Status and progress of membrane separation technology in water capture in flue gas

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Abstract. The flue gas of power plant contains a lot of water, it is of great significance to save water resources if the water can be recycled and utilized. Gas membrane separation technology, which integrates the advantages of low investment, small area, simple operation and no secondary pollution, is considered to be a feasible technology to effectively realize the recycling of water resources. It has become a research hot spot of flue gas treatment in power plants. This review refers to the completed and ongoing research on membrane separation of water in flue gas and summarizes the advantages and challenges of current membrane-based technologies in water resource recovery.

1. Introduction
As of April 2020, China’s installed capacity for power generation is 1.92 billion kilowatts, up 5.3 percent from the same period last year. 1.19 billion kilowatts of thermal power, 61.9% of total installed capacity, of which 1.04 billion kilowatts are generated from coal, account for 54.2% of the total installed capacity [1]. The proportion of installed capacity of different generation modes is showed in Fig. 1. Now, Renewable energy and nuclear energy still can not meet the energy demand worldwide. Conventional fossil fuels will remain the main energy for human survival in the coming decades[2]. The flue gas emitted from coal-fired power plants mainly contains nitrogen, water vapor and carbon dioxide. The quantity and proportion of each component of flue gas vary with the coal type, water vapor has considerable latent heat which accounts for about 5-11% of flue gas[3]. No doubt, The direct discharge of flue gas into the air can cause waste of energy and water. If waste heat and water vapor can be recovered from the flue gas, it can not only save considerable energy, but also save water resources, which is of great significance to power plants in water-deficient areas [4-6]. In China, it is estimated that 35 million tons of standard coal will be saved per year if half of the latent heat from the flue gas can be recovered [7, 8]. Therefore, the recovery of water from the flue gas of power plant is of great significance to solve the problem of water shortage.
1.1. Membrane separation technology and application

Recovery of heat and water from low-grade waste gases requires effective alternative technologies. Membrane separation technology has great potential in the recovery of heat and water in wet exhaust flue gas [9–12]. Membrane separation technology can overcome the shortcomings of traditional recovery technology. It gradually attracts more and more researchers’ attention because it has the advantages of extremely high gas-liquid interface area, minimal equipment size, enhanced process, easy operation and small environmental pollution.[13–15]. It has higher heat transfer efficiency than traditional recovery technology, and can also solve the problems of corrosion, scaling and high energy consumption [12,16–19]. It has great potential in water and heat recovery. However, the research of this recovery technology is still immature. Therefore, the research of membrane separation Technology to recover water from flue gas in power plant is helpful to its popularization and application as soon as possible, and to create considerable value in sustainable development and economy. To sum up, the membrane technology has important academic value and wide application prospect in recovering water from flue gas in power plant.

1.2. Types of membranes separation

Membrane plays an important role in the separation process when separate water from flue gas. The membrane shall have the following characteristics: (1) Good high temperature degradation resistance. (2) High porosity to minimize mass transfer resistance. (3) It has high mechanical strength to prevent deformation owing to high flow rate of flue gas, which affects separation performance. (4) Hydrophilicity.

1.2.1. Organic membranes. Macedonio et al.[20,21] reported an experiment on the water vapor in the waste gas recovered from the polyvinylidene fluoride(PVDF) membrane. All tests were carried out in the artificial flue gas flow without the remaining impurities. The aim of this study was to evaluate the ability of this new condenser to recover water from the artificial flue gas flow. The results show that the water recovery was 20%. However, this organic polymer membrane has poor mechanical properties, thermal stability and pollution resistance, and its service life in complex flue gas is very short.

With the researchers' experiments on different material membranes, they found that organic composite membranes have better properties. Chen et al. [22,23] used polyethersulfone-sulfone polyether ketone (PES-SPEEK) hollow fiber composite membrane to separate water, and studied the feasibility of such membrane to recover water in flue gas. Where the selective layer is the core structure of the separation membrane and plays a vital role in improving the permeability and separation performance of the membrane. When selecting selective layer materials, the permeability and separation performance of water vapor with other gases should be considered.

Both SPEEK and PES have extremely high water vapor permeability and selectivity in mixed gases as shown in Fig.2, while intermolecular interactions make the two substances more closely linked[24]. The results show that the SPEEK/PES composite membrane has excellent heat resistance and strong
mechanical stability, and can be used for environmental of high temperature flue gas[3,23]. The application of the composite membrane to recover water in flue gas has a good prospect.

