Powder Technology 206 (2011) 63–71

Contents lists available at ScienceDirect

Powder Technology

journal homepage: www.elsevier.com/locate/powtec

Granule nucleation and growth: Competing drop spreading and infiltration processes

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ARTICLE INFO

Article history:
Received 5 August 2009
Received in revised form 18 May 2010
Accepted 18 June 2010
Available online 25 June 2010

Keywords:
Granulation
Nucleation kinetics
Drop spreading
Drop penetration
Tanner Law

ABSTRACT

Wet granulation is the formation of powder particle assemblies bound by a liquid binder. Granules are produced in order to prevent mixture segregation, improve flow characteristics and ease of powder handling. During this process powder is agitated inside a high shear mixer or fluidised bed and a liquid binder is added onto the powder mixture. The binder droplets then penetrate into the powder and bind them together. The kinetics of this drop-powder interaction is strongly linked to the properties of the granules produced, with shorter penetration times often leading to a more desirable narrower distribution of properties, providing the assembly formed is strong enough. Whilst there have been several separate studies into drop penetration times into porous powders and drop spreading on non-porous surfaces, relatively little work has focussed on the competitive spreading–infiltration process that would occur in reality.

To investigate this, single drop penetration experiments were carried out into static dry and pre-wetted powder beds. Pre-wetted in this case means to have been previously wetted by one identical drop. Viscous binders have been shown to exhibit lesser degrees of spreading in shorter times on non-porous surfaces [1,2]. However, the current work has discovered that, on porous surfaces, the infiltration rate has a greater degree of dependence, to that of spreading, on changes in viscosity. This means that higher viscosity binders will spread comparably further but require a smaller reduction in 'remaining drop' volume for capillary induced suction to pin the contact line.

1. Introduction

Wet granulation is a Unit Operation commonly employed in the Pharmaceutical, Food and Chemical Industries and involves the formation of aggregates/granules by sticking together fine powdery materials using a liquid binder [3]. This term encompasses a number of mechanisms; 'wetting and nucleation', 'consolidation and growth' and 'breakage'.

This paper considers 'wetting and nucleation' which involves the initial interaction of the liquid binder droplets impacting on a powder bed surface in a high shear mixer. The liquid bridges that form between powder particles and capillary forces, due to the surface tension of the binder, will act to pull them together. The larger 'agglomerate particles' that result, are termed 'nuclei'. The importance of penetration time on nucleation and the quality of final granules has been highlighted in a number of studies [4,5]. In granulation processes, faster penetration times are desirable in order to maintain operation in a 'drop controlled' regime which produces a narrower distribution of final granule properties [6]. However, the drop penetration times can vary over several orders of magnitude and penetration behaviour on loosely packed porous beds is complex and highly dependent on the microstructure of the bed [3,7].

Drop penetration is driven by the Laplace Capillary Suction Pressure P_c, Eq. (9), where $R_{pore,}$ γ_{lf} and θ are the characteristic pore size of the bed, liquid surface tension and the solid–liquid thermodynamic contact angle respectively.

$$p_c = \frac{2\gamma_{lf}\,\mathrm{Cos}\theta}{R_{pore}} \tag{1}$$

Based on Eq. (1), theory presents that there are two limiting cases for the penetration of a droplet into a porous material, Fig. 1. In the first case A) the contact line of the drop with the solid surface remains anchored to its initial position, implying that the contact angle decreases and that the radius of curvature of the drop increases as penetration proceeds, this is often referred to as the constant drawing area (CDA) penetration. The other limiting situation B) assumes the contact angle of the drop remains constant as it penetrates the bed, and the base area decreases as the volume decreases, this is often referred to as the decreasing drawing area case (DDA) [8]. In this work τ refers to a predicted time, whereas t or T refer to actual times.

Equations predicting the time required for a drop to penetrate into a powder bed, Eq. (2) and Eq. (3), are based on the Washburn equation, assuming that the bed can be approximated as a bundle of

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doi:10.1016/j.powtec.2010.06.013

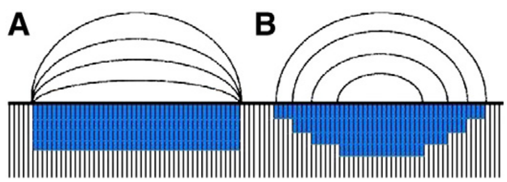

Fig. 1. Limiting cases of drop penetration on a porous surface, A) constant drawing area case, B) decreasing drawing area case. Modified from [3].

concentric non-interconnected tubes, of characteristic radius R_{pore} given by Eq. (4), and that the drop sits on the surface of the powder with a spreading diameter equal to the diameter of the original spherical drop. V_0 and μ are the drop volume and viscosity, Φ and d_{32} are the sphericity and surface-volume (Sauter) mean size of the powder particles, and ε is the bed porosity.

$$\tau_{CDA} = 1.35\frac{V_0^{2/3}}{\varepsilon^2 R_{pore}}\frac{\mu}{\gamma_{lf}\cos\theta} \tag{2}$$

$$\tau_{DDA} = 9\tau_{CDA} \tag{3}$$

$$R_{pore} = \frac{\Phi d_{32}}{3}\frac{\varepsilon}{(1-\varepsilon)} \tag{4}$$

A derivation for Eq (3) is provided by Denesuk at al [9]. Some of the limitations of the assumptions used in these equations have been presented previously [7]. In reality, there are competing drop spreading and infiltration processes taking place such as those considered by Clarke et al. [10], followed by a wetting inversion, or receding drop contact line. Fig. 2 illustrates this phenomenon for infiltration of a porous substrate, which could be anything from a loose powder bed to a consolidated granule produced in a high shear mixer.

