NOTE

SIMULTANEOUS EFFECTS OF ADHESION AND POLY-DISPERSITY ON MICROSTRUCTURES AND GEOMETRICAL PROPERTIES IN PARTICLE DEPOSITION

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Abstract—The simultaneous effects of adhesion and polydispersity on packing (or deposit) microstructures and their bulk properties are examined. The results show that the microstructures and bulk properties of the deposits vary sharply with the introduction of even small adhesion and polydispersity. A structural phase diagram is obtained as functions of adhesion and polydispersity. Increases in adhesion lead to noticeable or large fluctuations in packing fractions for polydisperse systems. However, the packing fraction can be stabilized and the fluctuations greatly reduced regardless of the magnitude of the polydispersity index by keeping the adhesion relatively low (i.e., $s \leq 0.1$).

INTRODUCTION

In the previous work the individual effects of either adhesion [1] or polydispersity [2] on the microstructures and their properties in particle deposition have been effectively investigated in terms of sticking probability (s) and polydispersity index (σ), respectively. The sticking probability is defined as the probability that a depositing particle sticks on contact with the other deposited particle, and the polydispersity index is defined as the standard deviation from the normal distribution of particle radii. In addition, it has been noted that a number of characterization methods developed are quite useful to analyze microstructures, even having anisotropic properties.

In the present work the simultaneous effects of adhesion and polydispersity on microstructure formation and on bulk properties of the structures are examined, which have been little studied previously. From close observations of the global and local density functions, diffraction patterns obtained from structure factors, and correlation lengths, a structural phase diagram are prepared. The packing fractions as a function of the polydispersity index are compared on a much fine scale for different sticking probilities in order to examine (i) the variation of the packing fraction with σ for each s and (ii) the extent of deviation of the packing fraction from the average at each σ. Also, the changes in coordination number are compared.

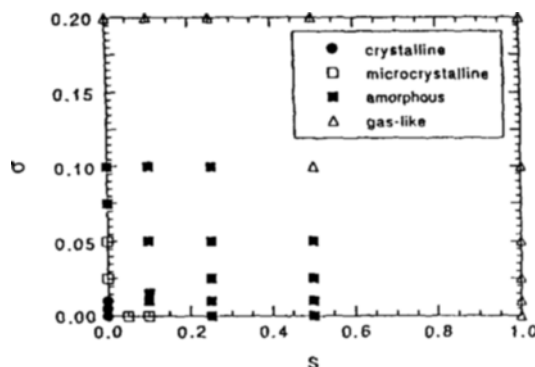[†]To whom all correspondences should be addressed.

Fig. 1. Phase diagram drawn in terms of the sticking probability and the polydispersity index.

Computational details of deposition algorithm and structure characterization used here are available elsewhere [1-3].

RESULTS AND DISCUSSION

1. Structural Phase Diagram

A structural phase diagram is prepared in terms of polydispersity and sticking probability, as shown in Fig. 1. Interestingly, one can see that, depending on the degree of adhesiveness and/or polydispersity, the packing structures can be classified into crystalline, microcrystalline, amorphous, or gas-like phase, though the boundaries among phases are not clearly defined here. Typical examples of diffraction patterns
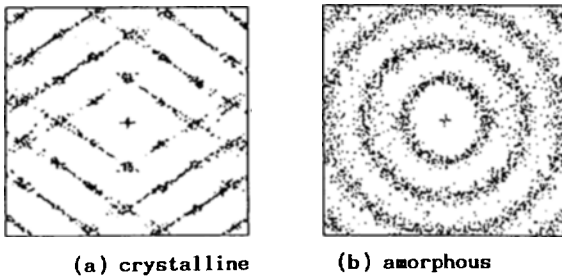
(a) crystalline   (b) amorphous

Fig. 2. Diffraction patterns observed for different phases.

observed for different phases are shown in Fig. 2. From this phase diagram it is noted that adhesion

and polydispersity prevent the formation of ordering. Only for small deviations from monodispersity and for the absence of adhesion does one have locally ordered regions separated by defects such as stacking faults, larger pores, etc. Beyond this region amorphous structures appear for broad ranges of the sticking probability and/or the polydispersity index.

