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The Asakura-Oosawa theory

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In 1954, Asakura and Oosawa[1] explained that nonadsorbing macromolecules can induce attractive forces between colloidal particles in a single page paper in this journal. The effective pair interaction between the colloidal particles mediated by the nonadsorbing species is nowadays often termed the Asakura–Oosawa (AO) potential.[2] Figure 1 illustrates such a colloid–polymer mixture and its tendency to phase separate into a phase enriched in colloids and a phase concentrated in polymers due to the attraction mediated by nonadsorbing polymer chains.

This special issue of *The Journal of Chemical Physics* contains a collection of papers related to the concept introduced by Asakura and Oosawa. We are very honored that the late professor Oosawa contributed to this issue with a historical overview[3] on the origin of the Asakura–Oosawa theory. In his synopsis, he explains that he came to Nagoya to "do some unorthodox physics" and that experimental results have stimulated him to theoretically consider the effects of adding macromolecules to colloids.

Kurihara and Vincent[4] discussed how, after a lag time of about 20 years, the theory was rediscovered in the 1970s via experimental studies on colloid–polymer mixtures.[5,6]

In the same period, Vrij[7] independently found that excluded volume interactions in a colloidal dispersion containing an additional component have important consequences for the effective interactions, structure,[8] and phase stability.[9,10] Vrij explicitly introduced the description of penetrable hard spheres (PHSs) to describe the nonadsorbing polymers, which was implicitly proposed by AO. In hindsight, the delayed recognition clarifies that Asakura and Oosawa were ahead of their time. Since the 1980s, especially in the 1990s, attention concerning the influence of nonadsorbing macromolecules on the interaction between colloidal particles and the resulting phase behavior of colloid–polymer and binary colloidal mixtures gained increasing interest (see Fig. 2 in Ref. 4).

In 1980, the term "depletion"[11] was introduced to describe the effect of nonadsorbing species near a surface. In the field of colloid chemistry, the accumulation of species (ions, polymers, and proteins) at (colloidal) surfaces received quite some attention after the 1960s (see, for instance, Refs. 12 and 13). Terms such as positive and negative adsorption were common, and the latter was also termed depletion.

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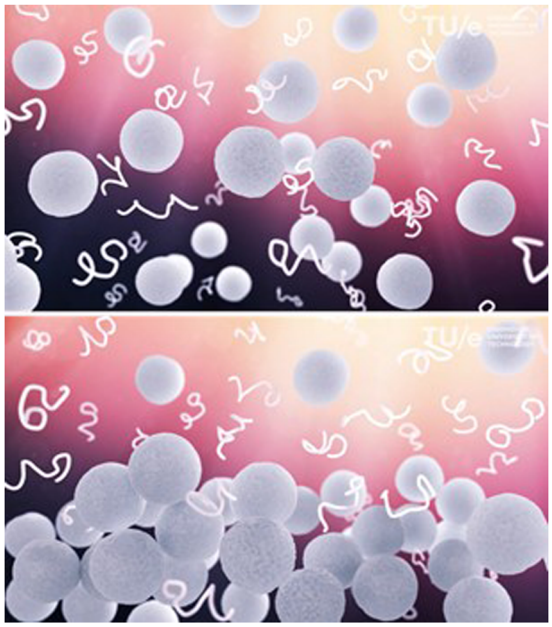

FIG. 1. Snapshots of an animation of a colloid–polymer mixture. The spheres are the colloidal particles that are mixed with the nonadsorbing polymer chains. The upper animation reflects the system just after mixing. The lower sketch shows that the dispersion gets inhomogeneous after some time if the concentration of the nonadsorbing polymers (sometimes called depletants) is sufficiently high. See also the simplified animation on http://youtu.be/tEsvuSnW-9Y.

In the decades that followed the work of Vincent[5,6,14,15] and Vrij[7–10] and their co-workers, various theoretical tools were applied to better understand the microstructure, pair depletion interactions, and phase stability of colloid–polymer mixtures. We summarize these below with a focus on bulk properties. For some relevant references on the interface physics in colloid–polymer mixtures, see Refs. 16–22.

A key step was the application of thermodynamic perturbation theory by Gast, Hall, and Russel[23] to predict the phase behavior of colloid–polymer mixtures to interpret systematic experiments[24,25] that were performed in several laboratories. At the same time, field theoretical methods were employed to understand the detailed polymer physics related to depletion.[26–28] Shaw and Thirumalai[29] formulated a reference interaction site model for colloids and combined it with the Edwards model for polymers to explain depletion stabilization effects:[11,30,31] at high polymer concentrations, repulsive contributions to the pair interactions appear.

