



One-step pasting method for preparation of flat-sheet ceramic membrane

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ABSTRACT

Flat-sheet ceramic membrane without any transition layer was prepared directly applied on the surface of the support layer by a one-step pasting method. The preparation of the different thickness of plastic clay slice as the separation layer is the key point in membrane fabrication. The result shows that the formation of the defect-free ceramic membrane with even pore-size distribution after calcinated at 1,350°C for 2 h. With increasing the thickness of the membrane layer, the average pore-size did not change notably. But, the permeability and the largest hole of the membrane decreased gradually. Additionally, the defects of the support had no effect on the performance of the separation layer with the one-step pasting method. Meanwhile, this method can improve the efficiency greatly and save the cost of the production.

Keywords: Ceramic membrane; One-step pasting method; Low-cost; Defect-free

1. Introduction

Membrane separation technology has been widely used in the fields of gas and liquid filtration, purification, separation, thermal insulation, and other applications, especially the ceramic membrane and it has the advantages of high porosity, high-temperature resistance, good corrosion resistance, and high chemical stability [1–5]. For higher permeability, the ceramic membrane should have an asymmetric architecture that consists of a separation layer, transition layer, and support layer with large pores [6–8]. The support layer provides high mechanical strength and permeability of the membrane layer [9–11]. The transition layer provides the appropriate pore-size to bridge the

support and separation layer. Hence, the separation properties of the membrane depend on the performance of the support and transition layer [12–15].

The fabrication process of the flat-sheet ceramic membrane is mainly carried by the dip-coating method [16–19]. During the dip-coating process, the formation of the membrane layer depends on the capillary force of support and viscosity of the coating slurry [20–24]. Therefore, the particles of slurry easily penetrate into the pores of the transition layer or support, which will significantly decrease the permeability. Meanwhile, the preparation of the transition layer will increase the fabrication cost and decrease the permeability of the membrane. Simply because of this, some researchers focused on the preparation method of the

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membrane layer to obtain the ceramic membrane with high permeability and low cost in recent years. For example, the sacrificial inter-layer method [25,26], which transition layer is decomposed during the sintering process. However, the decompose rate of the transition layer will lead to some cracks of the separation layer. The hot-coating method [27,28], which the support layer needs to be heated at 190°C, then the separation layer coating or spraying on the hot support, the particles can be prevented from penetrating into the pores of the support. However, the drying quickly of the separation layer easily causes defects in the separation layer. These limit the industrial preparation and application of the flat-sheet ceramic membrane seriously.

In this paper, the one-step pasting method was proposed to fabricate the high-performance and low cost flat-sheet ceramic membrane. In this method, the preparation of the different thickness of plastic clay slice as the separation layer is the key point to prepare the ceramic membrane without defect. The ceramic membrane without any transition layer, the plastic clay slice (separation layer) is pasted directly on the surface of support. In this way, the particles can be prevented from penetrating into the pores of support and the defects of support have no effect on the performance of the separation layer. Meanwhile, this method has some advantages of low-cost, simple process, and it suitable for industrial applications.

Based on our present work, this paper tries different methods including dip-coating method and one-step pasting method and investigates the effect of different thickness of plastic clay slice on the morphology, pore-size distribution, permeability, and separation performance of the flat-sheet ceramic membranes. We hope that this method may provide more options to prepare the different pore-size of the flat-sheet ceramic membrane with a lower cost.

2. Experimental

2.1. Materials and machines

Alumina (α -Al₂O₃) powders (Luoyang, He'nan, China, Purify $\geq 99\%$) were used without further treatment. The median particle sizes (d_{50}) of the four alumina powders are 29 μm (W40), 3.76 μm (W3.7), 0.22 μm (W0.2), and 0.13 μm (W0.1), respectively. Polyvinyl alcohol as the stabilizer or plasticizers and the concentration of 12 wt.% (PVA-1799, Sinopharm Chemical Reagent Co., Ltd.). Glycerine and polyethylene glycol (PEG-400) can be served as lubricants (Jiulong New Material Technology Co., Ltd., China). Hydroxypropyl methylcellulose can be served as the binder in the preparation process of the different thickness clay slice (RT-6000, Sinopharm Chemical Reagent Co., Ltd.). Rolling membrane machine (RM-350, East ran machinery, China), the roll spacing and speed can be adjusted. The wastewater was prepared by alumina powders (W0.1) and the solid content of 5 wt.%.

