

Influence of coal power plant exhaust gas on the structure and performance of ceramic nanostructured gas separation membranes



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ABSTRACT

In this work, we investigate the effect of coal power plant exhaust gas on amino-modified mesoporous ceramic membranes. The testing of ceramic membranes in the flue gas of coal-fired power plants represents a new approach, as testing under simulated flue gas conditions has already been undertaken, but not yet during direct exposure to exhaust gas. Flue gas exposure trials were carried out at a lignite-fueled power plant and a hard-coal-fueled power plant. Most experiments were conducted using a test rig designed to bring planar membrane samples in direct contact with unconditioned flue gas in the exhaust gas channel. Another test rig was designed to test membrane modules with pre-treated flue gas. The tested membranes had an asymmetrical structure consisting of a macroporous α -Al₂O₃ support coated with a mesoporous γ -Al₂O₃ or 8YSZ interlayer. The microporous functional top layer was made of amino-functionalized silica. The tests revealed different degradation mechanisms such as gypsum/fly ash deposition on the membrane surface, pore blocking by water condensation, chemical reactions and phase transformation. A detailed analysis was carried out to evaluate their impact on the membrane in order to assess membrane stability under real conditions. The suitability of these membranes for this application is critically discussed and an improved mode of membrane operation is proposed.

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1. Introduction

The retrofitting of carbon capture and storage (CCS) as well as carbon capture and utilization (CCU) technologies in existing coal-fueled power plants is currently under investigation aiming on reduction of CO₂ emissions. CO₂ separation by membrane technology is an attractive method of capture especially if partial separation of CO₂ from the exhaust gas is aspired (Zhao et al., 2008) or if turndown operation of the power plant is planned. The advantage of membrane technologies is that they achieve stationary operating conditions in short time periods which is ideal for handling the large variation of flue gas volume during turndown operation. Furthermore, easy scalability of membrane modules allows exact adjustment of the carbon capture plant to customer needs, which is attractive especially in the case of integrating CCU technologies in coal-fueled power plants. CO₂/N₂ selectivity of membranes is usually limited, therefore

CO₂ concentration in the permeate of single-stage membrane reactors will be lower than in the case of chemical adsorption, but this can be overcome by introducing multi-stage membrane concepts (Zhao et al., 2012). In multi-stage operation, CO₂ concentrations of more than 95 vol.% in the permeate become realistic.

Investigation of membrane technologies and materials for carbon capture from the flue gas of post-combustion power plants is already in progress. First promising results with polymer membranes have been reported in the European project NanoGloWa, in the work of Membrane Technology and Research Inc. (MTR, USA), which was partly funded by the Department of Energy (DOE, USA), as well as in the German METPORE II project funded by the German Federal Ministry for Economic Affairs and Energy (BMWi, Germany).

In the NanoGloWa project, Sandru et al. (2013) reported the successful operation for several months of a 1.5 m² membrane module equipped with facilitated transport membranes in a hard-coal-fueled power plant. The membrane performance was characterized by a CO₂/N₂ selectivity of 80 at moderate permeance of 0.6 Nm³/(m² h bar).

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MTR deals with carbon capture from the exhaust gases of gas-fueled industrial production processes, coal-fueled power plants or from syngas containing hydrogen and CO₂ (Liu et al., 2013; Brinkmann et al., 2013). All of these activities are accompanied by related process simulation aiming at optimum process integration of membrane technology while in parallel reducing the energy demand to a minimum. MTR operates a carbon capture pilot plant with a capture capacity of 1 t CO₂ per day in the National Carbon Capture Center (NCCC, Wilsonville, USA). A stable performance of spiral wound polymer modules was demonstrated for several months characterized by an almost constant CO₂ concentration of around 60 vol.% in the permeate and around 3 vol.% in the retentate. In sum, 85% of the CO₂ was captured from the exhaust gas.

In the German METPORE II project, polymer membranes (PolyActive™) – developed by our project partner Helmholtz-Zentrum Geesthacht (HZG, Germany) – were recently tested with exhaust gases. If a suitable pre-treatment of the exhaust gases was applied, membrane stability for up to 2250 h was demonstrated in the lignite-fueled power plant using a small-scale membrane test rig (membrane area approximately 1×10^{-3} m²). In a hard-coal-fueled power plant, HZG successfully operated a pilot plant equipped with a 12 m² membrane module for up to 743 h achieving a CO₂ concentration in the permeate in the range of 60–70 vol.%. Currently, these results are being prepared for publication. A more detailed description of the characteristic properties of the polymer membrane module used in the pilot plant can be found elsewhere (Brinkmann et al., 2013; Pohlmann and Brinkmann, 2014).

Alternatively, CO₂-selective ceramic membranes are regarded as promising candidates for carbon capture from exhaust gases. Due to their inorganic chemical bonding, it is expected that they will show clearly improved long-term stability in the highly corrosive environment of a coal-fueled power plant characterized by acidic conditions, deposition of fly ash and huge amounts of water vapor at slightly increased temperatures (60–70 °C). If ceramic membranes are able to withstand these conditions, the effort of pre-treating the exhaust gas as required for operation of polymer membranes (Pohlmann and Brinkmann, 2014) could be significantly reduced. Up to now, CO₂-selective ceramic membranes have only been successfully tested under simulated flue gas conditions mainly using binary CO₂/N₂ gas mixtures (Xomeritakis, 2005; Xomeritakis et al., 2009; Sakamoto et al., 2007; McCool and DeSisto, 2005), but little is known so far regarding their functionality and stability when in direct contact with real flue gases.

