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# Performance Analysis of Membrane Separation Process for Water and Heat Recovery from Flue Gas

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Membrane separation process can simultaneously recover water vapour and waste heat from flue gases, which has great potential for water and energy savings. Composite ceramic nanoporous membrane is one of the emerging technologies showing the good performance of water vapour and waste heat recovery from the flue gas of power plants. However, the flue gas has great property changes along a flue resulting in a dramatic performance change of membrane separation. Current experimental studies couldn't provide enough experience to explore the performance of membrane modules in a wide range of working conditions and structure configuration. In this paper, a calculation model is established based on thermodynamic mechanisms of membrane separation process to simulate the performance of ceramic membrane modules under different structure configuration and working conditions. Three cases are conducted to analyse the effects of module configuration and parameter selection in a ceramic membrane module. Results show that a ceramic membrane module placed before the desulphurisation tower can recover more heat, while after the desulphurisation tower it can recover more water. There would be an optimal tube diameter and an optimal tube spacing for water recovery under specific working conditions. These results show great adaptability of the calculation model and could guide the system design of membrane separation modules in water vapour and waste heat recovery.

## 1. Introduction

Zero-water-discharge target is an important trend in the development of coal-fired power plants, especially in water-scarce areas. For a typical power plant, flue gas contains a large amount of water vapour and low-temperature heat, which has a huge and significant recovery potential. A coal-fired power plant can be self-sufficient in water consumption if the water recovery rate of flue gas reaches 20 % (Judd and Jefferson, 2003). Membrane separation has been widely used in filtration, purification and other fields. Benefitted by the separation mechanisms, membrane separation technology could provide cleaner and more energy-efficient water compared with several typical methods like cooling condensation and liquid / solid adsorption. Recently, there are two kinds of membranes with good performance to recover water vapour and low-temperature heat from flue gases. One is the non-porous membranes. Sijbesma et al. (2018) proposed a membrane system to remove water vapour from flue gasses using composite hollow fibre membranes with a top layer of sulfonated polyetheretherketone (SPEEK). This kind of membrane system has high selectivity of water, but the mass flux is small and easy to scale. The other one is the porous membrane. Gas Technology Institute used nanoporous ceramic separation membranes to extract water vapour and latent heat from flue gases (Wang et al., 2012). The ceramic membrane has larger diameter, so it has higher mechanical strength and less solid fouling. Compared with the non-porous membranes, the capillary condensation mechanism of porous ceramic membranes enables the membrane module to reach a high recovery efficiency of water vapour and heat from flue gases, and the quality of recovered water is good enough to be directly used for industrial water supplement. At present, most research focuses on analysing experimental data of water vapour and low-temperature heat recovery to explore the characteristics of membrane separation. Chen et al. (2018) conducted a series of experiments with different pore sizes of ceramic membranes under different flue gas conditions and their results show that a 20 nm ceramic membrane could perform a 55 % water recovery rate and is most suitable for power plant flue gas moisture recovery. But a more critical field is to illustrate how parameters of a membrane module

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affect the performance of water vapour and latent heat recovery according to the existing knowledge of membrane separation, and to guide the parameter design for industrial application. Harasek et al. (2016) applied Computational Fluid Dynamics (CFD) to simulate detailed membrane processes of a hollow fibre membrane module and investigated the effect of geometric parameters on purification performance. Soleimanikutanaei et al. (2018) use a CFD model to numerically calculate the performance of a ceramic membrane module under typical working conditions and find out the optimal tube spacing for arrangement. However, different membrane materials vary in water permeability. Various structure parameters of membrane modules (like pipe diameter, tube spacing, channel length, etc.) would also lead to significant changes in performance. The operating conditions are quite extensive in power plants to simulate and test. It is still a challenge to understand the water recovery and heat recovery characteristics of a membrane separation module under different module structure and various working conditions to design an optimal module.
In this paper, a general model is proposed to calculate the water vapour and heat recovery performance of membrane separation modules considering both structure configuration and working conditions. Then, three important parameter combinations about arrangement and structure are selected for case studies to provide suggestions of membrane module configuration.

