Supplementary Information for Caged Colloids

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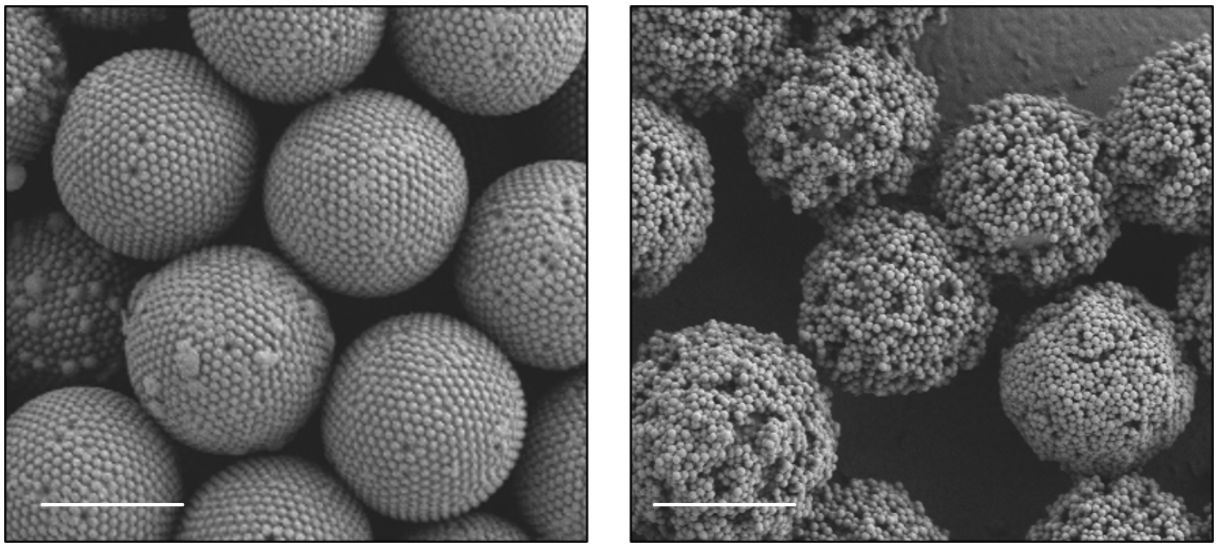


Figure S1: **Control of Layering.** Polymer-adsorption-based heteroaggregation between liquid cores and solid shell particles (satellites) results in a uniformly close-packed monolayer of satellites (*left*). Electrostatic-based heteroaggregation leads to randomly formed multi-layered clusters (*right*). This illustration represents a variation where the shell particles are composed of positively-charged polystyrene (PS) particles. These PS particles are synthesized using the cationic 2,2'-Azobis(2-methylpropionamidine) dihydrochloride (AIBA) initiator, in contrast to the anionic potassium persulfate (KPS) typically used. The measured zeta potential for these positively-charged PS particles is approximately +30 mV. Scale bars: 2 μm.

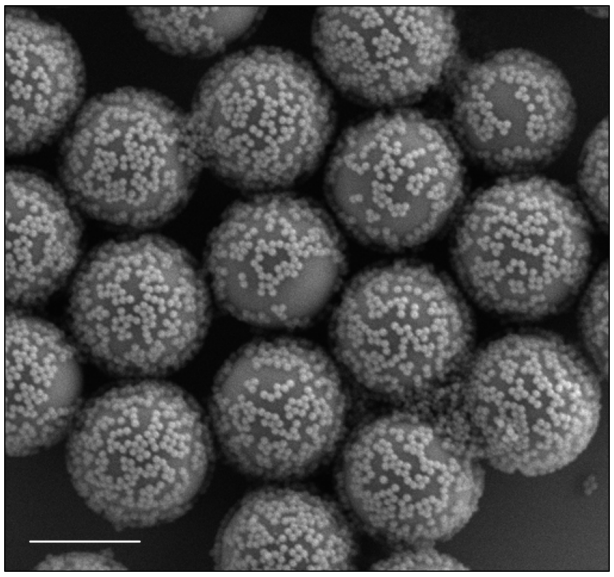


Figure S2: **Polymer-Adsorption on Solid Cores.** F108-grafted solid TPM cores interact with PS resulting in only sparse coating. F108 brushes are chemically locked in place upon polymerization, losing reconfigurability throughout coordination. Insufficient wetting into the liquid core also contributes to satellites detaching during drying. Scale bar: 5 μm.