To shed light on this question, in the present work the function and stability of amine-modified silica membranes were tested for 100 h and 1000 h in direct contact with the flue gas of a lignite-fueled power plant in Niederaußem, Germany (operated by RWE) and a hard-coal-fueled power plant in Karlsruhe, Germany (Rheinhafendampfkraftwerk, operated by EnBW). The amine-modified silica membranes were supported by a pore-graduated ceramic substrate consisting of a macroporous γ -alumina support and a mesoporous interlayer made of γ -Al₂O₃ or 8YSZ (Czyperek et al., 2010; Van Gestel et al., 2010, 2012). The membrane tests in the power plants are based on preliminary studies which were published recently (Van Gestel et al., 2010, 2012).

2. Experimental

2.1. Membrane and membrane module preparation

To achieve a high gas flux as well as a high selectivity, ceramic membranes have an asymmetric design. This means that they have

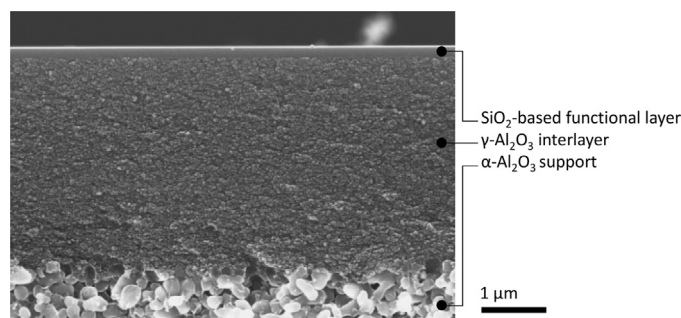


Fig. 1. Cross-fracture SEM micrograph of a planar gas separation membrane.

a macroporous support for mechanical strength, one or more mesoporous interlayers to gradually decrease the pore size and finally a functional top layer. A representative cross section of the multilayer structure of the membranes investigated in this study is shown in Fig. 1. All membranes tested were prepared in-house. Disk-shaped macroporous α -Al₂O₃ supports were prepared starting from a commercial powder (AKP-30, Sumitomo Chemicals, Japan) using a vacuum slip-casting process. After firing at 1100 °C and final machining, the supports had a diameter of 39 mm and a thickness of 2.5 mm. Two types of interlayers, γ -Al₂O₃ and 8YSZ, were tested. Mesoporous γ -Al₂O₃ membrane layers were made by dip-coating a colloidal sol and subsequent heat treatment at 600 °C, following the well-known method described Lin and Burggraaf (Lin et al., 1991; Lin and Burggraaf, 1991). Mesoporous 8YSZ membrane layers were made by subsequent dip-coating of a nanodispersion and a colloidal sol followed by heat treatment at 500 °C, based on a process developed by Van Gestel et al. (2012). Two types of microporous silica top layers were tested, a CO₂-selective amino-functionalized silica layer and – as a reference – a pure silica layer. Functionalization was done in order to enhance the CO₂ affinity of the membranes (Xomeritakis, 2005; Xomeritakis et al., 2009; Sakamoto et al., 2007; McCool and DeSisto, 2005; Leal et al., 1992, 1995). Pure silica layers were prepared by dip-coating a polymeric sol and subsequent heat treatment at 500 °C on the basis of the well-known process described, e.g. by several authors (Uhlhorn et al., 1992; de Lange et al., 1995; de Vos and Verweij, 1998). Functionalized silica layers were prepared in a two-step approach: first the preparation of a surfactant-templated porous silica layer followed by a post-synthesis grafting of amino functional alkoxysilanes on pore walls. Preparation of surfactant-templated membranes was achieved following the description provided by Besson et al. (2000), using hexadecyltrimethylammonium bromide (Sigma-Aldrich, $\geq 99\%$) as the surfactant. Thermal decomposition of the tenside was achieved at 400 °C. Functionalization of the membranes was carried out in the next step following the recommendations of Zhu (Zhu et al., 2011). The membranes were vacuum infiltrated with a 0.064 M solution of ((3-aminopropyl)triethoxysilane, Sigma-Aldrich, 99%) in 1-propanol (Sigma-Aldrich, anhydrous, 99.7%) and were afterwards kept at 80 °C for 5 h. After thorough rinsing with 1-propanol, the membranes were dried overnight at 150 °C.

Additionally, the processing of functionalized silica layers as described above was transferred to ceramic tubular supports. Coating was applied inside the tube. The inner surface was already modified by the manufacturer with a mesoporous γ -Al₂O₃ interlayer. The length of the tubes was 300 mm, the external diameter 10 mm and the wall thickness 1 mm. After processing, ten tubes with a total active membrane surface of approx. 500 cm² were assembled in a steel membrane module. The ceramic tubes and module were provided by Atech Innovations GmbH, Gladbeck, Germany. Both, planar and tubular supports are shown in Fig. 2.

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