## 2. Methodology

### 2.1 Mechanisms

There are different mechanisms of water permeation through membranes. For the porous membrane, the permeability coefficient is closely related to the Knudsen number (Alkhudhiri et al., 2012), which is the ratio of mean free path of transported molecules to mean membrane pore size. For Kn > 1, it is under Knudsen diffusion condition. For Kn < 0.01 it is under ordinary molecular diffusion condition. In the transition region, the capillary condensation phenomenon would occur and water vapour would condense into the pores of membrane tubes, which can enhance water permeability (Kim et al., 2018). The membrane pore size is often designed to meet the requirement of capillary condensation for water vapour recovery (Yang et al., 2019). Although many efforts are made on the research of capillary condensation, the theoretical formula for permeability calculation is not accurate enough until it is modified by experiments (Barsotti et al., 2016).
A typical membrane module for water vapour and heat recovery in flue gas has a set of membrane tubes in a staggered arrangement. Cooling water flows inside membrane tubes, and flue gas flows over the outer surface of membrane tubes. The cooling water and flue gas are in a counter-current arrangement along the direction of flue gas to get a better mass and heat exchange. The temperature of cooling water is lower than that of flue gas, and the water vapour in flue gas would condense onto the outer surface of tubes. Then the condensate would enter the nonporous or microporous membranes and get mixed with cooling water. Flue gas could exchange heat with cooling water in membrane tubes, and the latent heat released by the condensation of water vapour would also be absorbed by the cooling water.

### 2.2 Model Description

General representation about the model for heat and mass transfer performance calculation is shown in Figure 1. Flue gases are divided into two parts: one is the water vapour to be condensed, and the other is the rest non-condensed flue gas.

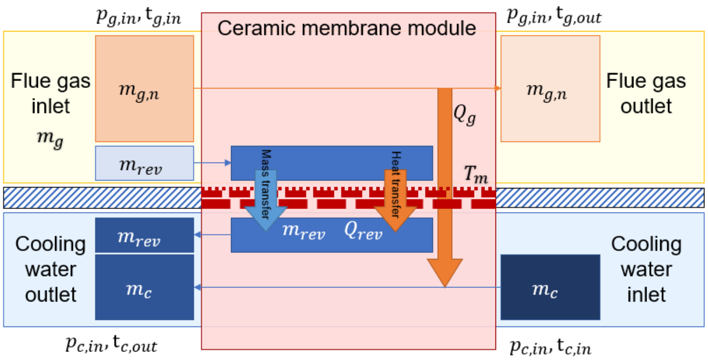


*Figure 1: Schematic diagram of the calculation model for heat and mass transfer*

The mass flux through membrane tubes is assumed to be proportional to the vapour pressure difference across the membrane tubes, which is given by Eq(1).

The mass transfer driving force cannot be infinitely large. So, there is a maximum limit and it varies with different species of membrane material, given by Eq(2)

$$J = C_m(p_g - p_c) \tag{1}$$

$$p_g - p_c \leq p_{max} \tag{2}$$

where $J$, $C_m$, $p_g$, $p_c$ and $p_{max}$ are mass flux through membrane tubes, membrane permeability coefficient, water vapour partial pressure of flue gas, water vapour partial pressure of cooling water, maximum limit of mass transfer driving force.
The membrane filling rate $\phi$ represents the ratio of the membrane area to the module volume. It is derived from the structure of the module as Eq(3)

$$\phi = \frac{\pi D}{2S_1S_2} \tag{3}$$

where $D$, $S_1$ and $S_2$ are outer tube diameter, lateral tube spacing, and longitudinal tube spacing.
The amount of recovered water through tubes $m_{rec}$ is calculated by the integration of mass flux along the flue and given by Eq(4)

$$m_{rec} = \int JdS = \int_L C_m(p_g - p_c)\phi WHdl \tag{4}$$

where $W$, $H$ and $L$ is, the width, height and length of the membrane module.
The heat transfer between the flue gas and the cooling water is regarded as the sum of convection heat transfer from non-condensed flue gas and condensation heat transfer from water vapour. And these heats are absorbed by cooling water. They are calculated by Eq(5) – (7)

