Defined core-shell particles as the key to complex interfacial self-assembly

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Self-assembly of colloidal particles can yield defined surface patterns over macroscopic areas. However, the prevalence of hexagonal symmetries limits the structural versatility of self-assembling monolayers. In 1998 E. A. Jagla computationally modelled core-shell particles with two interaction length-scales and predicted the formation of complex minimum energy configurations [1]. Despite the elegance of this approach, its experimental realization has remained largely elusive. Here, we use iniferter-type controlled radical polymerization to create core-shell particles consisting of a silica core surface-functionalized with a non-crosslinked polymer shell (figure 1a) [2]. Upon interfacial compression, the resulting core-shell particles arrange in well-defined dimer, trimer and tetramer lattices before transitioning into complex chain and cluster phases. The experimental phase behavior is accurately reproduced by Monte-Carlo simulations and minimum energy calculations, suggesting that the interfacial assembly interacts via a pair-wise additive Jagla-type potential (figure 1b). The possibility to control the interaction potential via the interfacial morphology provides a framework to realize structural features with unprecedented complexity from a simple, one-component system [3].

[1] E. A. Jagla, Phase behavior of a system of particles with core collapse, Phys. Rev. E. Stat. Phys. Plasmas Fluids Relat. Interdiscip, Topics 58, (1998), p. 1478–1486.
[2] J. Menath, J. Eatson, R. Brilmayer, A. Andrieu-Brunsen, D. M. A. Buzza, and N. Vogel, Defined core–shell particles as the key to complex interfacial self-assembly, PNAS, 118, (2021), p. 1-10
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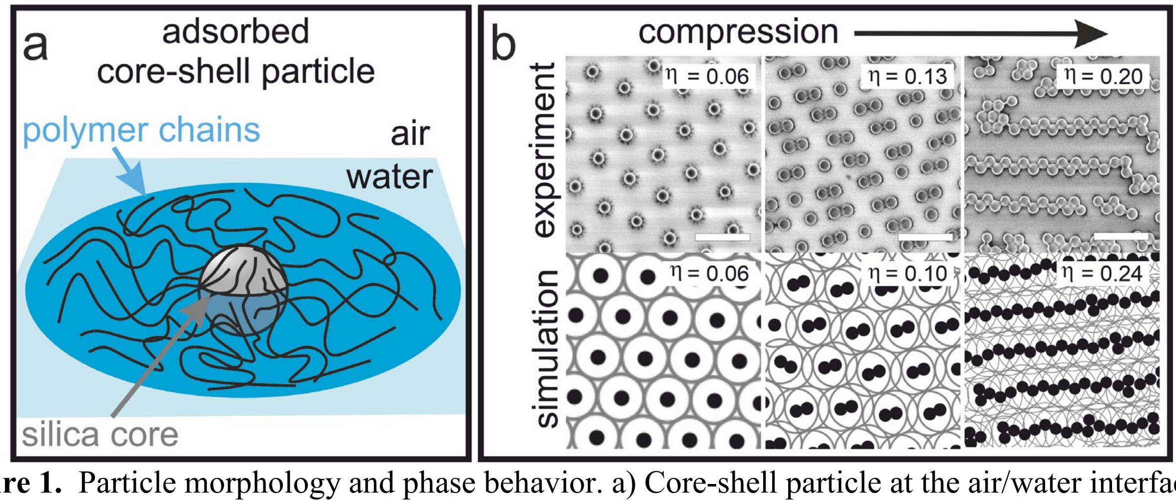

Figure 1. Particle morphology and phase behavior. a) Core-shell particle at the air/water interface. b) Scanning electron micrographs of the particle lattice upon interfacial compression and corresponding Monte-Carlo simulations. Scale bar: 1 µm.