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[P2.130] Functional dual-layer ceramic hollow fibre membranes for methane conversion

N.H. Othman*, Z. Wu, K. Li
Imperial College London, UK

Methane is considered as an attractive feedstock for the synthesis of chemical products due to the large amount of natural gas deposits. The process of converting natural gas into a marketable liquid can be carried out using two routes: direct and indirect conversion. The indirect routes involve the conversion of methane into an intermediate, known as syngas (a mixture of CO and H_2). Currently, the syngas is obtained from the methane steam reforming process, which is energy and capital intensive due to high operating temperature and pressure. Partial oxidation of methane (POM) is a promising alternative to the reforming process as it possesses higher energy efficiency and higher selectivity to CO and H_2 [1]. However, as the downstream of the POM process cannot tolerate nitrogen, pure oxygen is required. Pure oxygen is usually obtained from a pressure swing adsorption or cryogenic distillation process that is extremely expensive [2]. Therefore, alternative technologies are required to develop less expensive air separation processes.

The production of syngas through partial oxidation of methane (POM) can be enhanced by using a membrane reactor. In this study, a hollow fibre membrane reactor consisting of two functional layers and integrating oxygen separation (from the ambient air) and methane conversion into a single unit of operation (Figure 1) is developed, which significantly reduces the reactor volume due to the extremely high surface area/volume of the hollow fibre configuration.

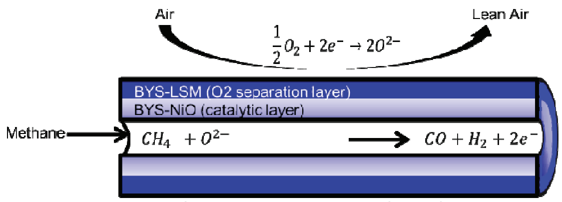

Figure 1. Schematic diagram of membrane reactor for POM

The dual-layer hollow fibre membranes were fabricated via a single-step co-extrusion and co-sintering technique, in which the thickness and the composition of each functional layer can be controlled in order for a better reactor performance. The thin dense outer layer consists of 50% bismuth yttrium samarium oxide (BYS) and 50% lanthanum samarium manganite (LSM), while the inner layer consists of 60% BYS and 40% nickel oxide (NiO). The inner layer not only acts as a support layer but also a catalyst layer for methane conversion. For dual layer ceramic membranes made of different materials, the sintering process is quite challenging due to the difference in sintering behaviour of the membrane materials. BYS was used for both layers to ensure high oxygen permeation and simultaneously acts as a ceramic matrix that contributes to match the sintering behaviour of the two layers. The sintering behaviour of membrane materials was analyzed using Dilatometer prior to the preparation of spinning suspensions.

In order to investigate the performance of BYS as a highly oxygen permeable material, a single layer hollow fibre membrane fabricated from BYS-LSM was prepared first. A series of oxygen permeation were studied between 650 and 950 °C (Figure 2) and the results show that the oxygen permeation is dependent on operating temperatures. The highest oxygen permeation was obtained at 950 °C (11.6 x 10^{-7} mol $cm^{-2}s^{-1}$). Further characterization using X-ray diffractometer (XRD) was carried out to study the changes of the membrane materials. No changes in material structures were observed in XRD patterns, when the membranes were sintered at 1100 °C for 10 hours. This indicates there is no inter-diffusion of BYS and LSM

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throughout the sintering process. A shorter air gap length was used during the fabrication of the single layer hollow fibre membrane due to the low viscosity of the spinning suspension. As a result, a sandwiched structure membrane was obtained (Figure 3). As oxygen permeation has been considered as the limiting step for methane conversion in our previous studies [3, 4], ideally, improving the oxygen permeation will significantly improve the syngas production.

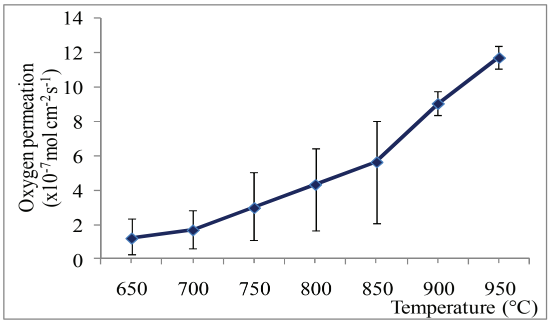

Figure 2. Oxygen permeation of single layer membrane

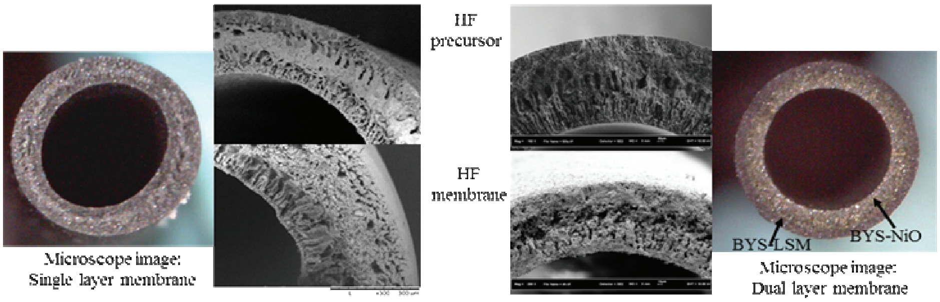

Figure 3. Microscope and SEM images of single and dual layer membrane

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Keywords: Ceramic hollow fibre membrane, Membrane reactor, Partial oxidation of methane (POM), Oxygen permeation