$$Q_g = m_{g,n}C_{p,g}(t_{g,in} - t_{g,out}) = h_gS(T_g - T_m) \tag{5}$$

$$Q_{rec} = m_{rec}\gamma(T_m) \tag{6}$$

$$Q_c = m_cC_{p,c}(t_{c,in} - t_{c,out}) = h_cS(T_m - T_c) \tag{7}$$

where $Q_g$, $Q_{rec}$ and $Q_c$ are the amount of heat transfer of flue gas, recovered water and cooling water. $m_{g,n}$and $m_c$ are the mass flow of non-condensed flue gas and cooling water. $T_g$, $T_m$ and $T_c$ are the average temperature of flue gas, membrane tubes and cooling water. $S$ is the membrane area. $\gamma(T_m)$ is the latent heat of water vaporisation at the temperature $T_m$.
The heat exchange coefficients $h_g$ and $h_c$ can be obtained by selecting a suitable heat transfer coefficient calculation formula, which could be determined by experiments. The water recovery rate $\eta$ is measured by the ratio of mass flow of recovered water to the total mass flow of water vapour in flue gas $m_{g,v}$ as shown in Eq(8).

$$\eta = \frac{m_{rec}}{m_{g,v}} \tag{8}$$

The software MATLAB 2019a is used to solving the coupled governing equations. The input is structural parameters of membrane module such as $W$, $H$, $L$, $D$, $S_1$ and $S_2$, material parameters of membrane tubes like the heat and mass transfer coefficient, and inlet parameters of cooling water and flue gas including temperature, pressure, flowrate, and water vapour content. A dichotomy iterative method is used to quickly derive the operating state of the membrane separation process, including $T_m$, $m_{rec}$, $Q_g$, $Q_{rec}$, $Q_c$ and $\eta$. The model has strong applicability to be applied to different kinds of membrane modules by changing the specific input and can be used to explore the influences of different module configuration.

## 3. Case studies

Recent research reported good performance using a composite ceramic membrane to recover water vapour and waste heat from flue gas (Gao et al., 2019). So, composite ceramic membrane modules are selected as the modelling object and set up three cases for analysis considering different working conditions of flue gas and specific parameters of membrane modules. As shown in Table 1 and Table 2, the data of porous ceramic membrane tubes is based on the experimental research (Chen and Yang, 2018), and propose a set of structural parameters of the membrane module according to Chen et al. (2019). The conditions of flue gas and cooling water are based on these studies and expanded appropriately.

*Table 1: Parameters of the porous ceramic membrane tube*

| Tube length (mm) | Tube diameter (mm) | Pore diameter (nm) | Membrane tortuosity | Membrane thickness (mm) |
|---|---|---|---|---|
| 400 | 12 | 20 | 2.8 | 1 |

*Table 2: Basic parameters of the membrane module*

| Module length (m) | Module height (m) | Module width (m) | Lateral tube spacing (m) | Longitudinal tube spacing (m) |
|---|---|---|---|---|
| 0.15 | 0.2 | 0.4 | 0.04 | 0.015 |

The working conditions of a tail flue vary greatly. Along the flow direction of a boiler flue, the temperature of flue gas decreases to around 130 °C, and the water vapour content keeps relatively constant at a low level. But after the desulphurisation tower, the temperature drops to around 45 °C and the water vapour content rises significantly to a nearly saturated state. To determine what operation modes of a ceramic membrane module placed before or after the desulphurisation tower are, the performance of water and heat recovery are observed under different flue gas temperatures and water vapour contents.
There are many optional parameters of a membrane module, such as tube diameter and tube spacing. They would determine the structure of a membrane module, affect the overall heat and mass transfer performance. The diameter of membrane tubes is a key parameter for membrane module design since it determines the total membrane area of a membrane module and would affect both heat and mass transfer processes and the economics of a module. The trend of water recovery performance changes under a series of flue gas flowrates is explored by fixing the number of ceramic membrane tubes and changing tube diameters.
Distance between two tubes is another important structure parameter for membrane module design. The change of the distance would affect the number of membrane tubes and heat transfer coefficient between the flue gas and tube bundles. Fixing the membrane tube diameter and the module size, the lateral tube spacing is changed to observe the trend of water recovery performance under different flue gas flowrates.