2.2. Sample preparation

The preparation process of support by follows:

Firstly, the alumina (W40) powders and the PVA (8 wt.%) was mixed evenly. Then the mixture was shaped into the

round piece of $\Phi 25$ mm by the dry pressing method under the pressure of 12 MPa. Finally, the porous alumina membrane supports were obtained after calcinated at 1,650°C for 2 h, the pore-size of the support is 4 μm and the porosity of 36%.

2.2.1. Dip-coating method

Firstly, the alumina powders were dispersed in pure water, the Dolapix CE-64 as a dispersant, and the addition amount is 0.05 wt.% of powder mass. Secondly, the suspension was mixed with a ball mill at 200 rpm for 3 h with a solid content of 15 wt.% (transition layer, W3.7) or 12 wt.% (separation layer, W0.2). Then the aqueous solution of 30 wt.% (transition layer) or 26 wt.% (separation layer) PVA was added into the above ball-milled suspension as stabilizer. Subsequently, the above suspension was re-ball milled for 2 h to prepare the stable slurry. In the ball milling, the mass ratio of suspension: alumina ball was 1:1.

The preparation process of the ceramic membrane can be operated as follows:

The coating time of transition layer was 40 s, and the samples were dried in an oven at 80°C overnight and sintered at 1,450°C for 2 h. The coating time of the separation layer was 30 s, and the samples were dried in an oven at 80°C overnight and sintered at 1,350°C for 2 h. Then the ceramic membrane was formed.

2.2.2. One-step pasting method

The preparation process of support is similar to that of the support in the above dip-coating method.

In this method, the transition layer does not need to be prepared. The alumina particles (W0.2), pure water, hydroxypropyl methylcellulose, polyethylene glycol, and glycerine were mixed evenly with the ratio of 74:18:4:2:2. Then, the above mixture was treated under vacuum. Lastly, the plastic clay slice with different thicknesses were prepared by rolling the membrane method by adjusting the distance between the roller. The plastic clay slice pasting the surface of the support, then the samples were dried in an oven at 80°C for overnight and sintered at 1,350°C for 2 h. The ceramic membrane was formed by one-step pasting method and the schematic diagram of the formation of the ceramic membrane are shown in Fig. 1.

2.3. Characterization

The particles size distribution of alumina powders was measured by a laser particle size analyzer (Bettersize2000, Dan dong, China).

The pore size distribution of the sintered compacts were measured by mercury intrusion porosimetry (auto-pore IV9500, Micromeritics, USA).

The fracture surfaces and cross-section of the sintered samples were observed by means of Field emission scanning electron microscopy (JSM-6700F, JEOL, Japan).

The water flux of membrane was tested in a cross-flow filtration apparatus (as shown in Fig. 2). The apparatus was capable of operating at a variety of temperatures and

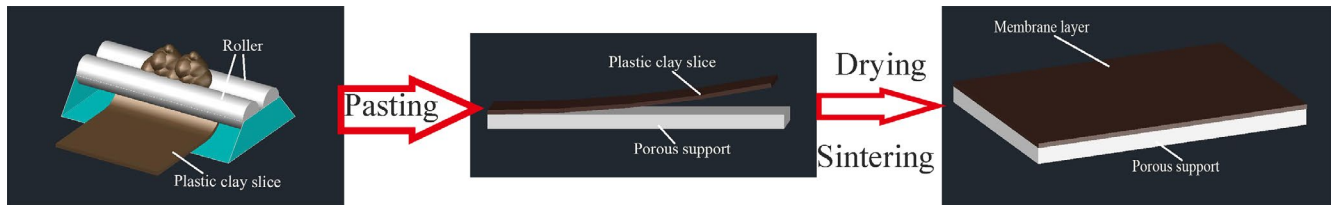


Fig. 1. Schematic diagram of the formation of the ceramic membrane by one-step pasting method.