1.2.2. Inorganic membranes. The American institute of natural gas technology (GTI) proposed the use of a transport membrane condenser (TMC) to recover water and heat from flue gas[25]. The TMC is located after the desulfurization tower of the power plant, which is composed of several ceramic membrane bundles. For micro-nanopore membranes, the special transmission mode is capillary condensation, in which steam condenses in the membrane pore structure and completely blocks the pores to prevent the transport of non-condensable gas components. Condensate water flows inside the tube and flue gas flows outside the tube. When the flue gas passes through the membrane tube, under the action of the driving force, the water will penetrate into the inner side of the membrane tube [26-29]. Throughout the membrane structure, the pore size of the selective layer is at the nanoscale, and the pore size of the support layer is at the microscale[30]. At the same time, it has good hydrophilicity, absorption flux, mechanical strength and long-term stability. For the application of nano-ceramic membrane to water recovery, the recovery efficiency of water and heat reached 20-60% and 33-85%, respectively [28]. Chen et al. [31] made coal ash ceramic membrane electric plant coal ash, which reduced the sintering temperature of production ceramic membrane. At the same time, the heat and mass transfer performance of coal ash ceramic membrane was tested. The experimental results show that the coal ash ceramic membranes and microporous ceramic membranes had similar mass transfer characteristics, and the heat transfer performance was lower than that of microporous ceramic membranes.

1.3. Membrane modification
In virtue of the composition in the flue gas is complex and the impurity is too much, the using effect of the membrane is seriously affected. It has become an urgent problem to improve the hydrophilicity and anti-fouling ability of the membrane. Membrane modification techniques include plasma modification, non-solvent assisted deposition, sol-gel coating and uv light grafting.

Muhammad et al. [32] added carboxylated TiO$_2$ nanoparticles into the polyamide membrane matrix to prepare thin-film nanocomposites (TFN) membranes for water vapor separation. The water vapor permeability of TFN membrane was tested on laboratory scale and carboxylation TiO$_2$ was studied, effect of nanoparticles on water vapor permeation and selectivity. As a result, adding carboxylation TiO$_2$ greatly improves water vapor selectivity and transmittance. Hu et al. [33] modified the surface of nanoporous ceramic film hydrophilic and hydrophobic, respectively. The contact angles of the membrane surface after hydrophobic and hydrophilic modification were 137.5° and 26.25°, respectively. The heat and mass transfer process of the modified nanoporous ceramic membrane was investigated. The experimental results show that the hydrophilic nano-porous ceramic membrane has high performance of condensation heat transfer and water recovery.

Although the hydrophilic modification of the membrane is mature, but few studies on membrane modification applied to dehydration in flue gas. Due to in practice, the membrane condenser will last
for months or even years in a power plant. After surface modification, it is of great significance to improve the long-term stability of membrane properties.

Up to now, there is a lack of a review on membrane-based separation techniques for water recovery in flue gas. This review summarizes the research work on membrane separation of water from flue gas in power plants, and reviews the articles published so far in experimental research and numerical simulation.

2. Theory of mass transfer in membrane

In the membrane condenser, the flue gas and cold water are separated by the membrane. The process of water vapor entering condensed water through the membrane in flue gas is generally divided into three steps: (1) the diffusion of gas to the gas-membrane interface; (2) the diffusion from the gas-membrane interface to the liquid-membrane interface through the membrane pore; (3) the transfer from the liquid-membrane interface to the liquid. With the continuous progress of membrane technology, the composite membrane improves the water separation performance of the membrane, so it is particularly important to study the mass transfer process in the composite membrane.

2.1. Basic mass transfer model

There are three main transport mechanisms for porous media: Knudsen diffusion, molecular diffusion, and viscous flow. The dominant mechanism can be judged by Kn number. The Fick model (FM), the Maxwell-Stefan model (SMM) and the dust-gas model (DGM) are the current commonly used gas transport models[34].

Given the complexity of the composite membrane structure, a certain model can no longer explain the whole mass transfer process. Pressure gradient difference is the main driving force for multi-matter transport. Nevertheless, Fick’s law is used to explain the component transfer process driven by concentration gradients. Therefore, combining Maxwell-Stefan theory with the DGM model can accurately explain the whole mass transfer process.

