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Experimental study of water recovery from flue gas using hollow micro–nano porous ceramic composite membranes

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ABSTRACT

A new method of water recovery from flue gas with hollow micro–nano porous ceramic composite membranes has been put forward in order to deal with the evaporated wastewater from thermal power plants. The feasibility of this method has been investigated in theory, and ceramic membranes of different pore sizes in selective layer have been prepared for experiments. The achieved results indicate that the ceramic membrane with selective layer of 20 nm pore size is appropriate for different flue gas conditions. The amount of recovered water augments with the increasing relative humidity of gas. When the temperature of flue gas reaches 70 °C, the amount of recovered water is above 1 L/(m^2 h) and the recovery ratio can be up to 55%. This method has great potentiality in the application of water recovery from flue gas in thermal power plants.

Introduction

It is estimated that the water vapor content of flue gas from coal-fired boilers is 4%–13%, and for gas-fired boilers the content can up to about 20%. In China, according to the statistical data in 2013 [1], the total amount of water vapor in flue gas emissions is about 1.01 billion tons per year, whereas the amount of water required in thermal power plants is about 8.44 billion tons per year and the wastewater amount is 392.55 million tons per year. Thus, it can be seen that a significant amount of water is discharged into the atmosphere along with flue gas. If this part of water resource can be recovered effectively and be available for power plants, the annual energy-saving benefit is considerable.

A lot of research has been carried on to reduce the temperature of flue gas below dew point and recover waste heat and water by the means of heat exchangers [2–6]. Meanwhile, as a new "green technology", membrane separation method is getting more and more attention in the field of dehumidification and water recovery [7–12]. Dehydration of natural gas is a main important study field, and it has been used in commercial application [13,14]. In the study of dehydration of flue gas, Sijbesma et al. [15] investigated two promising membrane materials (SPEEK and PEBAX® 1074) for flue gas dehydration in power plants and an average condensed water vapor flux of 0.2–0.46 kg/(m^2 h) was obtained with using SPEEK. However, it is a very low flux of water recovery and the inferior mechanical strength of prepared membranes is an important problem need to be solved in actual application. Wang et al. [16] developed the nanoporous ceramic membrane technology for waste heat and water recovery. A 40% recovery ratio of the exhaust water vapor has been achieved. Bao and Wang [17] and Wang [18,19] conducted a lot of tests with using ceramic membranes (pore size 15 nm) and result showed that high moisture content flue gas could provide high vapor transport driving force and create a higher water transfer rate. In the United States, Gas Technology Institute had developed a transport membrane condenser for recovering both energy and water from the low-grade waste heat streams [20], and the nanoporous ceramic membrane could recover water of 3.2–4.5 kg/m^2h when the temperature range of flue gas was 60–90 °C. Macedonio et al. [21] investigated a membrane condenser with microporous hydrophobic PVDF (Polyvinylidene Fluoride) hollow fibers. Simulated and experimental results achieved a 20% water recovery efficiency, meanwhile the temperature reduced about 50 °C when the temperature of flue gas was 50–90 °C and the RH was 90–100%. In addition, they proposed and compared three different possible membrane condenser configurations in terms of amount of recovered liquid water and energy consumption [22]. Wang et al. [23] also used a nanoporous membrane tube bundle to recover both water vapor and latent heat from the flue gas. In their study, the water and heat recovery efficiency reached 20–60% and 33–85%, respectively. Zhao

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Nomenclature

A	Superficial area of selective layer (m^2)
M	Relative molecular mass of liquid
p	Plane saturated vapor pressure (Pa)
p_r	Saturated vapor pressure in the capillary (Pa)
q	Amount of recovered water (L/m^2 h)
r	Pore size (m)
R	Universal gas constant
RH	Relative humidity (%)
T	Temperature (K)
V	Volume of recovered water (L)
ρ	Density of liquid (kg/m^3)
σ	Surface tension of liquid (N/m)
τ	Time (h)
φ	Contact angle

