Preparation, Characterization, and Modeling of Nanoporous Silicon Carbide Membranes

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Silicon carbide (SiC) is a promising material for the preparation of high temperature membranes [1-5] due to its many unique properties such as high thermochemical stability, high thermal conductivity and resistance to abrasion. In our studies, SiC microporous membranes are fabricated using two different techniques. The first approach involves the pyrolysis of pre-ceramic polymeric films [3,4], which are coated on tubular SiC macroporous supports using a combination of slip-casting and dip-coating techniques. Combining slip-casting with dip-coating significantly improved the reproducibility in preparing high quality membranes. In addition, a novel method, based on the use of sacrificial interlayers, was also developed for the preparation of these SiC membranes, which involves periodic and alternate coatings of polystyrene sacrificial interlayers and SiC pre-ceramic polymeric layers on the top of slip-casted tubular SiC supports. Membranes prepared by this technique exhibit single gas ideal separation factors of He and hydrogen over Ar in the range of (176-420) and (100-200), respectively, with permeances that are typically two to three times higher than those of SiC membranes prepared previously by the more conventional techniques. The second approach in the preparation of microporous SiC membranes involves chemical-vapor infiltration/chemical-vapor deposition (CVI/CVD) techniques [2]. We have again used macroporous SiC tubes as supports, and a number of SiC CVD precursors. A key aspect of our studies, furthermore, involves the preparation of appropriate macroporous supports. We make use of novel binders which overcome the challenge of the non-homogeneous distribution at the micro-scale one encounters with the current "state-of-the-art" approaches. In our study, we investigate the effect on the final performance of the SiC supports of different parameters such as the starting SiC particle size distribution, the sintering aid content and sintering temperature. Optimized supports show two orders of magnitudes improvement in gas permeance compared to the SiC supports we prepared previously using the traditional approaches [1].

In this talk, we will first describe the techniques that we utilize in our group for the preparation and characterization of such microporous membranes. These involve a variety of surface analytical techniques (SEM/TEM, EDAX, AFM, FTIR, XPS, etc.), TGA, and various chromatographic and mass spectrometric techniques useful for characterizing the membrane structure and its surface and bulk chemical composition. They are coupled to transport and sorption investigations and molecular simulations of such processes. We use equilibrium molecular dynamics (MD) techniques to model the sorption of single gases and mixtures of gases through such membranes, and non-equilibrium MD techniques to study the transport [6,7]. To describe the structure, we use

a novel three-dimensional molecular pore network model of the pore space based on a Voronoi tessellation of the simulation cell. The simulations with such a model allow us to investigate the effect of the morphology of the pore space, i.e., its pore size distribution and pore connectivity on the sorption and transport characteristics. Continuum models coupled with network representations of the membrane are used to simulate the preparation methods for some of these membranes [8-10]. Combining the experimental, network, equilibrium and non-equilibrium molecular dynamics techniques helps to provide unique insight into the key factors of the preparation procedures that determine membrane properties and performance.

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