2.2. Water mass transfer model for membrane separation

The flue gas contains N\textsubscript{2}, O\textsubscript{2}, CO\textsubscript{2}, H\textsubscript{2}O, SO\textsubscript{2}, NO\textsubscript{2} and other components. Because the flue gas has already been treated with desulphurization and denitrification when it enters the membrane separation assembly, the SO\textsubscript{2} and NO\textsubscript{2} in the flue gas are very small, so only the first four gases have been discussed[8,35,36]. Taking 110°C flue gas as an example, the mass fraction of each component in the flue gas is shown in Fig.3.

Figure 3. Volume fraction of flue gas components.

2.2.1. Mass transfer of the selective layer. The selective layer is relatively high, and the separation of water in flue gas mainly depends on the selective layer. By using the flux difference form of the Maxwell-Stefan equation, the mass transfer process is represented:

\[ -\bar{x}_i \frac{\Delta p_j}{\rho_j \Delta \delta} = \sum_{j=1}^{4} \frac{x_i N_j - c N_j}{D_{ij}^c c} + \frac{N_j}{D_{im} \bar{c}} \]

(1)

Where \( i,j=1,2,3,4 \), represent H\textsubscript{2}O, N\textsubscript{2}, O\textsubscript{2} and CO\textsubscript{2}, respectively; \( \bar{c} \) is the average concentration before and after the selective layer for the multicomponent mixture; \( N \) is the flux of the gas through
the selective layer; $\bar{x}$ is the molar fraction of the gas; $p$ is the gas separation pressure; $\Delta \delta$ is the thickness of the selective layer.

The friction between the gas can be ignored. Equation (1) is converted to:

$$N_i = -\frac{D_{i,m}^*}{\Delta z} He, \Delta p_i$$  \hspace{1cm} (2)

From equation (2), the mass transfer of the selective layer is affected by the solubility of the gas in the selective layer and the partial pressure of the gas. The material and gas partial pressures of the selective layer affect the membrane selectivity and flux size, respectively.

2.2.2. Mass transfer of the connecting layer. Assuming that the pore distribution is uniform, the flux equation of Knudsen diffusion in the connecting layer and the flux equation of dissolved penetration are:

$$-\frac{x_i \Delta p_i}{p_i \Delta z} \varepsilon = \sum_{i=1}^{m} \frac{x_i N_{i,j} - \bar{x}_i N_{i,j}}{D_{i,j}^* \varepsilon} + \frac{N_i}{D_{i,m}^* \varepsilon}$$  \hspace{1cm} (3)

$$N_{i,m} = -\frac{D_{i,m}^*}{\Delta z} H_m \Delta p_i (1-\varepsilon)$$  \hspace{1cm} (4)

By using the theory of Henis resistance model of the composite membrane, the mass transfer flux in the connecting layer is the sum of the two fluxes:

$$N_{i,sum} = N_{i,j} + N_{i,m}$$  \hspace{1cm} (5)

Obviously, equation (5) shows that the pressure difference and the thickness of the embedded part affect the flux size of the connecting layer.

2.2.3. Mass transfer of the support layer. The support layer is a porous region in which the mass transfer process is Knudsen diffusion and viscous flow.

In the support layer area, thanks to the effect of viscous flow, in addition to the diffusion velocity, there is also the effect of viscous flow on the total velocity. In the same way as Knudsen diffusion, viscous flow is regarded as driven by the interaction of multi-component flue gas mixture.

$$F = \xi_v v$$  \hspace{1cm} (6)

The coefficient of friction is related to the hydraulic permeability of the support layer structure.

$$\xi_v = \frac{\eta}{cB}$$  \hspace{1cm} (7)

$$v = \frac{1}{32} \frac{\Delta \delta}{\eta \Delta p^2} \hspace{1cm} (8)
$$

Where $\xi_v$ is the viscous friction coefficient; $d_p$ is the pore diameter; $B$ is hydraulically permeable coefficient.

$B$ is determined by the distribution of void diameter and pore size. The total velocity is the sum of the viscous flow velocity and the Knudsen diffusion velocity, and the total flux of the component is the sum of the flux of the two flow modes. Therefore, the main factors affecting mass transfer flux are porosity and pressure.

3. Numerical simulation and experimental study on mass transfer of membranes

In this section, the review aims to discuss the progress made by researchers in the membrane method for water capture simulation and to understand the modeling methods for water separation in flue gas with different membranes. It describes the research progress from treating porous membrane tubes as solid regions to porous regions, providing some assistance to new entrants and existing researchers in this field.
3.1. Numerical simulation of membrane mass transfer

The modeling and simulation of water in flue gas by membrane separation have been studied for more than ten years [35–38], and is only in the preliminary stage at present. It should be emphasized that for simplicity, almost all modeling methods treat water as the only mass transfer phenomenon.