Methods such as free volume theory (FVT),[32] polymer reference interaction site model (PRISM) integral equation theory,[33] integrating out the depletant using an effective one-component Hamiltonian,[34] density functional theory (DFT),[35–37] and a Gaussian core model[38] were applied to gain insight into depletion effects. Many theories treat polymers as effective soft spheres, which can be useful when considering dilute solutions of polymers much smaller than the particles, the so-called "colloid limit." New physics emerges in polymer semidilute solutions and melts, and it was found that the solvent quality also matters. Accounting for polymer physics is also relevant particularly in the "protein" limit,[39–41] where the particles are small with respect to the polymer chains. The "protein" limit regime is of particular interest also for (cell) biology.

Free volume theory (FVT)[32] (see also Ref. 42) is a simple, yet insightful and reasonably accurate, theory for the macroscopic phase behavior that also enables partitioning of colloids *and* depletants over the phases to be predicted. For colloid–polymer mixtures described as hard spheres plus PHS, the predicted phase diagrams correspond to computer simulation results.[43,44] FVT also allows accounting for interactions between the depletants[45,46] and to evaluate the rich phase behavior of anisotropic colloids mixed with nonadsorbing polymers.[47,48]

Microscopic equilibrium theories of thermodynamics, structure, and phase separation of polymer–particle suspensions that explicitly treat polymers and their conformational degrees of freedom were created by generalizing the PRISM integral equation approach.[33,49] The role of particle size (from the protein to colloid limits), polymer concentration (dilute and semidilute good and theta solvent conditions[50] and dense melts[51]), arbitrary particle volume fractions, and the full microstructural correlations were determined in a unified manner.

Experimental work focusing on measuring (i) the pair interaction (see, e.g., Refs. 52–55), (ii) the structure of the dispersion mediated by nonadsorbing polymers,[56] and the phase behavior[57,58] of well-defined colloid–polymer mixtures appeared. PRISM equilibrium predictions were successfully confronted against experiments.[59,60]

Soon, nonequilibrium phenomena in multi-component mixtures and the role of depletion effects[61–64] gained interest from both theoreticians and experimentalists.[65] Knowledge of the structural correlations computed using PRISM theory allowed microscopic dynamical theories of slow colloid dynamics to be constructed, at both the mode coupling and activated dynamics level. Quantitative predictions for the structural relaxation time, formation of glasses and gels, nonlinear rheology, and delayed gel collapse[66–68] were made and compared with experiments.[69,70]

The short-range and controlled strength of attraction induced by the AO interactions also offers an ideal playground for studying the glass transition and gelation of dense colloidal suspensions. In 2000, the mode-coupling theory (MCT), which is one of the most successful first-principles theories to describe the slow dynamics of the glass transition, has been applied to the hard-sphere system with short-range attraction.[71] It unveiled the existence of a series of singular dynamical ideal glass transitions as well as the re-entrance of the repulsive-to-attractive glass transitions. The theoretical prediction was soon verified by experiments for a mixture of colloidal particles interacting via the AO potential induced by depletant polymers.[63,72]

Progress on all the above elements was stimulated by computer simulation. Several works have focused on the influence of depletion-induced attraction on the structural and dynamical behavior of colloids, highlighting, for example, the onset of attractive and repulsive glasses and the occurrence of reentrant melting when the range of the depletion attraction is very small.[73,74] The idea to tune the "sticky" interaction by changing the concentration of the depletant has been extended to colloidal suspensions with lower

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densities and sparked a series of experimental and numerical studies on colloidal gels (see Refs. 75 and 76 and references therein). These studies succeeded in explaining the route from the glass transition at high densities to gelation of colloidal particles by tuning the concentration of depletant.

Another important research topic concerns the fate of the attractive glass/gel line at lower densities and the interplay with phase separation.[77] For the latter, an appropriate combination of simulations and confocal microscopy experiments was able to show that such a line intersects the binodal at high densities, giving rise to so-called arrested phase separation.[76] Arrested states induced by depletion have also been studied extensively in the context of hard-sphere/star polymer[78] or star/star mixtures.[79] The introduction of soft interactions is found to enrich the phenomenology of glass transitions and the interplay between the two species[80] compared to binary mixtures of hard spheres.[81]

As indicated above, the interest was broadening and systems such as colloidal spheres plus multi-component depletants[82] and dispersions of star polymers (soft colloids) plus linear polymers dispersions[83,84] received interest, as well as mixtures of different types of star polymers.[85] Besides studying the effects of nonadsorbing polymers as depletants, it is also of interest to treat colloids themselves as depletants added to a dispersion of larger or different colloids. Depletion effects also can be encountered in mixtures of self-assembling block copolymers in a selective solvent under the influence of nonadsorbing polymers.[86,87] The phase behavior of hard-sphere binary asymmetric mixtures gained attention in the 1990s.[88–91] Fundamental studies using DFT[37] helped to quantify the effective interactions and microstructural effects.[35] Studies of depletion effects in mixtures of different particle shapes are also gaining interest.[92–94] It also became clear that more specific effects, such as charges,[95,96] the presence of polymer brushes,[97,98] solvent quality,[99,100] and polydispersity,[101–104] are important.