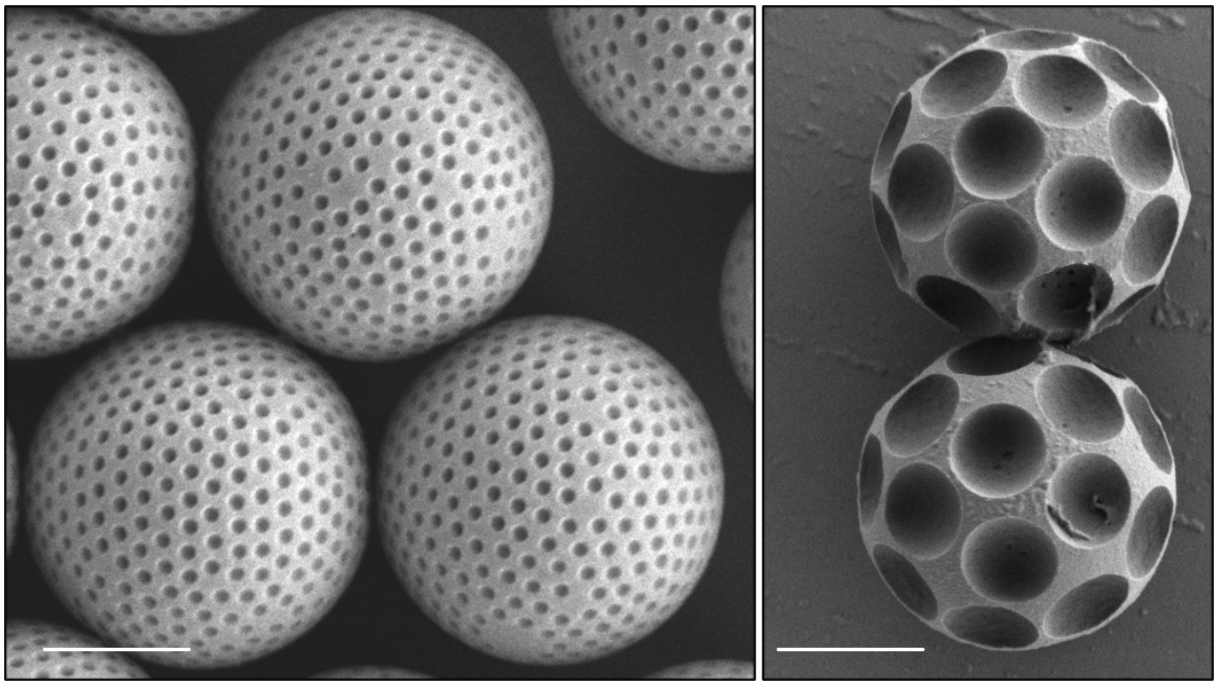


Figure S3: **Dimple Analysis.** We assemble colloidal cages using polymer-mediated bonds, which form when a polymer-coated particle comes into contact with a polymer-free particle. The two species are initially stabilized by electrostatic repulsion, preventing them from making physical contact. By screening the charge, however, the particles reach a proximity where the polymer can form bridges and bind them together. This assembly approach is effective between both solid-solid as well as solid-liquid particles because it doesn't require wetting. In our system, however, wetting occurs after attachment as demonstrated by the imprints left by the PS particles on the polymerized oil. The resulting capillary forces are sufficiently weak to allow for some degree of surface reconfiguration. We hypothesize that the polymer coating and residual electrostatic repulsion between PS particles contribute to mitigating these capillary interactions. Low satellite/core size ratio (*left*) reveals a majority of symmetric six-neighbored close packing, whereas larger size ratios (*right*) result in many distorted or five-neighbored coordinations. Scale bars: 2 μm.