3.1.1. Modelling framework. A general framework diagram for modeling and simulation of water processes in membrane separation flue gas is shown in Fig.5. The key variables that must be defined when simulating the water performance in the membrane separation flue gas and the different objectives of modeling simulation research are summarized in the figure. Any modeling problem starts with a set of assumptions about the physical and chemical properties of the system.

![Figure 4. Process modeling and simulation frame diagram.](image)

![Figure 5. Model of membrane tube bundle [38].](image)

As shown in Fig.4: the input variables in the simulation study can be divided into two main categories: (i) operating conditions; (ii) membrane and module properties. Usually, the mixture gas with water vapor content of 10-15% is selected to simulate the flue gas of coal-fired power plants. Using ambient temperature, the gas pressure is the standard atmospheric pressure, usually the Model of membrane tube bundle is shown in Fig.5.

3.1.2. Numerical simulation. Because the simulation of water in the flue gas of membrane separation is basically in the preliminary research stage, only two models are listed in this paper to evaluate.

3.1.2.1. Numerical simulation using two-step condensation-evaporation model. Lin et al.[36] developed a multi-species transport model to simulate the process of flue gas swept transport membrane condensing tube bundle according to practical problem. As shown in Fig.6, it can be seen that there is a virtual water membrane outside the tube wall, in which evaporative condensation occurs. This process is considered as a two-step reaction.

\[
H_2O(gas) \xrightarrow{\text{Condensation/Evaporation}} H_2O(\text{virtual mass}) + \text{heat}
\]  

(9)
Figure 6. Multi-species transport model (STM) [36].

The introduction of virtual mass in the reaction is only for the convenience of calculation. In the numerical simulation, there is no actual liquid membrane outside the tube wall. After simulation, the model was verified and the two extreme cases were compared. Scheme 1 and 2 were the minimum condensation rate and the maximum condensation rate, respectively. The comparison of the average values of outlet water temperature, outlet flue gas temperature and outlet water vapor mass fraction obtained from the experiment and computational models are listed in Table 1. It can be seen from the table that the error between the experimental results and the simulation results is between -7.76% and 11.0%. It can be said that the simulation results are basically consistent with the experimental results, and the model is relatively close to the actual mass transfer process.

Table 1. Experiment and simulation results compare, modified from [36].

| Case | EXP(℉) | SIM(℉) | Error (%) |
|------|--------|--------|-----------|
| Flue gas exit temperature (℉) |       |        |           |
| 1    | 138.9  | 145.0  | 4.39      |
| 2    | 113.3  | 125.8  | 11.0      |
| Liquid water exit temperature (℉) |       |        |           |
| 1    | 134.5  | 133.8  | -0.52     |
| 2    | 95.5   | 100.6  | 5.34      |
| Water vapor exit mass fraction (℉) |       |        |           |
| 1    | 1.031  | 9.51   | -7.76     |
| 2    | 5.19   | 4.79   | -7.71     |

3.1.2.2. Semi-empirical model combining capillary and wall condensation. After optimizing and improving the model, Soheil et al.[37,38] used a semi-empirical condensation model based on capillary condensation model and wall condensation model. Since the two models simulate the same process, only the advanced nature of the model is discussed. The model is based on capillary condensation model and wall condensation model. The water and heat transfer from the flue gas side to the porous media is realized during the simulation. The related source term needs to be applied to the unit near the porous media wall. The model is implemented in Ansys Fluent using a user-defined function (UDFs).

By comparing the simulation results of this mixed model with the previous experimental results, the relationship is given in the form of combination diagram by analyzing the of literature data[15,38]. As shown in Fig.7 and Fig.8, the experimental and simulation results of condensate water and flue gas outlet temperature are compared respectively. It is very intuitive to see that after using the new simulation model, the temperature error of condensate water and flue gas outlet is much smaller than that of the previous two-step condensation-evaporation model. It shows that the mixing model can predict the condensation rate on the membrane tube more accurately.
3.1.3. Unsolved problems in simulation. Despite great efforts and improvements in modeling and simulation of water in membrane separation flue gas, there are still some unsolved problems.

In most simulation studies, the flue gas is composed of only a few gases, which is very different from the actual composition. A few tiny particles were suspended in the flue gas without considering the influence of its attachment to the surface of the membrane tube bundle on separation effect.

