Coexistence of gas-liquid-solid phases in colloidal uncharged hard spheres induced by a disc-like depletion agent

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The semi-grand canonical ensemble theory within the free volume approximation was applied to construct a composite Helmholtz free energy density function which describes a mixture of colloidal neutral hard spheres and a disc-like depletion agent in its liquid and solid phases. Instead of calculating the pressure and chemical potential that are commonly employed to construct the phase-diagram boundaries, we study this kind of colloidal particles whose interparticle interactions are of entropic origin by the more experimentally appealing means of free energy density minimization method [1]. The method yields the phase-diagram domains (rather than the phase boundaries) of homogeneous single phases (gas, liquid and solid) as well as their coexisting bi- and triphases. The calculated phase-coexisting domains have the same patterns as one often sees in laboratory experiments. Our theoretical colloid-disc phase diagrams contain the well-known triangular area of coexisting gas-liquid-solid three phases, but it has to be realized as some kind of a kinetic coalescence of two sets of biphases. One spectacular feature in our findings is that the minimized coexisting three phases (gas, liquid and solid) always assume the same vertices of a triangle for any set of initial concentrations of colloids and discs that falls inside this triangular area, but on a closer examination, the spatial volume of each phase varies with different initial set of concentrations. It would thus be interesting and a challenge if laboratory experiments at the same quantitative level as those reported previously for the colloid-polymer mixture [2] can be carried out to confirm this theoretical scenario.

Acknowledgements This work is supported by the Ministry of Science and Technology (MOST103-2112-M-008-015-MY3), Taiwan.

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[2] F. Renth, W.C.K. Poon and R.M.L. Evans, *Phys. Rev. E*, 2001, **64**, 031402.