et al. investigated a tubular ceramic membrane as the condenser for simultaneous heat and water recovery from flue gas [24]. Results showed that water and heat transfer efficiencies and recoveries declined due to the reduced residence time when the gas flow rate increased. Increasing the temperature of the inlet gas could enhance water and heat fluxes and recoveries, but did not improve the overall heat transfer efficiency. Hu et al. [25] carried out condensation experiments to investigate the simultaneous heat and mass transfer across the nanoporous ceramic membranes that were treated to be hydrophilic and hydrophobic surfaces using the semicontinuous supercritical reactions. Experimental results showed that the hydrophilic membrane exhibited higher performances of condensation heat transfer and condensate recovery. However, the hydrophobic modification resulted in remarkable degradation of heat and condensate recovery from the mixture. Lin et al. [26] carried out a numerical study to investigate the heat and mass transfer characteristics of a condensing combustion flue gas in a cross-flow membrane tube bundle. The flue gas consisted of one condensable water vapor and three noncondensable gases (CO_2, O_2, and N_2), and the flue gas inlet temperature is within the range of 60–88 °C. Result showed that the heat and mass depletion levels decreased with the increase of the flue gas Reynolds numbers. Chen et al. [27] and Zhou et al. [28] presented the application of a 20 nm pore-sized porous ceramic membrane for condensation heat transfer in artificial flue gas. Results indicated that the water recovery rate could reach above 20% with the help of capillary condensation when the cooling water temperature was higher than the feed gas dew point. A modified heat and mass transfer model for calculating the wall temperatures of the membrane showed good agreement with experimental results.

It can be concluded that different membranes have been prepared to be studied and used for different gas dehydration. Research shows the permeation flux of organic membrane is lower than the one of inorganic membrane [29–32]. Thus, the inorganic membrane can be a potential alternative for processing large flux gas in power plants. Inorganic membranes, porous and nonporous, possess a large number of advantages in engineering application, such as corrosion resistance, high mechanical strength and long service life, while most organic membranes are inferior to them [33–35]. As a kind of inorganic membrane, the porous ceramic membrane shows high stability in both high temperature and acid/alkali environment or microbial erosion environment [36]. This advantage makes ceramic membranes to be applicable to work perfectly in flue gas. Porous media transmission mainly depends on three kinds of mechanisms: gas diffusion, surface flow and capillary condensation [7]. The primary purpose is to effectively separate water vapor and other gas components of flue gas. Capillary condensation mechanism shows that the vapor separation characteristics of porous membrane can be greatly improved and the transport of the non-condensable gas can be prevented [37]. Until now, research of capillary condensation is not enough, and there is hardly any research about the permeation mechanism of micro–nano porous ceramic membrane for water recovery. For this technology, much attention should be paid to the undesirable condensation on the gas side or within the membrane pores [38,39].

This work is aim to use membranes to recycle water from the low-temperature saturated flue gas after FGD, and make the recovered water to be boiler make-up water in power plants. In the present work, based on the mixed nitrogen and water vapor, ceramic membranes of different pore sizes in selective layer are prepared for experiments and analyses. With the help of Kelvin equation, theoretical study illustrates the feasibility of water recovery from flue gas by using hollow micro–nano porous ceramic membrane. For membranes, the recycle ratio is an important parameter to be considered in the industrial design process since it affects the gas processing cost significantly [40]. Thus, it is necessary to study the transport flux of membrane and water recovery ratio under different operating conditions.

Ceramic membrane and transport mechanisms

Multilayer ceramic membrane structure

Hollow porous ceramic composite membranes are manufactured after being sintered several times, which consist of three layers (selective layer, intermediate layer and substrate) according to the pore size, as schematically shown in Fig. 1. According to the different locations of selective layer, the ceramic composite membrane can be divided into two categories: inner side coating and outer side coating. Membrane of inner side coating means the innermost layer is the selective layer and it can be shaped into the multi-channel membrane tube. The outer side coating indicates the selective layer of membrane is located on the outermost layer, and it is often the single channel membrane. Hollow micro–nano porous ceramic composite membranes used in this study refer to single-channel multilayer ceramic membranes. The pore sizes of selective layer and substrate are at the nanoscale and the microscale, respectively.

Structural characterization

In Figs. 2 and 3, the scanning electron microscopy (SEM) pictures of hollow micro–nano porous ceramic composite membranes are taken by JSM6490LV SEM which is produced by JEOL Electronics Corporation. The structures of ceramic membranes with four different pore sizes are investigated.

As shown in Fig. 2, the inner structures of ceramic membranes sintered from particulate matters are compact and the surfaces are smooth. There is a certain relationship between the pore size and

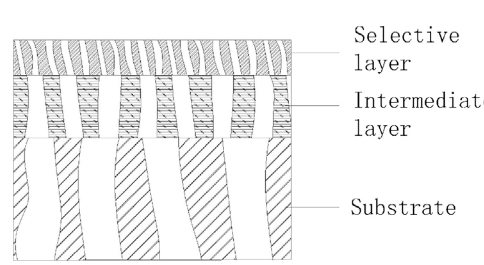

Fig. 1. Multilayer ceramic membrane structure.

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