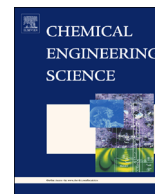




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# The fluid dynamic effect on the driving force for a cobalt oxide silica membrane module at high temperatures

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## HIGHLIGHTS

- The effect of binary gas (H<sub>2</sub>/Ar) composition and fluid dynamics up to 500 °C.
- The H<sub>2</sub> molar fraction was the most influential factor affecting the driving force.
- The decline of the H<sub>2</sub> molar fraction in the feed domain was substantial at 45.3%.
- The decline of the H<sub>2</sub> molar fraction in the permeate domain was marginal by 2.2%.
- Concentration–polarization in both feed and permeate domains was very weak.

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## ABSTRACT

This work presents a computational fluid dynamic (CFD) model to investigate the effect of binary gas (H<sub>2</sub>/Ar) composition and fluid dynamics at high temperatures (from 200 up to 500 °C) for a reasonably sized membrane module containing two cobalt oxide silica membrane tubes in series for H<sub>2</sub> separation. The model provided the local information of velocity, pressure and H<sub>2</sub> fraction for the driving force analysis. The H<sub>2</sub> molar fraction was found to be the most influential factor affecting the driving force, though the total pressure varied slightly along the axial length. In feed domain the H<sub>2</sub> molar fraction showed a clear decline in the axial direction from feed inlet to retentate outlet by 45.34% (for the case of feed fraction 50% H<sub>2</sub> and feed flow rate 100 N ml min<sup>-1</sup>). In permeate domain, H<sub>2</sub> fraction showed the same trend but the decline slope was much less than feed domain being 2.22%. Concentration–polarizations in both feed and permeate domains were very weak with the concentration polarization degree less than 0.4% and can be ignored in cobalt-oxide-silica membrane module. High temperature promoted the performance of pure gas permeation, but had little impact on mixed gas separation as the driving force reduction at higher temperature is more significant.

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## 1. Introduction

Hydrogen is an element of major interest in chemical engineering processes. Hydrogen is extensively used for hydrogenation reactions in the production of diesel (Kan et al., 2012), alkanes (Cordero et al., 2008; Slater, 1964) and Bergius process (Molyneux, 2001), or as a clean energy carrier in fuel cell vehicles (Bahukudumbi and Ford, 2006). Hydrogen can be produced from a variety of sources including fossil fuels, biomass, water and some industrial waste chemicals (McLellan et al., 2005). Many of the conventional hydrogen processes involve fossil fuel as the primary feed stock, such as natural gas

reforming and coal gasification. These processes are characterized by high temperatures, which require cooling down the gas stream for hydrogen separation if traditional gas separation processes are used such as pressure swing adsorption or solvent absorption. Although these separation processes are very effective with a long history of successful application in the chemical engineering industry, cooling down large volumes of gases is a costly exercise with high operating cost implications.

Therefore, separating hydrogen at high temperatures is becoming attractive, as it reduces the energy penalties associated with the cooling requirements encountered with conventional gas separation processes (Bracht et al., 1997; Smart et al., 2010). Among potential novel gas separation technologies, inorganic membrane based metal oxide silica molecular structures have performed well at high temperatures (Miller et al., 2013; Tsuru et al., 2011; Wang and Tsuru, 2011; Yacou et al., 2012), and have

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demonstrated good stability under the exposure of steam (Igi et al., 2008) and hydrogen sulfide (Uhlmann et al., 2011). These membranes have also been deployed in membrane reactor configurations for hydrogen production via the water gas shift (WGS) reaction (Battersby et al., 2010; Brunetti et al., 2007), and dehydrogenation reactions (Battersby et al., 2006). Many of the previous works in this field have primarily focused on materials development and membrane performance using single gas permeation in terms of hydrogen fluxes (e.g. as a production measure) and hydrogen selectivity (e.g. as a measure of product quality). However, industrial gas streams contain multi-component gas mixtures, which cannot be represented by an ideal single gas experimental work. In order to shift the inorganic membranes from a laboratory scale to an industrial scale, it is paramount to develop simulations for large membrane modules and validate against experimental data.

In the case of small membrane modules with high space velocities, the changes in driving forces for gas separation are not significant and analytical solutions are applicable. In the case of large membrane modules, there are major changes in the driving force along the axial direction. As the boundary conditions between the feed interfaces and permeate interfaces are not constant, the mass transfer resistances are significantly affected at the membrane. Therefore, CFD has superior features to describe and model gas transport in both axial and radial directions for the permeate and feed domains.