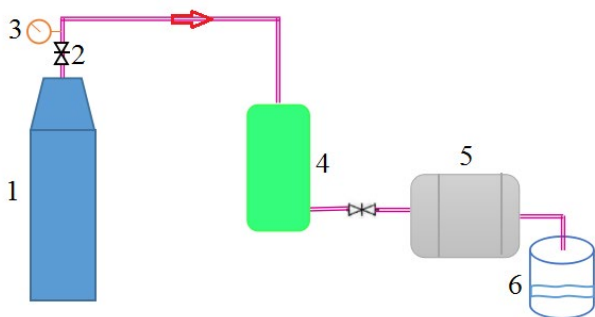


Fig. 2. Schematic diagram of the performance-testing apparatus for ceramic membrane (1) N_2 cylinder, (2) counterbalance valve, (3) pressure gauge, (4) water cylinder, (5) membrane module, and (6) beaker.

pressures. During the test, the volume of the water permeated and the collection time were recorded.

3. Results and discussion

3.1. Effect of the prepared method on the performance of ceramic membrane

Fig. 3 shows the schematic diagram of the formation and the pore size distribution of ceramic membrane by different methods. The dip-coating method is made of two-step coating, and followed by drying and sintering. This method results in an uneven pore size distribution on the surface of the membrane. The largest hole was 500 nm in diameter. This can be explained that the dip-coating method depends on capillary colloidal filtration [15]. Under the action of capillary force, the particles penetrate into the pores of the support layer at two-step dip-coating process (as shown in Fig. 3-A1 and Fig. 4-A1). The surface of membrane layer is uneven, then the big-hole appears in the surface of membrane layer and it leads to the largest hole with 500 nm in diameter (as shown in Fig. 4-A2). Meanwhile, the greater shrinkage occurs while drying and sintering, and the presence of bubbles during membrane layer formation is due to the lower solid content in the slurry.

On the contrary, membranes by one-step pasting method showed even pore size distribution and the largest hole was about 200 nm in diameter. This indicated that the one-step pasting method did not affect by the capillary force. The particles were evenly distributed in the plastic clay slice by the rolling membrane method, the particles bind tightly to each other and it can prevent the particles

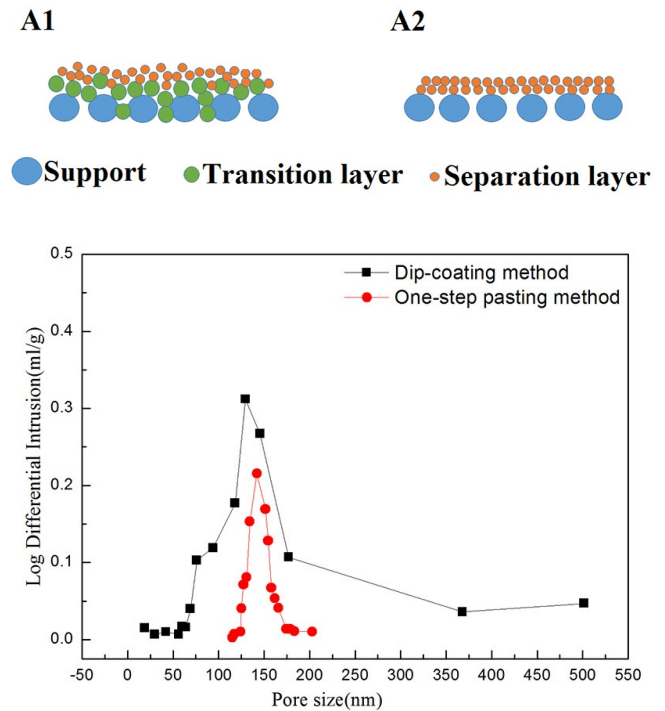


Fig. 3. Schematic diagram of the formation and pore size distribution of ceramic membrane (A1) dip-coating method and (A2) one-step pasting method.

from penetrating into the pores of the support layer. The clay slice can be evenly pasted on the surface of the support layer (as shown in Fig. 3-A2 and Fig. 4-B1). The surface of the membrane layer is smooth (as shown in Fig. 4-B2). Therefore, the formation of the membrane layer can cover the defects and uneven pore-size distribution on the support layer with the largest pores. It is crucial to prepare defect-free membranes with uniform pore-size distribution.

