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A semi-empirical model relating flow properties to particle contacts in fine binary powder mixtures

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ABSTRACT

Binary powder mixtures, consisting of large and small diameter particles, are used in electrophotographic (EP) printing devices where control of the relationship between the flow properties and mixing ratio is of great importance. In this study, we have experimentally investigated the bulk flow properties (*i.e.* bulk cohesion and internal friction) of large-and-small fine binary powder mixtures (size ratio: $d_l/d_s = 9.17$) as a function of the volume ratio of the smaller component, x_s. Using polymer spheres as the smaller constituents, whose bulk cohesion was controlled by surface additives, we have obtained systematic data on the effect of the smaller constituent. A semi-empirical model was then developed, in which the total bulk cohesion is expressed as a linear superposition of the numbers of the three types of particle–particle contacts (*i.e.* large–large, small–small, and large–small) involved in the shear motion. The model successfully reproduced the experimental data after fitting appropriate values of a parameter representing the adhesion at large–small particle contacts. The rapid increase of the number of particles involved in the shear motion as a function of x_s near $x_s = 0$ was also considered in the model. The model clarifies the contributions of the three types of particle–particle contacts to the total bulk flow properties. The effect of large–small particle contacts on the bulk cohesion was observed for almost all values of x_s. The value of x_s at which the contribution of large–small particle contacts in the total bulk cohesion becomes a maximum shifted in the direction of larger x_s when a less cohesive small powder was used. This also resulted in a shift of x_s at which the internal friction reached a minimum.

1. Introduction

Two-component dry powder developers, comprised of binary mixtures of small and large particles, have been used in electrophotographic (EP) printing devices such as laser printers and photocopier machines for more than half a decade [1]. Two-component developers are composed of toner – resin particles with a diameter of 5–10 μm functioning as colouring materials – and carrier particles, usually made of magnetic materials with a mean diameter of 50–100 μm, which promote the electrostatic charging of the toner particles and carry the toners onto the latent images. Inside the EP printing device, a mixture of toner and carrier particles is intermittently circulated under applied shear and normal stresses in the range of 1–10 kPa. Since the flow behaviour of the developer affects the image quality, it is crucial to control their flow properties appropriately [2,3].

Whilst many of the factors affecting the flow properties of two-component developers, such as particle size and shape distributions, composition, and mechanical characteristics, do not change significantly during normal operation of EP printing devices, the mixing ratio, or compositional balance, of the constituent particles is one of the most important variables to control in the process [1]. Therefore, understanding the relationship between the flow properties and mixing ratio of binary powder mixtures is of great importance for proper control of the flow behaviour of EP two-component developers.

In this study, we present a semi-empirical model which describes the relationship between the bulk flow properties of large-and-small dry fine binary powder mixtures and the ratio of the component powders, incorporating the contacts between the constituent particles. Specifically, the model estimates the number of particle contacts involved in the incipient flow motion, taking into account the change in the width of the incipient shear zone, and links these to changes in the flow properties. In fact, an understanding of the fundamental physics connecting changes in the particle contact network and width of the incipient shear zone to the onset of incipient flow is of wide benefit to a range of granular systems beyond EP printing devices such as the processes of powder materials in pharmaceutical and food industrials.

However, despite its importance, the number of fundamental studies exploring the bulk flow properties of large-and-small binary powder mixtures in terms of the mixing ratio of the constituent particles is still limited. There exists some experimental research investigating the relationship between the bulk flow properties and mixing ratio of the binary powder mixtures [4–6], but the analyses of the mechanisms were rather qualitative. Computational modelling studies also exist

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(*e.g.* Ref. [7,8]) but the results are again rather limited due to the high complexity of such systems, although this might potentially be a useful approach with further improvements of computational speed and modelling techniques in the near future. The current study aims to address the gap between experimental measurements and numerical simulations towards the ultimate goal of an improved understanding of their linkage.

In this study, we firstly investigated the bulk flow properties of fine binary powder mixtures experimentally. We used fine powders to construct model systems of two-component developers used in EP printing devices, thus eliminating any confounding factors such as the varying geometric shapes of the constituent powder particles and unknown surface treatments. In a previous study, we successfully performed systematic measurements by using commercially-available toner powders as the smaller constituent of the mixtures [2]. However, in present work, in order to develop a model which correctly describes the adhesive property between the constituent particles, we utilised spherical polymer powders as the smaller constituents whose bulk cohesions were systematically controlled.

Then, a semi-empirical model, which explains the bulk flow properties of the binary powder mixtures in terms of the contacts of the constituent particles, was developed by estimating the number of particle contacts involved at the point of incipient flow. There exist some studies discussing the particle packing of two- or multi-component powder mixtures from the microscopic viewpoint, relating the bulk packing density to the number of particle contacts (*i.e.* coordination number), including experimental measurements [9,10], analytical modelling [11–16], and computational simulations [17]. In this study, a geometrical model proposed by Suzuki et al. [12–14], in which the coordination number of the constituent particles in the binary powder mixtures is calculated from the bulk density (or voidage) of the systems, was used for the estimation of the numbers of particle contacts in the system. The experimental data were then analysed further using the proposed model to determine its validity.

