Nitrogen-Selective Metallic Membranes for Post-Combustion Capture

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Abstract

Carbon (CO$_2$) capture represents an important role in the reduction greenhouse gas emissions. Among various CO$_2$ capture technologies currently investigated, post-combustion capture allows for the retrofitting of existing plants and industrial units. Today, the amine scrubbing process is considered the most competitive method for CO$_2$ removal in the flue gases from power plants in comparison to other technologies. Nevertheless, recent work has shown that the energy requirement for solvent recovery could decrease the overall efficiency of the power plants up to 16%. Moreover, additional costs may occur in the solvent absorption technology because of solvent disposal and its continuous replacement due to chemical deterioration.

In contrast, membrane systems usually do not require chemicals or solvents. In addition, membranes offer higher energy efficiencies, greater operational flexibility as well as simplicity of operation and maintenance. Polymeric membrane operations are currently being explored for CO$_2$ capture in power plants. However, some issues still remain regarding the scalability and reliability of the polymeric materials under real operational conditions where the temperature is often too high for polymer stability. Metallic membranes, by contrast, usually require a high temperature for operation and may be more beneficial in saving energy under high temperature conditions. In particular, high temperature N$_2$ removal from coal-fired flue gases located nearby the boiler exit may result in increased concentrations of CO$_2$ and pollutants with a significantly reduced gas volume in the downstream, allowing for traditional emissions controls to perform more efficiently and, consequently, lowering the overall energy consumption and capital and operating costs.

The aim of this work is to explore the potentiality of metallic membranes for N$_2$ separation to enhance the post-combustion capture of CO$_2$. Based on preliminary theoretical investigations using density functional theory, the Group V transition metals (e.g., vanadium (V), niobium (Nb) and tantalum (Ta)) show strong affinity toward N$_2$. Moreover, from solubility and diffusivity values taken from literature, Iron (Fe) seems to be a good fit for this application. Therefore, V, Nb, Ta and Fe foils with a thickness of 40 µm are chosen as membrane materials in this study.

The membrane module used for housing the aforementioned membranes consists of two disc blocks of Hastelloy X, between which a graphite gasket seals the membrane to ensure the separation of the permeate and retentate streams. The entire experimental apparatus consists of mass-flow controllers to regulate the flow rates of the feed gases and sweep-gas, the membrane module containing the membrane, and mass spectrometer for the gas analysis on the permeate stream. The membrane module is heated up with a ramp of 2 °C/min in an Ar atmosphere. The temperature and pressure in the retentate side were varied from 400 to 600 °C and from 2.0 to 60 bar, respectively. A back-pressure regulator placed at the outlet side maintains the retentate pressure while the pressure in the
permeate side was kept constant at ambient pressure. Permeation tests with pure gases (He, N₂, CO₂ and CH₄) are performed to characterize the membranes in terms of N₂ permeating flux and ideal selectivity at different temperature and pressure.

With the instrumental sensitivity considered, the ideal selectivity of nitrogen to methane and nitrogen to carbon dioxide was at least 1,000. Vanadium shows the highest permeability, followed by Ta and Fe at 400 °C and high pressure. Ta results are reported only for 400 °C since it became broken at 500 °C. The permeability did not significantly depend on the pressure but more impacted by temperature. Moreover, scanning electron microscopy, and an electron microprobe analyzer are used to investigate the effect of the different operating conditions on the membrane surface.

From the experimental results, all the membranes have shown infinite selectivity towards N₂ permeation at each operating condition and V showed better performance with respect to the other metal membranes.