The mass transfer of water will change with time in practice, but most experiments on membrane separation are conducted on a limited time scale (usually days). A significant mass transfer performance change may occur at long time scales[39-41]. The limitation is regarded as a major problem in simulation.

3.2. Experimental study of membrane water recovery
Researchers have carried out a large number of experimental studies on water in membrane separation flue gas, covering many aspects.

3.2.1. Comparison of performance between membrane tube and stainless steel tube heat exchanger.
Bao et al.[15] compared the condensation performance of membrane condenser and stainless steel tube heat exchanger.
Table 2. Comparison of nano-porous membrane bundles of the same characteristic size and non-permeable stainless steel bundles.

| Unit                      | Nano-porous membrane bundles | Stainless steel tube |
|---------------------------|------------------------------|----------------------|
| Flow velocity of inlet flue gas (m/s) | 0.50                         | 0.44                 |
| Reynolds number           | 260                          | 280                  |
| Smoke inlet temperature (°C) | 65-95                        | 65-95                |
| Steam mass fraction (%)   | 11.3                         | 11.3                 |
| Nusselt number            | 50-80% higher than stainless steel tube bundle |
| Condensation rate         | The porous membrane tube bundle is 60-80% higher than the stainless steel tube bundle |

The comparison of nano-porous membrane bundles and non-permeable stainless steel bundles with the same characteristic size is shown in Table 2. Experimental results show that the convection Nusselt of porous membrane tube bundle is 50-80% higher than that of impervious stainless steel tube bundle. The condensation rate of porous membrane bundles is 60–80% higher than that of stainless steel bundles.

3.2.2. Effects of different pore size membranes on water recovery. Owing to the pore size of the membrane selected in the experimental study is different, the water flux of porous ceramic membranes with different pore sizes is compared according to the previous experimental results. As shown in Table 3, the highest water flux recovered increases with the increase of pore size.

Table 3. Recovered water flux of ceramic membranes with different pore sizes.

| Research | Pore size [nm] | Highest recovered water flux [kg/(m²·h)] | Gas type | Surface area of membranes [m²] |
|----------|----------------|------------------------------------------|----------|-------------------------------|
| Ref[42]  | 0.4            | 0.8-0.9                                   | True flue gas | 0.015                         |
| Ref [26] | 6-8            | 7.76                                     | True flue gas | 0.58                          |
| Ref [7]  | 7              | 8                                        | Nitrogen and water vapor | 0.0021                       |
| Ref [13] | 20             | 8.5                                      | Nitrogen and water vapor | 0.052                        |
| Ref [43] | 40             | 10                                       | Water vapor, sulfur dioxide and air | 0.00716                      |
| Ref [43] | 90             | 12                                       | Water vapor, sulfur dioxide and air | 0.00532                      |

To sum up, most of the research on the water in the membrane recovery flue gas only stays at the laboratory level. The test object is simulated flue gas, which lacks the performance research under the actual flue gas, so it is necessary to carry out the actual test in the power plant.

4. Challenges and research direction of membrane technology

4.1. Membrane fouling
Membrane fouling will cause the mass transfer flux to decrease gradually, and it is difficult to recover to the original flux level. At the same time, it will cause the membrane hole to be blocked, and the interception performance will change, which will reduce the capacity of the membrane to recover water, and the service life of the membrane will be greatly reduced, and its original application value will be lost. Membrane fouling phenomenon is the main obstacle to the further development of the application field of global membrane technology. A large number of studies on membrane fouling are very worthwhile.

4.2. Selection of membrane system parameters
The recovery of water from flue gas in power plants by membrane separation technology has been gradually accepted by the public, but the service life of membrane separation components in actual flue gas is unknown. The membrane tube will be deformed by the smoke flow, which will affect the performance of the membrane assembly. Therefore, it is necessary to analyze the stress and strain of
the membrane tube. According to the analysis results, the membrane module is optimized, which will extend the service life of the membrane, thus the performance of the whole system will be improved.

5. Conclusions
Membrane separation is a very promising technique, which has attracted great attention in flue gas water recovery. It integrates heat transfer and membrane separation (modularity and high selectivity). Because the key problems such as membrane fouling have not been solved satisfactorily, most of the research is still carried out in laboratory scale. Up to present, the experimental studies have verified the technical feasibility of membrane separation of water in flue gas. The future work is continuing to apply this experience to commercial production.

Acknowledgments
This work was financially supported by the research Start-up fee project of Shandong University of Science and Technology "study on the mechanism of acid-ash coupling based on mesoscopic scale”

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