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Aperiodic Crystals at the Mesoscale

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Materials with mesoscale structural characteristics have attracted great attention across the fields of chemistry, physics, and materials science. A typical example is mesoporous silica, which are synthesized in water/surfactant/silica systems, and has welldefined mesopores resulting in high surface area. Mesoporous silicas have two defining structural characteristics: (i) disorder at the atomic scale, i.e. only short-range order; and (ii) distinct order at the mesoscale, i.e. long-range order. Atomic-scale structural characterization by common diffraction techniques, such as X-ray single crystal diffraction, is challenging for these partially ordered materials. This is because of the difficulty in obtaining large (> 10 µm) single crystals, and because large-distance periodic features cause diffraction intensities to fall off rapidly with scattering angle, so that only limited small-angle data are available. On the other hand, transmission electron microscopy (TEM) is a powerful tool for structural characterization at the mesoscale level due to the stronger interaction of electrons with matter compared to X-rays, enabling the use of very small crystals. In particular, high-resolution TEM (HRTEM) images give the phase and the amplitude of the crystal structure factors experimentally, leading to a 3D structural model by electron crystallography. Cage-type anionic-surfactant-templated mesoporous silicas display rich structural diversity. Among them, cage-type mesoporous silica with tetrahedrally close-packed (TCP) structures can be described by four types of polyhedra, 5^12, 5^12 6^2, 5^12 6^3, and 5^12 6^4.[1] A variety of structural polymorph have been observed and characterized by TEM. Their structures show a close resemblance to the Frank-Kasper phases, which are well known in intermetallic compounds. We found mesoporous silica with dodecagonal quasicrystalline ordering as one of the TCP structures (Figure).[2] In this presentation, I will discuss structural characterization of aperiodic crystals at the mesoscale, such as mesoporous silicas and binary colloidal crystals, by electron microscopy.

[1] Y. Sakamoto, L. Han, S. Che, et al., Chem. Mater., 2009, 21, 223-229., [2] C. Xiao, N. Fujita, K. Miyasaka, et al., Nature, 2012, 487, 349-353.



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