Fig. 4 shows the scanning electron microscopy (SEM) images of the cross-section and surface of the membrane by different methods. As can be seen from Fig. 4, the particles are loosely contacted and some pores appear on the surface of the membrane. Meanwhile, it has leakages in the transition or separation layer in the membrane prepared by the dip-coating method. On the other hand, in the one-step pasting method, the particles contacted closely and the surface of the membrane is smooth, the defects and largest pores in the support had no effect on the formation of the separation layer and its performance. This further verified that discussed in Fig. 3. Consequently, the one-step

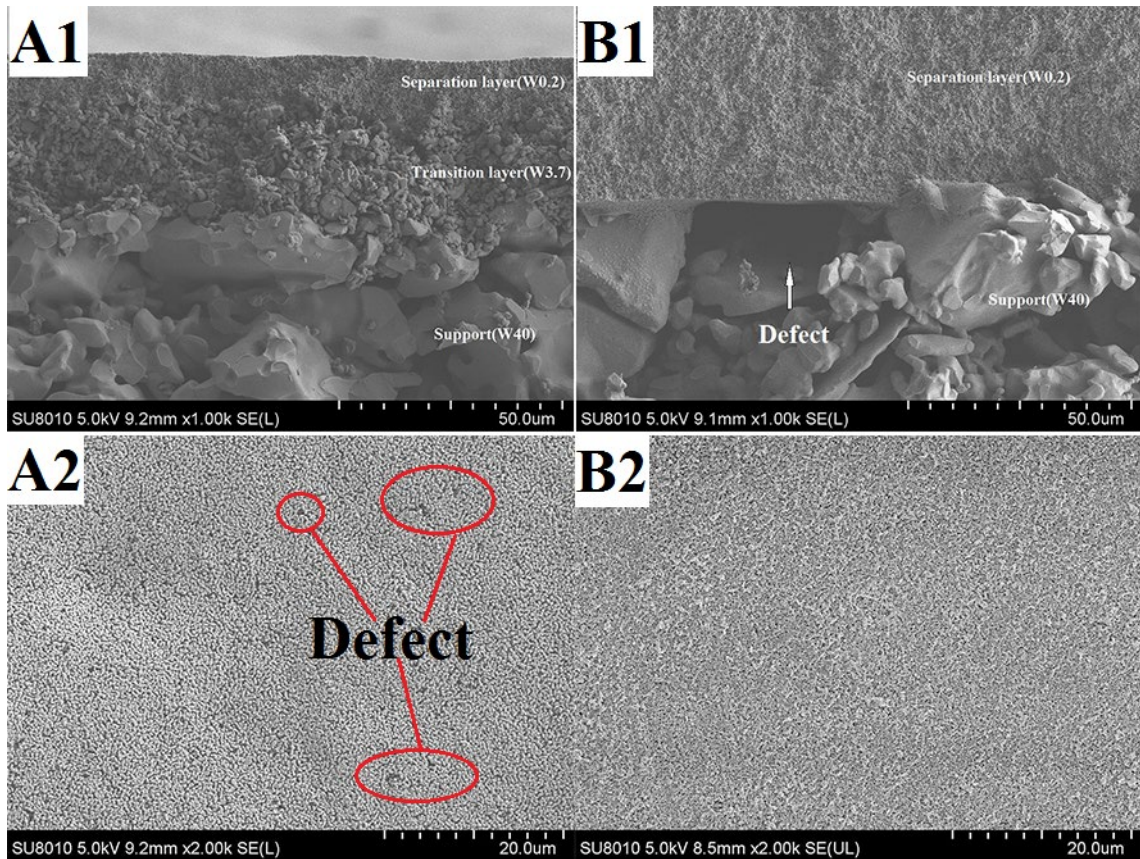


Fig. 4. SEM images of the cross-section and surface of the ceramic membrane by (A1 and B1) dip-coating method and (A2 and B2) one-step pasting method.

pasting method is preferred as an alternative method to prepare the flat-sheet ceramic membrane.