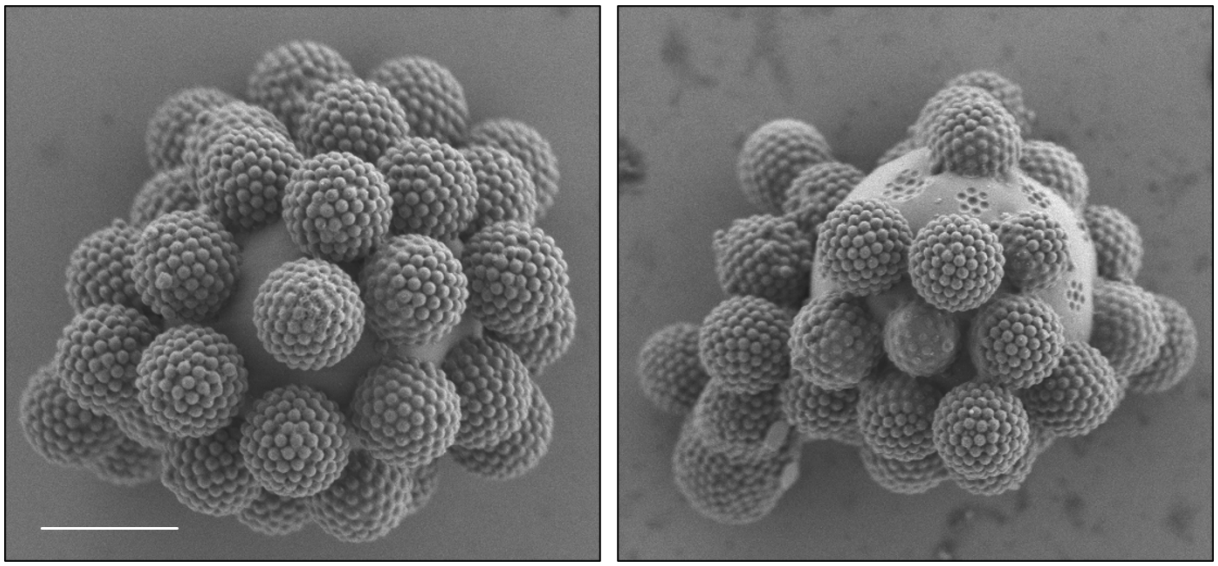


Figure S4: **Cluster of Clusters.** Close-packed clusters can be sequentially employed as new satellites for higher-order coordination. Distinctive flower-patterned dimples indicate the wetting condition for cluster of clusters. Scale bar: 1 μm.

