

This thesis presents an in-depth study on the effect of colloidal particle shape and formation mechanism on self-organization and the final crystal symmetries that can be achieved. It demonstrates how state-of-the-art X-ray diffraction techniques can be used to produce detailed characterizations of colloidal crystal structures prepared using different self-assembly techniques, and how smart systems can be used to investigate defect formation and diffusion in-situ. One of the most remarkable phenomena exhibited by concentrated suspensions of colloidal particles is the spontaneous self-organization into structures with long-range spatial and/or orientational orders. The study also reveals the subtle structural variations that arise by changing the particle shape from spherical to that of a rounded cube. In particular, the roundness of the cube corners, when combined with the self-organization pathway, convective assembly or sedimentation, was shown to influence the final crystal symmetries.

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