2. Packing Fraction

First, the change in packing fraction as a function of polydispersity is considered for the packing of non-adhesive particles, as shown in Fig. 3a. At very small values of σ the packing fraction shows a sharp decrease from that for monosized particles and goes through a minimum as increases. Beyond the mini-
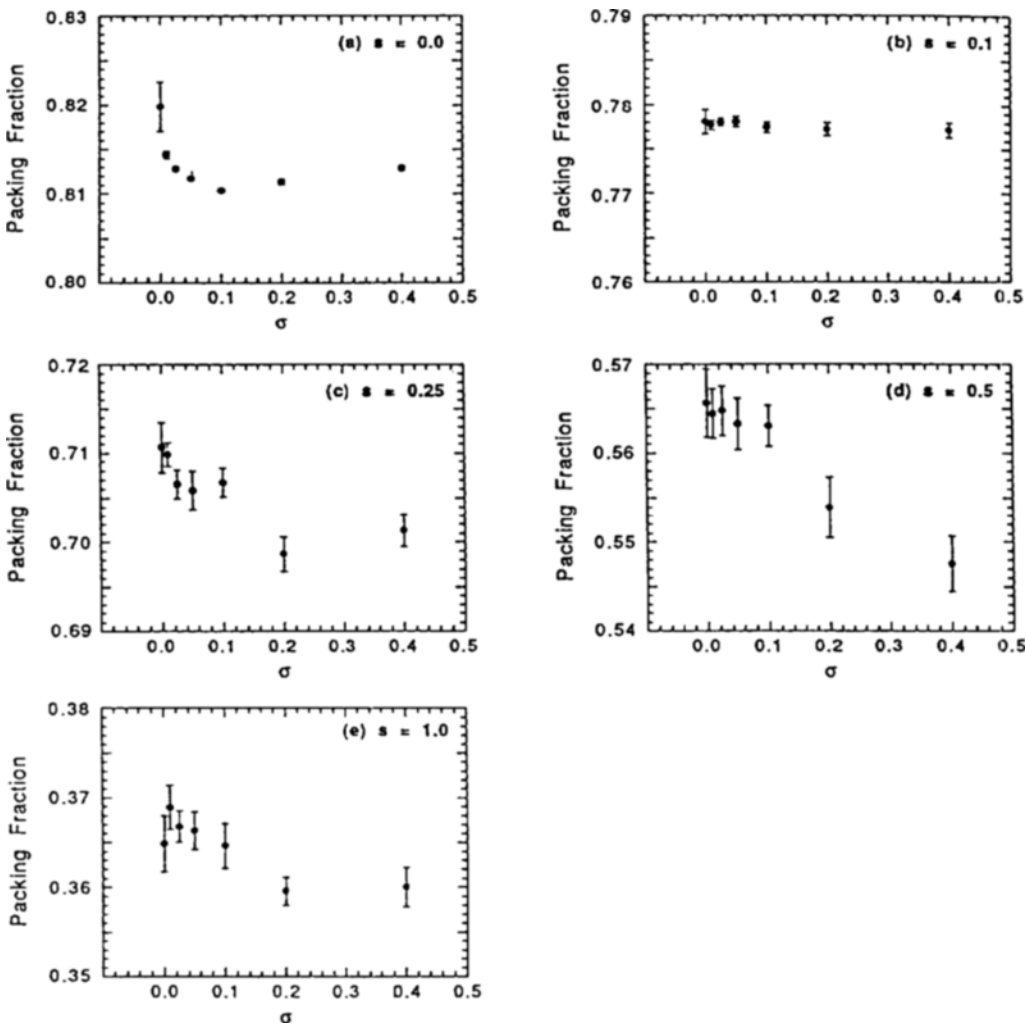

Fig. 3. Comparison of the variations in packing fraction as functions of the polydispersity indices for different sticking probabilities.
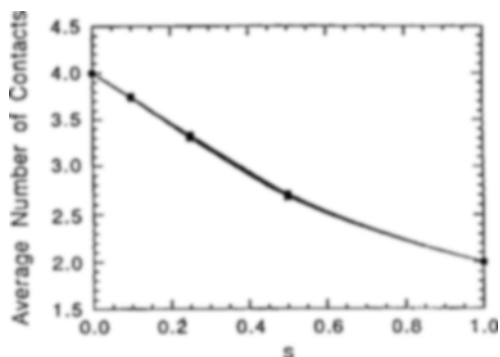
Fig. 4. The changes in average number of contacts as a function of the sticking probability for different polydispersity indices ($\sigma = 0.0$, 0.1, 0.2, and 0.4).

mum a slow monotonic increase is seen. A limiting, asymptotic value is reached when σ becomes infinite (i.e., for uniform particle distribution). This asymptotic value is lower than the value at $\sigma = 0$. Such a behavior is found only in two-dimensional gravitational packing, in contrast to what is observed in three dimensional packing (where the introduction of polydispersity always increases the packing fractions). The reason for this difference is that in the case of two-dimensional packing smaller particles have a reduced degree of freedom, because of the reduced dimensionality, to permeate through the network of larger particles (For this reason, segregation in three dimensions cannot be investigated using two-dimensional simulations.). Such a minimum in the packing fraction always appears in two-dimensional packings when polydispersity is present. The minimum obtained in this work is about 0.810, at a value of σ near 0.1. This value compares well with those available from the literature (0.8096 ± 0.02 for binary mixtures [4] and 0.8113 ± 0.0013 for triangular size distribution with σ near 0.1 [5]). Also one can see from Fig. 3a that the packing fraction for monosized particles (i.e., for $\sigma = 0$) shows large fluctuations (i.e., large variance) from the average value, whereas the fluctuations for polydisperse particles are very small. This again confirms an important property of monodisperse packings. Monodisperse packings in general may show considerable variations in the packing fraction, and, as a consequence, properties

and phenomena that are influenced by packing fraction may also vary widely. In contrast, when polydisperse particles are used, packing fractions show very small variations and the resulting properties also show only correspondingly small variations.

The variations in packing fraction with the sticking probability and the polydispersity index are illustrated in Figs. 3b-e. From these figures two points can be emphasized: (i) with increases in the 'stickiness' of the particles, the packing may show noticeable or large fluctuations in packing fractions and associated properties, and (ii) for small (but nonzero) values of s the packing fraction becomes uniform regardless of the extent of polydispersity.

3. Coordination Number

Fig. 4 shows that the average number of contacts (i.e., coordination number) is nearly constant for different polydispersity indices. This indicates that the coordination number for polydisperse packings remains close to that for monodisperse packings and is relatively insensitive to particle size distributions. This behavior is consistent with that observed in experiments [6]. Also, the coordination number decreases monotonically with increasing sticking probability.

NOMENCLATURE

s   : sticking probability

Greek Letter

σ : polydispersity index

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