High-temperature hydrogen gas permeation properties of precursor-derived microporous amorphous silica-based membranes

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Microporous amorphous silica-based membranes exhibit an excellent high-temperature hydrogen perm-selectivity, which can be achieved by the molecular sieve-like property derived from in-situ formed microporous amorphous silica network having a mean micropore diameter of approximately 0.3 nm. In this paper, recent progress in the synthesis of microporous amorphous silica-based membranes using various chemical processing techniques, their multilayered structures and gas permeation properties will be briefly described. Then, hydrogen gas permeation properties of novel transition metal cation-doped amorphous silica-based membranes will be shown and discussed. The ternary Si-M-O and quaternary Si-M-Al-O (M=Ni, Co) silica-based materials were synthesized by pyrolysis of metal-organic precursors at 873 K in air followed by heat treatment under a hydrogen flow at 773 K. The XRD and TEM/EDS analyses indicated the in-situ formation of transition metal nanocrystallites within the Si-M-O having the M/Si atomic ratio >1/8, while some of the quaternary Si-M-Al-O materials were found to keep amorphous state. The TPR/TPD analysis under the hydrogen/argon atmosphere was performed on the synthesized samples, and the transition metal /amorphous silica nanocomposites showed a typical TPD profile due to the hydrogen desorption from the nanocrystallites below 573 K and that assigned to spillover hydrogen above 673 K, while the amorphous Si-M-Al-O materials showed a unique hydrogen desorption peak at 573 to 773 K, and the hydrogen adsorption/desorption observed in this temperature range was found to be reversible. The Si-M-O/ -alumina layered membrane was successfully fabricated by dip-coating of the metal-organic precursor solution on a mesoporous -alumina layer placed on a macroporous -alumina support followed by the two-step heat treatment. The synthesized membrane exhibited a unique enhanced hydrogen permeance at 573 to 773 K. Further results obtained by the FT-IR measurement using DRIFTS technique, and the simultaneous TPR/TPD and mass spectrometry analyses for the desorbed gas species will be shown and discussed from a viewpoint to develop novel hydrogen-triggered chemical valve silica-based membranes.

Keywords: Metal-organic precursor, transition metal cation, amorphous silica, hydrogen, membrane, hydrogen affinity