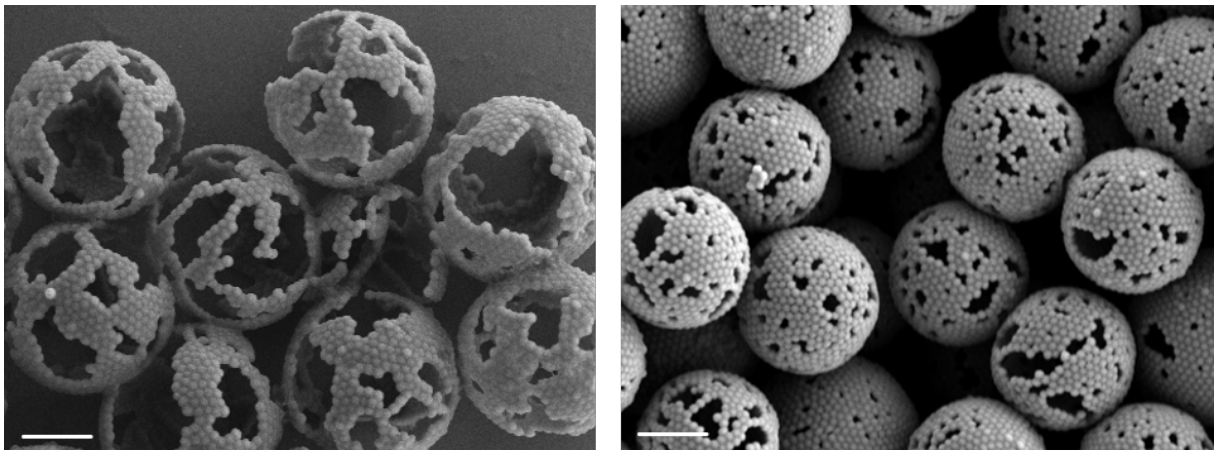


Figure S5: **Tunable Fissures.** TPM core droplets aged for 30 (*left*) and 60 (*right*) minutes demonstrate decreasing capacity for swelling and reduced fissure formation, resulting from increasing degree of network crosslinking. Scale bars: 1 μm.

