# 6–5 Membrane Technique Advancing the IS Process

## —Enhancement of the HI Conversion by Silica-Based Membrane Reactor—

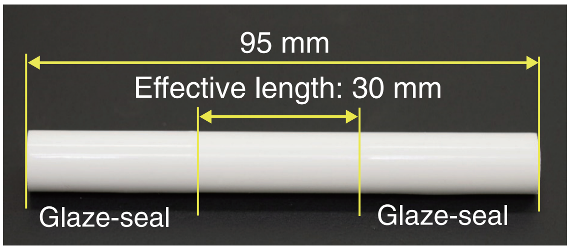


**Fig.6-11 Image of a silica membrane**
Silica membranes were prepared via counter-diffusion chemical vapor deposition on γ-alumina-coated α-alumina support tubes.

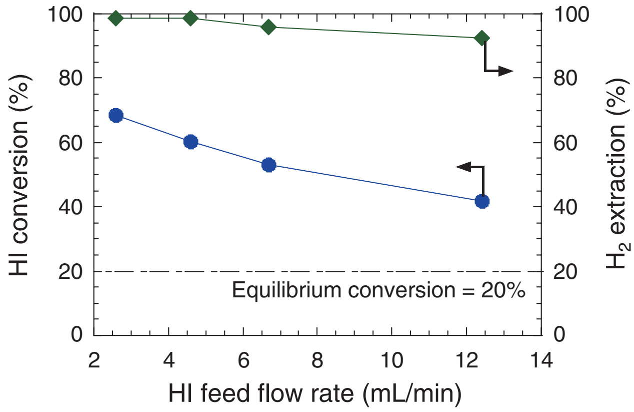


**Fig.6-13 HI decomposition performances of the silica membrane reactor at T = 400 °C and HI feed flow rates of 2.6–12.4 mL/min**
HI conversion greater than 70% was achieved, which is 3.5 times higher than the equilibrium conversion.

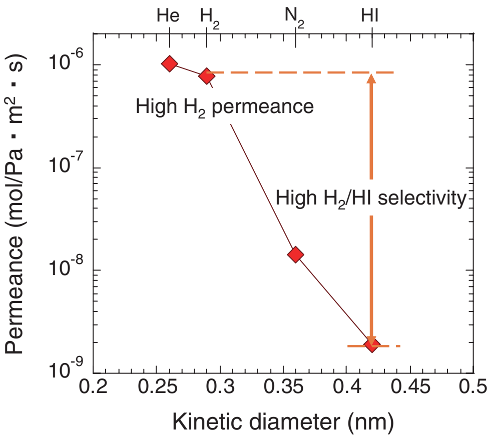


**Fig.6-12 Single-gas permeation performance of the silica membrane**
The prepared silica membrane showed a high $H_2$ permeance of $7.7 \times 10^{-7}$ mol/Pa・m²・s and a high $H_2$/HI selectivity of 403.6.

The thermochemical sulfur-iodine (IS) process is studied by the JAEA to realize massive hydrogen production using nuclear heat at high temperature. This process consists of three coupled chemical reactions:

Bunsen reaction (ca. 100 °C):

$$SO_2 + I_2 + 2H_2O \rightarrow H_2SO_4 + 2HI \quad (1)$$

Sulfuric acid decomposition (ca. 850 °C):

$$H_2SO_4 \rightarrow H_2O + SO_2 + 0.5O_2 \quad (2)$$

Hydrogen iodide decomposition (ca. 400 °C):

$$2HI \rightarrow H_2 + I_2 \quad (3)$$

The HI decomposition (Eq. 3) is a key reaction in the IS process. However, the equilibrium conversion of HI to $H_2$ and $I_2$ is very low (e.g., approximately 20% at 400 °C), leading to an increase in the amount of recycled materials and a decrease in the thermal efficiency of the process. The usage of a membrane reactor that combines a catalytic reaction and $H_2$ separation in one unit can shift the equilibrium and enhance the HI conversion. The membrane reactor demonstration comprises two stages: i) fabrication of a membrane possessing high $H_2$ separation performance and high corrosion stability; and ii) reactor assembly with membrane and catalyst integration.

In this study, silica membranes consisting of a three-layer structure comprising a porous α-alumina ceramic support, an intermediate γ-alumina layer, and a top silica layer that is $H_2$ selective were prepared via the counter-diffusion chemical vapor deposition of hexyltrimethoxysilane (Fig.6-11).

Fig.6-12 presents the He, $H_2$, $N_2$, and HI permeances through the developed silica membranes. The silica membranes exhibited high $H_2$ permeance and high $H_2$/HI selectivity and were stable in the corrosive gas of HI for 11 h. Therefore, the research extended to the assembly of a membrane reactor with the integration of the silica membrane and active carbon catalyst and investigated its potential for $H_2$ production from the HI decomposition.

Fig.6-13 shows the results of the HI decomposition tests varying the HI feed flow rate. The influence of the HI feed flow rate on the membrane reactor performance was significant in the lower range of the flow rate. Moreover, better than 70% conversion of the HI decomposition and 98% $H_2$ extraction were attained. The results suggest that the developed membrane reactor equipped with the silica membrane would reduce the recycled flow rate in the HI decomposition by up to approximately 80% and improve the total thermal efficiency of the IS process by 1%.

We are currently working on the improvement of the thermal efficiency of the IS process by developing a membrane reactor equipped with industrial-size silica membranes in HI decomposition.

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**Reference**

Myagmarjav, O. et al., Comparison of Experimental and Simulation Results on Catalytic HI Decomposition in a Silica-Based Ceramic Membrane Reactor, International Journal of Hydrogen Energy, vol.44, issue 59, 2019, p.30832–30839.