Novel crystallization mechanic of the zeolite silicalite-1 at high temperature: a study of nuclear spectroscopies

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Abstract: This paper, for the first time, presents on the full and detail mechanics for formation, aggregation, and densification of the amorphous nanoparticles as well as nucleation and crystal growth of the zeolite silicalite-1 in thermal condition at high temperature (170 ^{0}C). By combination of positron lifetime annihilation measurements and related techniques such as XRD, SEM and FTIR, it is the first time we have found that the inorganic-organic composites formed immediately after the initial gel mixture was prepared through fast agglomeration of silicate pieces around cation TPA^+ by van der Waals interaction. In hydrothermal environment, rearrangement of van der Waals interactions occurred to increase volume and structural order of composites before aggregating to inform the primary particles (size ~ 2.1 nm) that continues to construct into the amorphous nanoparticles in size ~ 10 - 50nm. Aggregation of the amorphous nanoparticles occurred, later, to form the particle clusters in size ~ 100 – 300 nm before being densified through narrowing space and distance between the primary and amorphous nanoparticles by addition of monomers from solution to surface of the particles through classical pathway. Nucleation mechanic occurred by rearrangement of the low order silicate molecule and forming sheets of the double five-membered rings through Si-O-Si bonds around a TPA^+ molecular. Densification process promotes connection between sheets of the double five-membered rings to form into the initial pentasil structures of the zeolite silicalite-1. In this mechanic, formation of TPABr clusters created mesopores having radius ~ 6 nm in significant amount. Crystal growth at high temperature occured in two mechanics including growing on surfaces of the nanoparticles and aggregating the particles clusters to inform the poly-crystalline core/single-crystalline shell structure of the zeolite silicalite-1 by the addition of both silica molecules and nanoparticles on their surface (simultaneous mechanics of classical and nonclassical pathways).