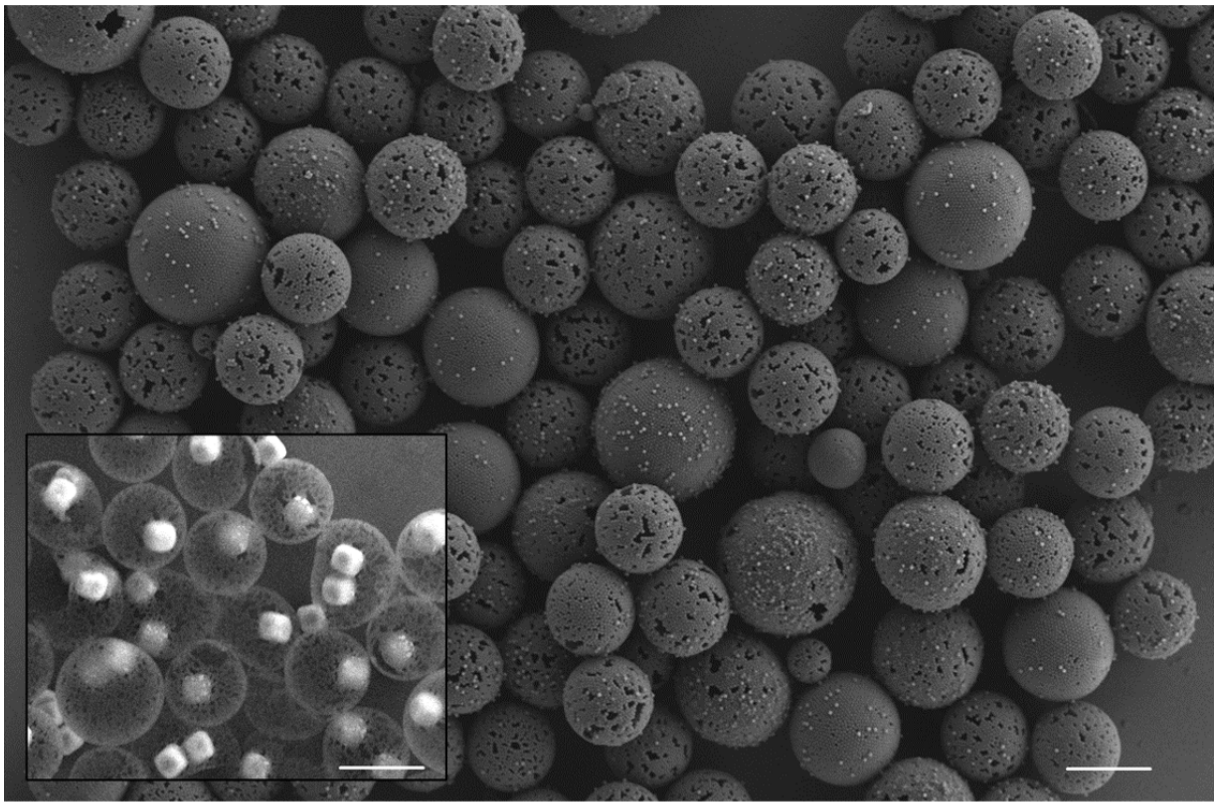


Figure S6: **Scalability.** A wide field of view of a typical cargo-loaded cage sample. Small fissures observed are due to TPM core swelling. Large particles without any apparent defect or fissure are direct outcome of interparticle coalescence and satellite overpacking, which can be determined by the number of cargos inside. The inset shows a high voltage SEM image, highlighting the number of hematite cargos within. Scale bars: 4 μm.

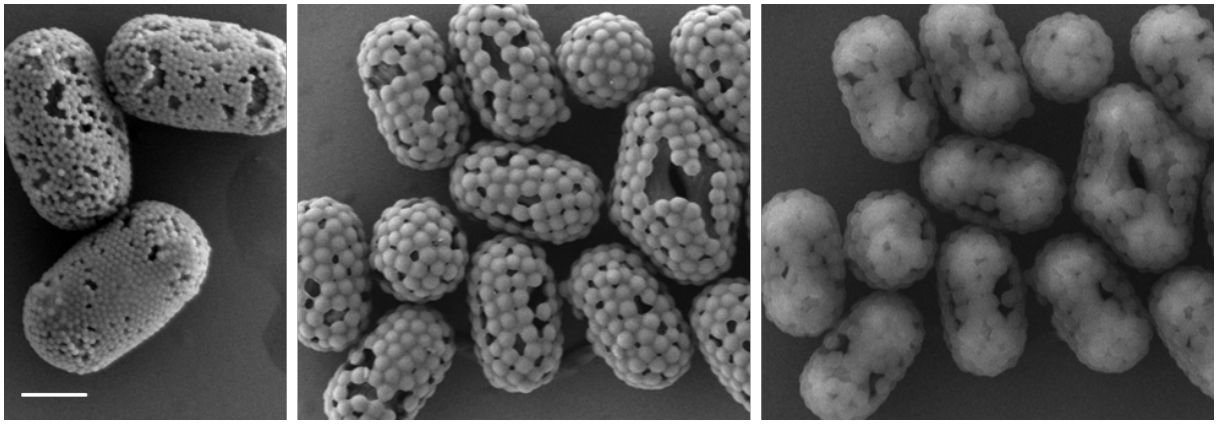


Figure S7: **Ellipsoidosomes.** Ellipsoidal-shaped cages are produced using peanut-shaped hematite cargo and PS sphere satellites (*left* and *middle*). SEM performed under high voltage reveals the orientation of embedded peanut (*right*). Scale bar: 1 μm.

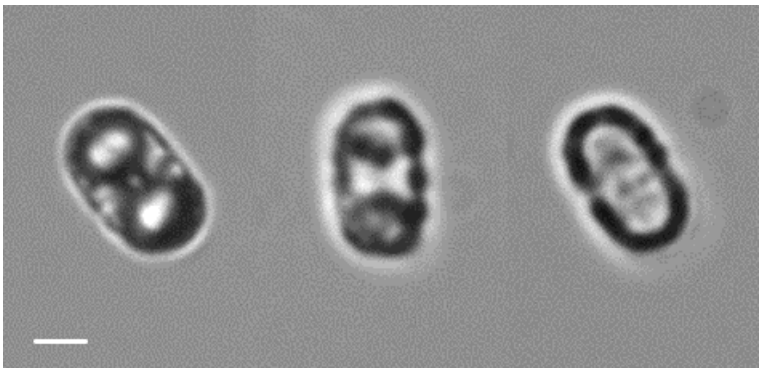


Figure S8: **Etched Delivery of Hematite Peanut.** A hematite peanut, loaded inside an ellipsoidal cage, dissolves preferentially at the thinner belt first, followed by the two ends. The particles undergo etching in 1M HCl. Scale bar: 1 μm.