## 4. Results and discussion

### 4.1 Location of ceramic membrane module arrangement

The results of different flue gas conditions are shown in Figure 2.

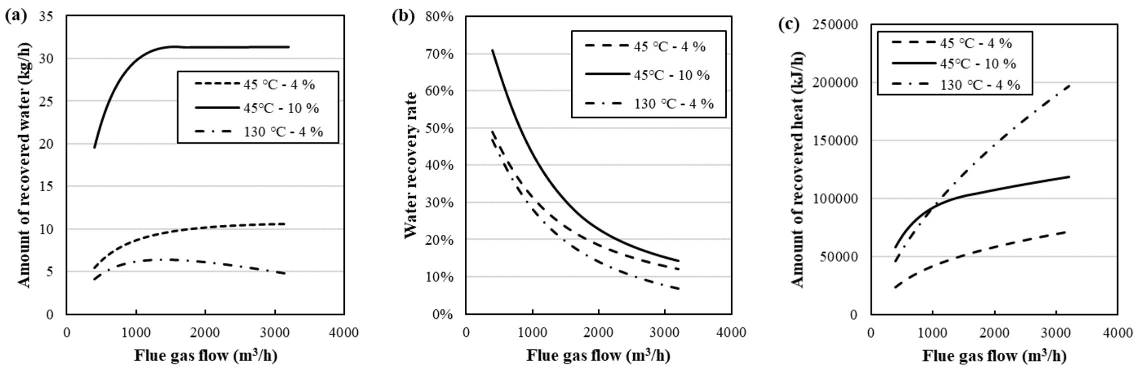


*Figure 2: Effect of different flue gas temperature and water vapour content on (a) amount of recovered water, (b) water recovery rate, (c) the amount of recovered heat.*

In the case of high water vapour content with low flue gas temperature (corresponding to the flue gas status after desulphurisation tower), the amount of recovered water and water recovery rate of the membrane module are both highest, and the heat recovery is moderate due to the release of the latent heat from the condensed water vapour. Under the condition of low water vapour content and low temperature, the water and heat recovery are both low because of the decrease of the partial pressure of water vapour and the temperature difference. In the case of low water vapour content and high flue gas temperature (corresponding to the flue gas status before the desulphurisation tower), the convective heat transfer between the flue gas and the cooling water increases. It makes the temperature of cooling water higher than that under the lower temperature, which means the driving force of mass transfer would decrease and lead to a lower water recovery rate. A worsening water recovery would occur at high flowrates due to a sharp rise in convective heat transfer. If a ceramic membrane module is arranged before the desulphurisation tower, it would mainly work in a heat recovery mode with a good heat recovery performance. If it is arranged after the desulphurisation tower, it would work in a water recovery mode with good water recovery performance.

### 4.2 Diameters of ceramic membrane tubes

It's shown in Figure 3 that there are a series of optimal diameters that maximise water recovery and water recovery rate simultaneously for every flue gas flowrate at certain module structure and membrane condition.

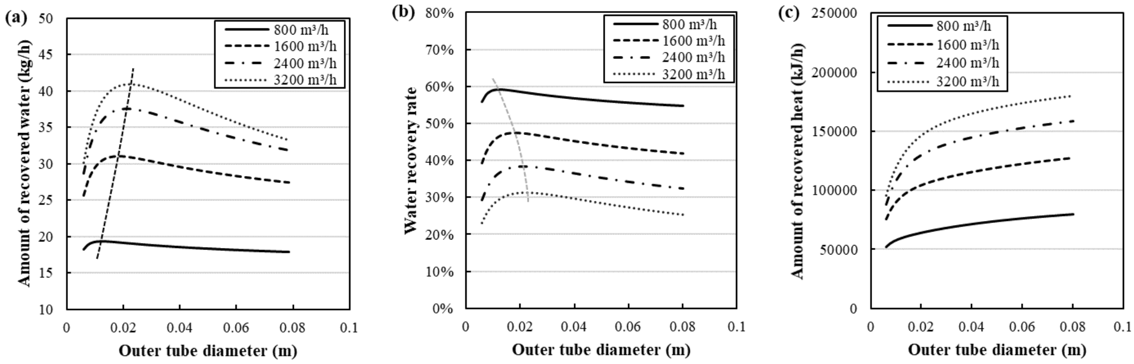


*Figure 3: Effect of different outer tube diameter on (a) amount of recovered water, (b) water recovery rate, (c) the amount of recovered heat.*

