning formulation for the binder solution may be interesting in coating applications as the viscosity decreases with increasing shear rate [50–52]. Extensive research has also focused on the droplet impact of non-Newtonian fluids with a low shear viscosity and high viscoelasticity. For example, the effect of adding flexible polymers on rebound and splashing suppression has been reported by different workers [19,47,53–58]. This phenomenon was initially explained by the energy dissipation caused by an extensional viscosity within the droplet. However, recent works showed that dissipation was mainly caused by the *coil–stretch transition* of polymer molecules near the droplet contact line [59,60]. To conclude, polymers reinforce the stability of the drop and change the mechanisms of energy distribution and dissipation within the fluid. However, further investigations are needed to describe the phenomena occurring during droplet impact of non-Newtonian fluids especially on hydrophobic surfaces [4,46,61].

The present work deals with different phenomena that occur during impact of aqueous droplets of sodium CarboxyMethylCellulose solutions (CMC) on a hydrophobic surface. This cellulose derivative polymer is widely used in powder coating processes for its good film forming ability. In addition, CMC solutions exhibit both elastic and shear thinning effects which is interesting in film forming applications. However, little attention has been paid to the investigation of droplet impact of such solutions [62–64]. The aim of this study is to understand the effect of droplet velocity, size and formulation on droplet behavior upon impact. These factors are varied to study droplet impact for a wide range of Ohnesorge (0.002–0.9) and Weber (1–760) numbers. For example, high *We* values are obtained for high droplet diameters or velocities; while high *Oh* values are obtained either for high viscosities (corresponding to high solution concentrations) or for low droplet diameters. These dimensionless numbers are used to describe the predominant forces during the different phases (impact, spreading and retraction) for each injected droplet and to plot a regime diagram showing the different droplet outcomes according to *Oh* and *We* numbers.

2. Material and methods

2.1. Experimental apparatus

A high speed camera (i-speed 3, Olympus) was used to visualize the impact of single droplets onto a hydrophobic and rough surface. The zoom was made using a 105 mm F2.8 lens and the droplets were illuminated by (ILP2, Olympus) source light. The hydrophobic substrate purchased from *Thermo Scientific Corporation* was made by the coating of glass slides by a Teflon layer. The millimeter sized droplets were generated using a syringe with a 0.7 mm orifice needle and injected from different heights varying from 8 to 80 cm to obtain different velocities. Drop-on-demand technology was used to generate micron sized droplets. Fig. 1 shows that the micro-droplet generator contains a cylindrical chamber where the liquid is trapped. The chamber is connected to the nozzle by a 2 mm tube and contains a piezoelectric disk linked to a signal amplifier. When an electrical field is applied to the piezoelectric element, the piezoelectric disk shape changes and the deformations cause a pressure pulse in the fluid which ejects a droplet from the nozzle.

During our experiments, the signal coming from the amplifier was controlled by a waveform generator (SYSCOM-WGM 201) linked to a computer by a USB cable. The waveform settings could then be varied through the graphical user interface control panel of the "WGM-201" software by choosing the shape of the waveform, signal amplitude, signal duration…etc. Micron droplets were generated using a square wave voltage pulse. A typical example of the waveform is provided in Fig. 1 showing that the signal was divided into a positive part, a negative part and a neutral part. The duration of the positive part determined the duration of the absorption phase, in which a depression was created inside the nozzle. In the same manner, the duration of the negative part determined the length of the pressure phase after which a droplet was ejected from the nozzle. The ejected droplet size and velocity depended