The remainder of this article is structured as follows: after describing the experimental methods and materials in Section 2, the results of the measurement of the powder packing structures and flow properties such as bulk cohesion and internal friction of the binary powder mixtures are presented in Sections 3 and 4, respectively. A model equation to relate the particle contacts in the powder mixtures and their flow properties is then introduced in Section 5, and further analyses based on the model are given in Section 6. We conclude by summarising the achievements in Section 7.

2. Experimental methods and materials

Several types of powders composed of smaller particles with different bulk cohesions were prepared for the purpose of developing a model describing the adhesive property between the constituent particles properly. Binary powder mixtures were then made using each small-particle powder together with a large-particle powder, in which the mixing ratio was systematically changed, and the incipient flow properties of the powder mixtures were measured under the same experimental conditions. The details of the measurement methods of the flow properties, as well as the powder materials used for the binary mixtures, are described in the remainder of this section.

2.1. Measurement of flow properties

The incipient flow properties of the powder samples were evaluated using a ring shear tester (RST) RST-XS (Dietmer Schulze, Germany). The fundamental principles of shear testing, as well as a description of the RST equipment, are summarised elsewhere in various textbooks and papers (*e.g.* Refs. [18,19]).

The series of measurements were conducted under the same experimental conditions as Ref. [2]. The powder sample was at first placed into the ring-shaped shear cell of height 13 mm, inner and outer diameters of 32 and 64 mm, respectively, and the lid with radial vanes of height 3 mm distributed every 22.5° on its lower surface, was loaded onto the sample. In each measurement, the incipient shear stresses, τ_{sh}, were measured under normal stresses of $\sigma_{sh} = 0.6$, 1.1 and 1.6 kPa after pre-shear consolidation with a normal stress of $\sigma_{pre} = 2.0$ kPa.

In all cases, the obtained data showed good linearity (correlation coefficients, R^2, of the linear fitting were greater than 0.994). Based on this, two parameters representing major incipient flow properties, bulk cohesion and internal friction, were obtained from the yield locus, a line fitted to the three measured (σ_{sh}, τ_{sh}) points in the σ–τ plane (*i.e.* a Mohr–Coulomb yield locus) and expressed by the following equation:

$$\tau = \sigma \cdot \tan\varphi_{lin} + \tau_c. \tag{1}$$

In Eq. (1), τ_c, the τ-intercept of the yield locus, represents the bulk cohesive property of the powder sample and is called 'bulk cohesion'. Also, the angle of the slope of the yield locus, φ_{lin}, is a measure of the internal friction of the powder sample and is termed the 'linearised friction angle (LFA)'.

The bulk structure of every powder sample was also evaluated from the voidage, ε, which represents the volume ratio of the voids in the total volume of powder system. The value of ε was calculated from the bulk density of the powder, ρ_b, obtained in the RST measurement, particle densities of the large and small components: $\rho_{p,l}$ and $\rho_{p,s}$, and the volume ratio of the small powder, x_s, by using the following relationship:

$$\varepsilon = 1 - \frac{\rho_b}{\rho_{p,l}(1-x_s) + \rho_{p,l}x_s}. \tag{2}$$

Each measurement was carried out under standard conditions of 21 ± 2 °C and 38–55% relative humidity (RH). The fluctuations of room temperature and RH did not affect the results.

2.2. Powder materials

Glass ballotini (Ballotini solid soda glass balls, diameter 40–70 μm, Sigmund Lindner, Germany) of the same type as in a previous study [2] were used as the larger component in the binary mixtures. Before being used for the flow measurement, the ballotini powder was washed using liquid detergent and rinsed thoroughly in distilled water in order to eliminate fine impurities on the particle surface. After being dried in the cabinet with an internal temperature of approximately 80 °C for at least 8 h, it was then sieved using a set of three nested sieves with different mesh sizes (38, 90 and 100 μm from bottom) in order to obtain a sufficiently uniform and narrow size distribution. Only the powder remaining on the 38 μm sieve was used for the evaluation of the flow properties. For the smaller component of the binary mixtures, a polymer powder composed of poly(styrene-*co*-divinylbenzene) (PS-DVB) microspheres (Sigma-Aldrich, USA) was used without pre-treatment process.

The cumulative size distributions of the powders obtained by image analysis are shown in Fig. 1, together with representative SEM images of each type of particles (SEM: JEOL 6340 F FEG–SEM). Under the SEM, the constituent particles of both glass ballotini and PS-DVB powders appeared almost spherical, and also the surface of the glass ballotini was confirmed to be almost smooth and impurity-free. The size parameters such as volume mean diameter, d_{50}, and geometric standard deviation, σ_g, of each powder were obtained by fitting a log-normal function, which closely followed the cumulative size distribution of each powder. The particle density of each powder was measured by a gas pycnometer (Accupyc 1330, Micromeritics, USA). The values of size parameters and particle density are summarised in Table 1.