Experimental and/or numerical simulations of pure gas permeation are often performed to provide an indication of optimized performance of membranes. However, the transport of a component in a gas mixture through the membrane is affected by the presence of other penetrants either due to the interactions among the permeating species or additional resistance in the gas phase (Coroneo et al., 2009a; Krishna, 1990; Krishna and van Baten, 2011a). As a consequence, the preferential permeation of a gas like hydrogen from a multi-component gas mixture is significantly affected resulting in much lower hydrogen fluxes for binary gas mixtures as compared to single gas permeation; as recently demonstrated by Yacou et al. (2012) for multi-tube membrane modules. In view of the multi-component gas effect, several research groups (Burggraaf et al., 1998; Chang and Lee, 2006; Kapteijn, 2006; Keizer et al., 1998; Krishna, 2012; Xiao and Wei, 1992a, 1992b) have developed gas permeation models that specifically deal with selective permeation through membranes of feed gas mixtures. These models have fixed boundary conditions on the feed interface and permeate interface. However, this assumption is not realistic when considering gas separation processes, as the boundary conditions are not constant along the membrane.

In order to better understand mixed gas separation processes, detailed information (e.g. gas concentration at every point) inside a membrane module should be investigated, though it is very difficult to obtain such information through experimental work. To address this problem, computational fluid dynamics (CFD) is a feasible approach to simulate detailed gas flow characteristics at any point of a membrane separation system (Ghidossi et al., 2006; Ji et al., 2012; Wiley and Fletcher, 2002). This has been demonstrated by previous CFD works for palladium (Chen et al., 2011, 2012; Coroneo et al., 2009b), ceramic (Koukou et al., 1996, 1999), polymeric (Kawachale et al., 2010; Sean and Liu, 2004; Siegel, 2008) and silica derived membranes (Abdel-jawad et al., 2007). A general feature of these CFD works is the application for small laboratory scales.

In the case of large scale membrane modules, Ji et al. (2013) established a one-dimensional model to investigate the impact of feed flow rate, feed pressure and module size on hydrogen recovery, hydrogen product purity and hydrogen product yield. This work was validated against a large multi-tube module containing cobalt

oxide silica membranes as reported elsewhere (Yacou et al., 2012). However, this model considered an almost constant radial gas concentration which simplified calculations and computational time. Questions still remain on the effect of the radial gas concentration together with the axial gas concentration, thus necessitating the development of a two-dimensional model to better describe the effect of gas dispersion in membrane modules for gas separation. In this work, we extend the one-dimensional to two-dimensional model as a more realistic CFD approach for the simulation of hydrogen separation in binary gas feed stream at high temperatures. The two-dimensional model is validated against a cobalt oxide silica membrane module. Further, the two-dimensional model is used to carry out simulations to investigate the gas concentration distribution in both radial and axial directions. Ultimately, this leads to the determination of the driving force distribution also in both axial and radial directions, thus explaining the phenomena observed for gas mixture separation at varying high temperatures.

## 2. Modeling details

The gas separation module depicted in Fig. 1 consists of two parts named as feed domain and permeate domain. Two cobalt silica membranes with length 160 mm each are fixed in series inside the module. Gas permeates from the outside of the membranes to the inside. The two virtual cross sections are located at the middle of each membrane which is used to display the radial profile of H<sub>2</sub> fraction. All other geometrical parameters are also displayed in Fig. 1. In the gas separation process, a multi-component feed gas is introduced from the inlet to the feed domain. The preferentially permeable gas molecules diffuse across the membrane, though not totally as a fraction diffuses along the axis towards the permeate stream, at the permeate outlet. The impermeable gas still flows from feed domain to the retentate stream and finally exits at the retentate outlet. The gases in both domains flow in the same direction as the cocurrent configuration. Binary gas mixtures consisting of H<sub>2</sub> and Ar were fed to the system at different temperatures using cobalt oxide silica (CoO<sub>x</sub>Si) membranes.

### 2.1. The CFD model

In order to properly model the membrane separation performance, fluid dynamics was considered since pressure, velocity and concentration fields have significant influences on the separation process. In hydrogen gas separation with membranes, the feed flow rate is usually much slower than other gas separation processes, as a consideration to maximize hydrogen recovery. The fluid flow in the membranes is generally featured by laminar flow as the Reynolds number is always below 10. This fluid flow phenomenon can be described by a cylindrical two-dimensional and steady state model, which is the basis of this work.

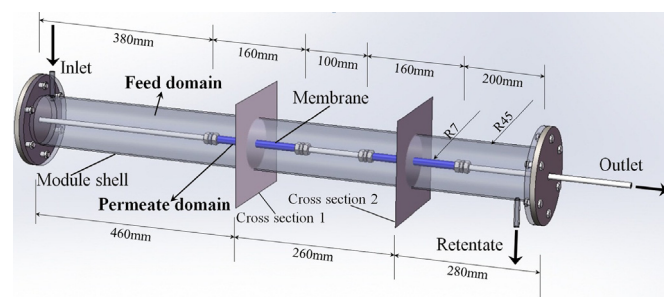


Fig. 1. The structure of the membrane separation module.

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