The value of optimal diameter increases with the increase of flue gas flowrate. When the outer diameter increases, the membrane area becomes larger and enhances the heat and mass exchange across the membrane. The higher flowrate, the larger membrane area required. However, the enhancement of heat exchange may result in the rise of mean membrane surface temperature, which could restrain the driving force of mass transfer. From Figure 3, it is observed that although the water recovery increases, the water recovery rate decreases with the rise in flue gas flowrate. This phenomenon has been reported by previous experimental studies (Chen et al. 2017). For a certain flue gas flowrate, the tube diameter should be selected around the optimal diameter to obtain both good water recovery and heat recovery efficiency.

### 4.3 Tube spacing of the ceramic membrane module

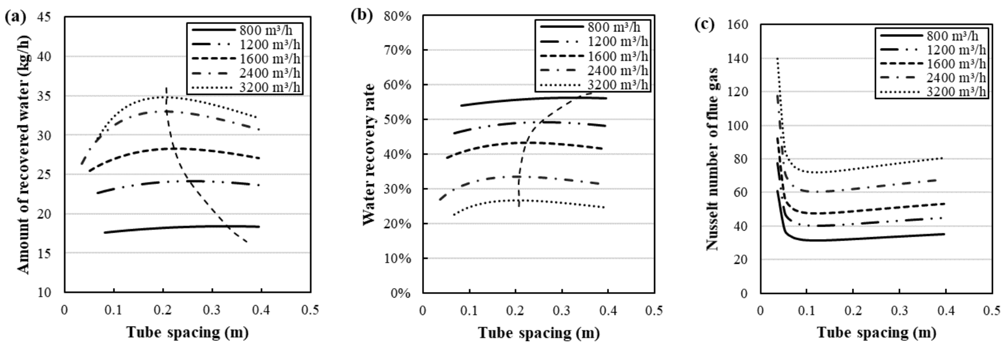


*Figure 4: Effect of different tube spacing on (a) amount of recovered water, (b) water recovery rate, (c) Nusselt number of flue gas.*

The optimal distance decreases with the increase of flue gas flowrate. The reason is that when the distance is very small, the membrane tubes are arranged densely, and the heat exchange is violent between the flue gas and the cooling water. It would raise the cooling water temperature and reduce the driving force of mass transfer. The heat exchange gradually becomes moderate when the tube spacing gets larger. Consequently, the driving force of mass transfer increases so that the water recovery increases. However, when the tube spacing keeps increasing, the heat transfer coefficient changes little, but the number of membrane tubes decreases in inverse proportion. The decrease of membrane area would lead to a reduction of water recovery. For determined flue gas flowrate, the tube spacing should be set in a certain range to ensure a high water recovery efficiency and avoid heat exchange exacerbation.

## 5. Conclusions

The application of membrane separation in power plants requires performance data under wide operating conditions and structure configuration. To support the design of membrane modules, a general model is established in this paper to calculate water vapour and waste heat recovery performance of the membrane separation process under wide-range working conditions and structure parameters. The model investigates the impacts of module location, tube diameter, and tube spacing in a ceramic membrane module. Results showed that a ceramic membrane module arranged before the desulphurisation tower recovers more heat while arranged after the desulphurisation tower it recovers more water. The optimal tube diameter and tube spacing for water recovery would be altered by the competition between the membrane area and the driving force of mass or heat transfer under different working conditions. The appropriate tube diameter and spacing should be selected based on the performance calculation under required working conditions to realise better water or heat recovery. Although only ceramic membrane modules are investigated in this study, the proposed model has strong applicability and generality and is adaptable to other types of membrane modules by adjusting the corresponding mechanism equations of heat and mass transfer.

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