3.2. Effect of the membrane layer thickness on the pore-size distribution of ceramic membrane

Fig. 5 shows the pore-size distribution of ceramic membrane with different thickness of the membrane layer. It can be seen that the pore-size of the membrane decreased gradually with increasing the thickness of membrane layer, especially the largest hole decreased. In particular, the membrane thickness was increased from 90 to 130 μm , the pore-size of the membrane did not change significantly. Meanwhile, the membrane with thickness of 150 μm , the median pore-size was 125 nm in diameter with the largest pore-size of only 165 nm. This indicates that the one-step pasting method can reduce the defects from the largest holes. The membrane thickness has no effect on the pore-size of the membrane. However, the pore-size depends on the particle size of raw materials [29].

The ceramic membrane layer prepared by the one-step pasting method can be regarded as the spreading and layer by layer accumulation of particles. Consequently, the membrane layer thickness increases, the number of particle layers increases gradually. As mentioned above, the particles are evenly distributed and more closely each other after the plastic clay slice by the rolling membrane method. Thus,

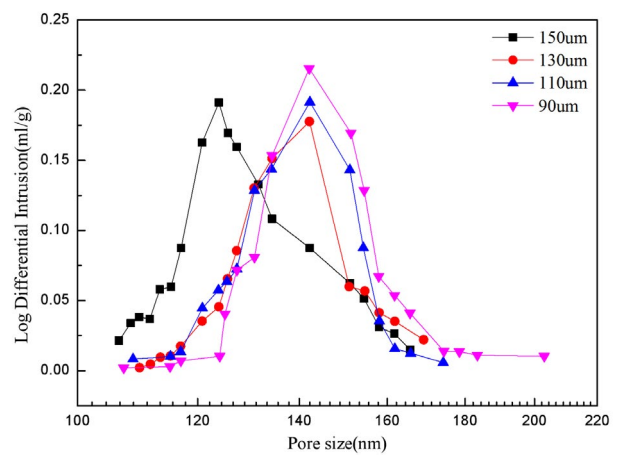


Fig. 5. Pore-size distribution of ceramic membrane with different membrane thickness by one-step pasting method.

as the number of layers increases, the particle rearrangement increases, which only leads to the membrane thickness as affected without altering the particle state of contact. Consequently, the pore-size of the membrane doesn't change, considerably and only the largest hole decreases in size.

Fig. 6 shows the SEM images of the cross-section and surface of the ceramic membrane with different thickness

