

174f Nanotiling: Coatings from Suspensions of Hexagonal Zeolite Particles

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Zeolites are known for their molecular sieving property, which arises from a directional crystalline structure that results in pore/channel networks of molecular dimensions. Not limited to zeolites, these molecular sieve crystals can be grown as thin films for applications in continuous molecular and isomeric membrane separations, which are difficult or expensive with traditional non-continuous methods. Because the crystal structure and molecular sieving property are directional or anisotropic, the film must be uniformly oriented to preserve sieving functionality [1].

Arrays of uniformly oriented crystal nanoparticles arranged in close-packed monolayers [2] (nanotiles) are precursors to such highly uniformly oriented crystalline thin films. The particle geometry is generally anisometric, corresponding to the anisotropy of the crystal structure in such a way that uniform geometrical particle orientation ensures uniform crystallographic orientation. Thus, nanotiled particles, or very small particles uniformly oriented in all three dimensions, can be grown into very thin polycrystalline films with highly uniformly oriented domains.

We are investigating coating processes that result in nanotiled particle deposition. Our approach is based on the convective assembly of hexagonal zeolite nanoplatelets to the contact line between a suspension and substrate driven by evaporation of the volatile component. Micrometric areas of nanotiling, in this case hexagonally close packed monolayers, have been observed in coatings made by the recession of the contact line down the substrate, either forced by withdrawal of the substrate, or resulting from evaporation of the bulk suspension bath. On a larger scale (submillimeter), multilayered deposition stripes are preceded and followed by stripes devoid of any particle deposition. The stripes are parallel to the contact line, and regularly spaced. At sufficiently high suspension concentrations, the stripes can be suppressed, and a continuous uniformly oriented multilayer is obtained. We will present scanning electron microscopy and X-ray diffractometry data supporting preferred orientation and periodic ordering of the zeolite nanotiles, and we will discuss the physical principles that govern the deposition process.

[1] Lai, Z., G. Bonilla, I. Diaz, J.G. Nery, K. Sujaoti, M.A. Amat, E. Kokkoli, O. Terasaki, R.W. Thompson, M. Tsapatsis, and D.G. Vlachos, "Microstructural optimization of a zeolite membrane for organic vapor separation," *Science*, 300, 456 (2003).

[2] Ha, K., Y.J. Lee, H.J. Lee, and K.B. Yoon, "Facile assembly of zeolite monolayers on glass, silica, alumina, and other zeolites using 3-halopropylsilyl reagents as covalent linkers," *Adv. Mater.*, 23, 1114 (2000).