The schematic shown in Fig. 3 describes the way in which the depression induced by the suction of the liquid acts as an additional resistance to spreading. Whereas the hydrostatic pressure generated by a drop volume that is too large would act as an additional driving force for spreading [11,12]. Too large here refers to a drop size greater than the capillary length k^{-1}, for which flow is dominated by gravity, where the capillary length defines the transition from the dominance of capillary force to body forces. When gravity dominates the drop shape becomes essentially flat with curvature near the drop edge [12].

This work will focus on drops that are below this transition, which occurs at drop diameters equal to the capillary length given by Eq. (5) where g is the acceleration due to gravity and ρ is the liquid density.

$$k^{-1} = \left(\gamma_{lf}/\rho.g\right)^{1/2} \tag{5}$$

In work by Sikalo et al. [1], whose work looked at effect of varying Weber number Eq. (6) and Reynolds number Eq. (7), where U_0 and D_0 are the impact velocity and spherical droplet diameter respectively, it was determined that higher viscosity produced a smaller maximum spread. This is because the increased viscous dissipation decreases the rate of spread. Moreover, a viscous droplet approaches its maximum spread in shorter time. A droplet with a lower surface tension was found to splash at smaller Weber numbers than a droplet with a higher surface tension [1]. Werner et al. made similar observations [2].

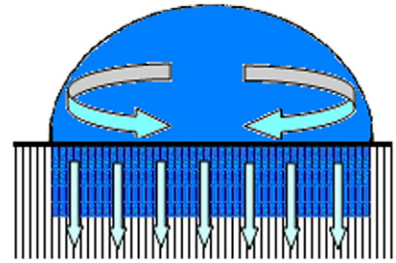

Fig. 3. Imbibition-induced liquid suction (small vertical arrows) and resulting hydrostatic depression inside the drop (large arrows). This depression can overcome the spreading, causing pinning of the triple line and eventually the retraction of the drop (wetting inversion), modified from [11].

$$We = \rho U_0^2 D_0 / \gamma_{lf} \tag{6}$$

$$Re = \rho U_0 D_0 / \mu \tag{7}$$

The time (t) based evolution of spreading diameter (d) and dynamic apparent contact angle (θ_D), on a smooth porous substrate has been studied by a number of authors [13–17]. The apparent contact angle here is termed 'dynamic' since the system is not in equilibrium during spreading.

$$d \propto t^{0.1} \tag{8}$$

$$\theta_D \propto t^{-0.3} \tag{9}$$

The speed of the contact line is given by a competition between capillary driving forces and viscous dissipation only, yielding Tanner's law shown in Eqs. (8) and (9), and reflects the competition between capillary forces whose amplitude is given by the surface tension and viscous dissipation given by the viscosity [13,15]. Although Tanner developed this proportionality for conditions where the surface tension was the only driving force considered e.g. no 'impact' effects, the same relationship was also obtained by Brochard-Wyart et al. [17] from a balance between the viscous dissipation and the work done by the surface tension force [14].

Although Tanner Law has been shown to adequately describe the evolution of diameter and contact angle with time during spreading on non-porous surfaces it is unable to explain why these kinetics are independent of the spreading parameter S, see Eq. (10) (where subscripts s, l and f denote solid, liquid and gas respectively), which should be the driving energy for spreading [18]. This independence can be explained by the precursor film formation. The spreading

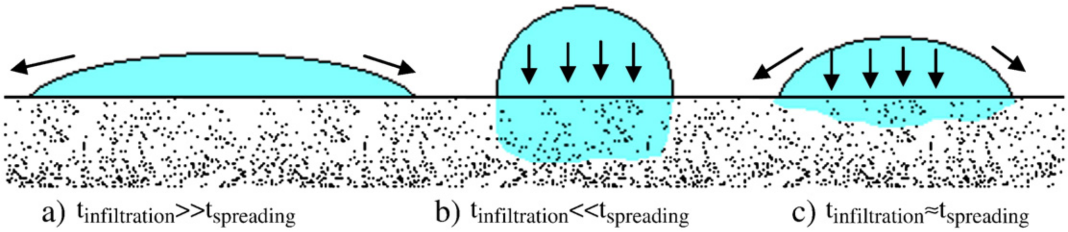

Fig. 2. Spreading and infiltration of a droplet on a porous, wettable powder surface: (a) spreading without infiltration; (b) infiltration without spreading; (c) simultaneous spreading and infiltration, where t is time.