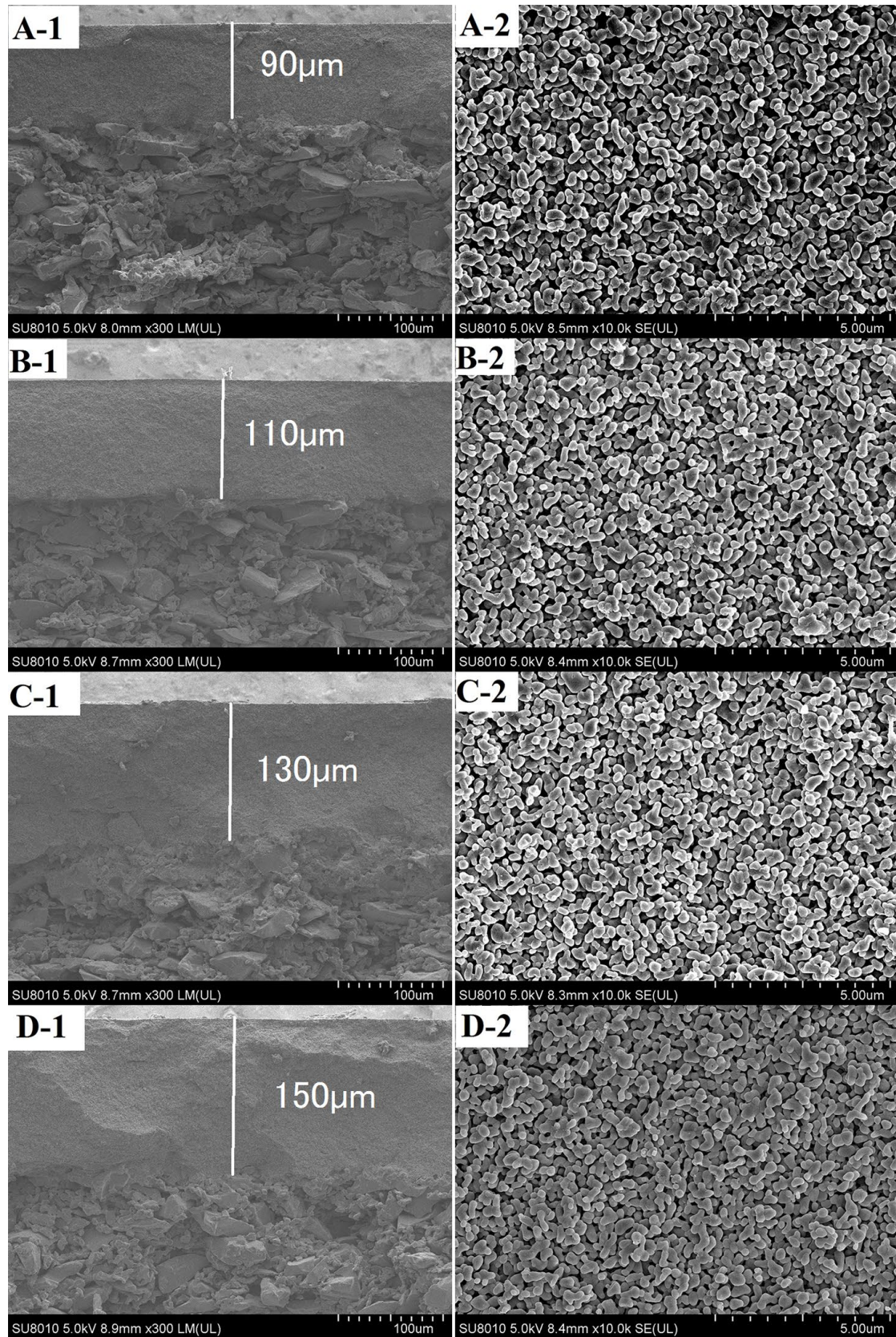


Fig. 6. SEM images of the cross-section and surface of the ceramic membrane with different thickness of membrane by one-step pasting method (A-1 and A-2) 90 μm, (B-1 and B-2) 110 μm, (C1 and C2) 130 μm, and (D1 and D2) 150 μm.

membrane layer. The SEM images clearly show the membrane layer has no phenomenon of particle leakage with increasing the membrane thickness. The separation layer and the support has a clear boundary, and it was firmly combined. Meanwhile, the surface of the membrane layer is smooth and the particles distributed uniformly.

3.3. Permeability of the ceramic membrane

Ceramic membrane is a kind of pressure-driven membrane and its water flux is affected by pressure, membrane thickness and so on [30]. Fig. 7 shows the water flux of the membranes with different membrane thicknesses. As it can be seen, the increase in pressure results increases in water flux. It indicates that the pressure is the only driving force for permeation. In addition, the water flux decreases with the increase of the thickness of membrane layer. The water flux and pressure are linearly related. It implies that the ceramic membrane has a high permeability, even pore-size distribution, and lower tortuosity [31].

Table 1 lists the pure water permeability of the membranes in this work and literature. Along with the increases in membrane thickness, the trans-membrane pressure difference increased accordingly, and the pure water permeability of the membrane decreased in this work. Particularly, the fabrication of the membrane with a one-step pasting method showed a little different permeability than those reported in literature. But, the membrane thickness varies greatly, even by tens of times (Table 1). This is due to that the one-step pasting method can prevent the particles from penetrating into the pores of support and keep the high permeability (shown in Fig. 4B). According to the Hagen Poiseuille Eq. (1) [37], if the membrane thickness decreases by one-step pasting method, the permeability may increase greatly.

$$J = \frac{\varepsilon}{\tau} \frac{r^2}{8\eta} \frac{\Delta P}{\Delta x} \tag{1}$$

where J is the water flux through the membrane, ε is the porosity, τ is the tortuosity, r is the pore-size of the membrane, η is a permeable medium viscosity, ΔP is the trans-membrane pressure difference across the membrane, and Δx is the thickness of the membrane layer.

