192c Spatially Resolved Measurements of Transport of Guest Molecules in Nanoporous Molecular Sieve Membranes

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We report spatially resolved, quantitative, non-destructive, in situ, measurements of the transport of organic molecules through a polycrystalline, anisotropic, nanoporous molecular sieve membrane, with micron-scale resolution. Nanoporous and nanocomposite molecular sieving membranes have potential to revolutionize several industrially important separations such as hydrocarbon mixtures, water/alcohol mixtures, and others.[1-4] Methods for studying nanoporous membrane transport phenomena depend heavily on theoretical models (e.g., the Maxwell-Stefan equations), since the transmembrane flux is the only experimental information available.[5, 6] To avoid transport-model-dependent analysis and probe directly and quantitatively the intramembrane transport, we have developed an experimental method based on step scan photoacoustic spectroscopy (SS-PAS) and the requisite data analysis techniques for this purpose.[7, 8] An important application is the transport-model-independent experimental description of membrane transport, by simultaneous measurement of the concentration profile, membrane thickness and membrane flux.

We demonstrate here the steady-state concentration profile of the organic molecules p-xylene and n-hexane during their permeation through an MFI zeolite membrane. In SS-PAS, the use of an infra-red incident beam modulated at a large number of frequencies, in combination with a step scan interferometer and a specially designed sample cell, allows us to extract concentration profiles from the membrane in situ. The in situ photoacoustic sample cell has two separated chambers between a membrane; guest molecules feed with He carrier gas into upper chamber and permeant gases sweep out from bottomed chamber. Depth-sensitive information is obtained by varying the chopping (modulation) frequency. The concentration profile of guest molecules is extracted by combining absorption band intensities of the guest and the zeolite with the theory of photoacoustic signal generation. The guest concentration profiles of p-xylene and n-hexane are obtained in the calcined MFI membrane under steady-state permeation conditions. We demonstrate the ability to measure the concentration profiles under different conditions of feed partial pressures and sweep gas flow rates, so as to directly probe the transport phenomena in the membrane. If coupled with conventional measurements of the transmembrane flux (via gas chromatography or other methods), the transport properties such as the concentration-dependent diffusivity D(C) can be directly obtained from experiments without involvement of any transport models.

It is proposed that this experimental method can allow a powerful new way of studying membrane transport based on detailed experimental knowledge of the concentration profile, the membrane flux, and the membrane thickness. With careful measurements, it will now be possible to validate the large number of theories and simulations for transport in polycrystalline, anisotropic, nanoporous membranes, that previously relied on only limited experimental data. The method is of sufficient generality to be applied to different classes of membranes – e.g., polymeric, mixed matrix, and nanocomposite membranes.

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