A very interesting research direction involves systems where depletion attraction competes with a long-range repulsion, often of electrostatic nature, giving rise to the onset of equilibrium cluster phases and arrested states where clusters are dominant,[105] with important implications to understand features of solutions of globular proteins. Recent reviews on this topic can be found in Refs. 106 and 107. In addition, simulations have been very useful to locate and characterize gas–liquid (colloid-rich/colloid poor) phase separation of the Asakura–Oosawa effective potential,[108] clarifying that this belongs, as expected, to the Ising universality class. Investigations on binary mixtures of colloids and polymers[109] where non-ideality effects of the polymer are taken into account have also been explored.

Another rich direction of work in simulations is to calculate effective interactions between colloids immersed in different kinds of solutes that are more complex than polymers. This can be achieved by umbrella sampling or by exploiting virtual moves in Monte Carlo simulations. With these methods, depletion induced by soft spheres or microgels has been investigated.[110] The latter have also been recently used in experiments to modify the depletion interactions *in situ*, exploiting the thermoresponsive character of microgels.[111,112] Furthermore, a promising class of depletants involves a self-assembling medium, such as a patchy co-solute, forming supramolecular chains[113–115] or clusters[116] or even in the vicinity of a critical point,[117,118] thus providing a connection between depletion interactions and critical Casimir forces.

Colloid synthesis has evolved to such a degree[119,120] that it is nowadays possible to make colloidal particles of a wide range of shapes.[121–125] This, and the fact that anisotropic shapes occur in nature, has triggered studies on mixtures of non-spherical colloids plus added nonadsorbing polymers. Hence, insights have been obtained into the phase behavior in mixtures of rods,[93,126–129] platelets,[47,130–132] and cubes[133–135] plus added polymers. In addition, nonequilibrium phenomena are quite relevant here.[136,137]

Insights into depletion effects inspired by the Asakura–Oosawa concepts have also gained attention in the life science field. Already at an early stage,[138–140] it was appreciated that the large volume fraction occupied by the macromolecules in living cells has consequences. Walter and Brooks[141] suggested that macromolecular crowding is the basis for microcompartmentation. As summarized a few years ago,[142] excluded volume effects are thought to be of importance to explain several intracellular processes.[143,144] Hence, depletion effects are suggested to mediate several types of biological processes, including dynamics.[145,146]

It is abundantly clear that macromolecular crowding affects all aspects of biological processes ranging from transcription to self-organization of the molecules of life. Nowhere is it more transparent than in crowding-driven structural transitions in protein-like polymers,[147,148] conformational switches between active states of RNA,[149] and depletion effects on the conformations of DNA.[150–152] Depletion effects play a similar role in protein dispersions[153,154] and dispersed bacteria[155] as in colloidal suspensions. There are many more biological processes in which crowding effects, especially the consequences of polydispersity, have not been explored at all. Quantifying the effects of entropic forces in biology remains a virgin area for additional research.

In materials science, depletion effects were used in various ways to self-organize colloidal systems. An example is to select the strength of the attraction by introducing colloidal surface roughness,[156–158] which allows the creation of colloidal micelles.[159] The use of the different shapes can help to tune the strength of the depletion attraction. This can facilitate the use of depletion effects to make colloidal "key-lock" systems.[160] An interesting element that is gaining interest in this field is the influence of colloidal shape on the self-assembly of colloidal particles[119,161] and how shape can induce "entropic patchiness."

It is clear that the depletion field has begun to develop in many different new directions, of which we mention a few that connect to the contributions in this special issue:

- Influence of solvent quality and solvation on depletion effects mediated by polymers (see Refs. 162 and 163).
- Macromolecular crowding. In this issue, some novel insights are presented (see Refs. 164–169).
- Interesting findings on charged colloids, proteins, and bacteria upon addition of nonadsorbing polymers, while also specific effects of polyelectrolytes are considered (see Refs. 170–174).
- Confinement of multi-component mixtures (see Refs. 175 and 176).
- Complex (hard/patchy) colloidal mixtures (see Refs. 177 and 178).

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- Fundamental insights into (i) the fluid–fluid phase transitions using DFT (see Refs. 179 and 180) and (ii) equilibrium cluster fluids.[181]
- Anisotropic colloids and depletion effects (see Refs. 182–185).
- Glasses in colloid–polymer mixtures (see Refs. 186–188).
- Nonequilibrium phenomena in colloidal mixtures (see Refs. 189 and 190).

After almost 70 years, it turns out that the classical theory of Asakura and Oosawa is very much alive. This collection of papers highlights the relevance of the Asakura–Oosawa theory and shows its promise to further understand multi-component soft matter systems, with significant relevance for science, technology, and biology.

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