Table 1
Pure water permeability of the membranes in this work and literatures

Average pore-size (μm)	Membrane thickness (μm)	Pure water permeability ($\text{Lm}^{-2} \text{h}^{-1} \text{bar}^{-1}$)	Reference
0.141	90	525	This work
0.140	110	507	
0.138	130	408	
0.124	150	377	
0.130	15	550	[32]
0.120	20	740	[33]
0.100	45	450	[34]
1.000	–	450.7	[35]
0.140	–	505	[36]

3.4. Separation properties of the ceramic membrane

Fig. 8 shows the properties of the ceramic membrane with a pore size of 141 nm for the wastewater treatment at a trans-membrane pressure of 0.2 Mpa. As shown in Fig. 8, when the filtration time reaches to 2 h, the ceramic membrane can completely intercept the particles in the wastewater. It can be explained that the cake layer is mainly formed within the first hour of filtration. However, with the increases of the filtration time, a new filter layer was formed, which can completely intercept the particles in the wastewater. This further proves that the flat-sheet ceramic membrane were prepared by one-step pasting method and it has a better performance of separation.

4. Conclusions

This work only provides a one-step pasting method for preparing the high-performance flat-sheet ceramic membrane. The ceramic membrane fabricated without any transition layer, but the separation layer was pasted directly on the surface of the support. Meanwhile, this method can prevent the particles from penetrating into the pores of

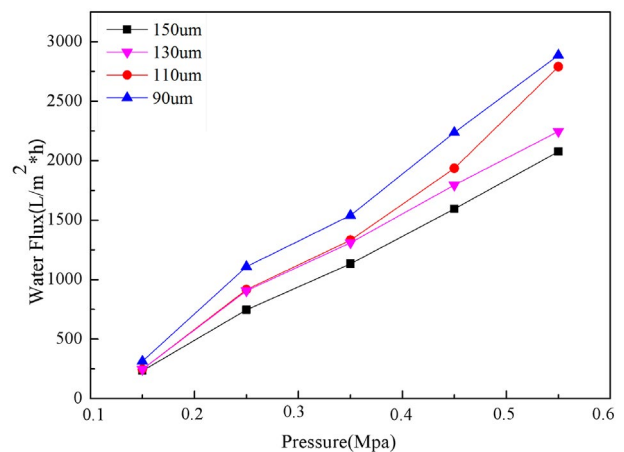


Fig. 7. Pure water flux of membrane with different thickness of membrane by one-step pasting method.

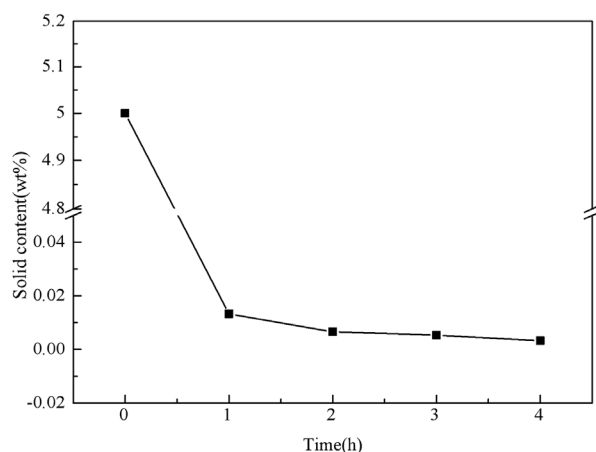


Fig. 8. Separation properties of the ceramic membrane by one-step pasting method.

the support, the defects of the support have no effect on the properties of the separation layer. With increasing the membrane thickness from 90 to 130 μm , the permeability of the membrane was decreased and the pore-size did not changed considerably. This further indicates that the membrane thickness doesn't have an effect on the pore-size of the membrane and the pore-size depends on the particle size of the materials by one-step pasting method. The ceramic membrane with a thickness of 90 μm and a pore-size of 0.141 μm was prepared after sintering at 1,350°C for 2 h. The efficiency of the membrane for separating the 130 nm particles is performed very well.

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