

Fundamental parameters of nanoporous filtration membranes

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Abstract. The design theory for nanoporous filtration membranes needs to be established. The present study shows that the performance and technical advancement of nanoporous filtration membranes are determined by the fundamental parameter I (in the unit $Watt^{1/2}$) which is formulated as a function of the shear strength of the liquid-pore wall interface, the radius of the filtration pore, the membrane thickness, and the bulk dynamic viscosity of the flowing liquid. This parameter determines the critical power loss on a single filtration pore for initiating the wall slippage, which is important for the flux of the membrane. It also relates the membrane permeability to the power cost by the filtration pore. It is shown that for biological cellular membranes its values are on the scale $1.0E-8Watt^{1/2}$, for mono-layer graphene membranes its values are on the scale $1.0E-9Watt^{1/2}$, and for nanoporous membranes made of silica, silicon nitride or silicon carbonized its values are on the scale $1.0E-5Watt^{1/2}$. The scale of the value of this parameter directly measures the level of the performance of a nanoporous filtration membrane. The carbon nanotube membrane has the similar performance with biological cellular membranes, as it also has the value of I on the scale $1.0E-8Watt^{1/2}$.

Keywords: filtration; membrane; nanopore; permeability; power loss; water

1. Introduction

Nanoporous filtration membranes have been in fast development particularly in manufacturing for their important applications in super filtration such as in hemofiltration, water purification, DNA analysis, and biosensors etc (Ariono *et al.* 2018, Baker and Bird 2008, Bottino *et al.* 2011, Brown *et al.* 1975, El-ghizel *et al.* 2019, Elizabeth *et al.* 2012, Fissel *et al.* 2009, Jackson and Hillmyer 2010, Jin *et al.* 2019, Sanjay *et al.* 2021, Sofos 2021, Stavrogiannis *et al.* 2022, Surwade *et al.* 2015). The fluxes of these membranes are expected to be as high as possible, though it seems very challenging to yield a high flux from a nanopore. The thin membrane is required for high flux, while the mechanical strength of the membrane demands the membrane thickness. For achieving these two goals, the composite membranes have been developed, consisting of both the very thin nanoporous filtration membrane and the microporous supporting layer (Cadotte *et al.* 1980, Tiraferri *et al.* 2011, Yang *et al.* 2006, Yip *et al.* 2010). The third concern is the power cost on a nanoporous filtration membrane for driving the liquid to flow through, which ought to be as small as possible for energy conservation. It is critical to combine limited power cost and high flux from a nanoporous filtration membrane. Although the manufacturing of nanoporous filtration membranes has evolved (Ariono *et al.* 2018, Baker and Bird 2008, Bottino *et al.* 2011, Brown *et al.* 1975, El-ghizel *et al.* 2019, Elizabeth *et al.* 2012, Fissel *et al.* 2009, Jackson

and Hillmyer 2010, Jin *et al.* 2019, Sanjay *et al.* 2021, Surwade *et al.* 2015), theoretical researches still remain open.

In animals and plants, water permeates through cells smoothly. For example, the human cell membrane contains the nanopore with the radii between 0.2nm and 0.5nm for water to flow through (Lin and Zhang 2022). While a human cellular connexon has the cylindrical nanotube with the diameter around 1.5nm for water flow (Wang and Zhang 2021). The classical fluid mechanics obviously can not explain the enhanced water permeation through biological cellular membranes, as it calculates large flow resistances of the biological membranes (Pinkus and Sternlicht 1961).

In experiments or molecular dynamics simulations (MDS), water has been found to flow through 1nm carbon nanotubes with flux several orders higher than the classical hydrodynamic flow theory calculation (Holt *et al.* 2006, Kannam *et al.* 2013, Majumder *et al.* 2005, Wang *et al.* 2012). These ultra fast water transports were ascribed to the water slippage on the nanopore wall. The nanoporous filtration membranes with higher fluxes are in development. Recently, Itoh *et al.* (2022) showed by experiment the fast water permeation through the fluorinated nanorings with the diameters between 0.9nm and 1.9nm, two orders higher than those through carbon nanotubes. They explained the fast flow by the breaking of the water clusters nearby the pore wall owing to the hydrophobic pore wall. However, by experiments, Nair *et al.* (2012) suggested that water should be blocked and unable to flow through the separation below 0.6nm because one mono water molecule layer even can not be filled in. Nevertheless, they detected the unexpectedly fast water flow through the nano slit pore with the clearance around 1nm made from graphene oxide. It was also

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observed that the graphene membrane with mono layer or even multilayer has good permeability for water (Cohen-Tanugi *et al.* 2016, Huang *et al.* 2015, Jang *et al.* 2017, Kim *et al.* 2016, Surwade *et al.* 2015). It was found that hydrophobic nanopores such as carbon nanotubes, fluorinated nanorings and graphene nanopores have small flow resistances to water, while hydrophilic nanopores such as alumina nanopores have much higher flow resistances (Koklu *et al.* 2017).

Researchers have tried to improve the flux of nanoporous filtration membranes by using conical pores (Lan *et al.* 2015, Li *et al.* 2004, Harrell *et al.* 2006, Zhang 2019a), optimized complex pores (Zhang 2018) and optimized nanopore trees (Zhang 2019b, c) or by ultimately reducing the membrane thickness (Surwade *et al.* 2015) and covering hydrophobic coatings on the nanopore (Itoh *et al.* 2022). However, the theoretical study on the mechanism of the permeation of nanoporous filtration membranes is still scarce, though a lot of experiments on such membranes have been made. It is of significant interest to theoretically explore the performance of nanoporous filtration membranes.

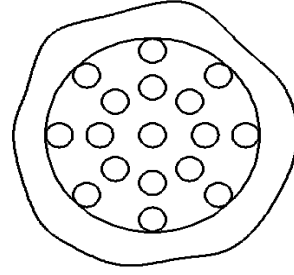
In this paper we have developed the mathematical equation for formulating the flow enhancement factor through super filtration nanopores based on the wall slippage and the nanoscale flow equation (Zhang 2016). In the case of the wall slippage, the permeability through a nanopore is directly related to the power cost by the flow in the nanopore. We obtained the critically important fundamental parameter I for characterizing the performance of a nanoporous filtration membrane. This parameter measures both the permeability and the energy efficiency of the membrane. It incorporates the important parameters for permeation observed in experiments such as the radius of the nanopore, the membrane thickness and the liquid-pore wall interfacial shear strength. Our theory shows that the dependences of the membrane permeation on these parameters all agree with the experimental results; the permeability of a nanoporous membrane is actually finally determined by the parameter I . We calculated the values of I for some typical membranes and made evaluation of the technical performances of these membranes. The evaluated merits of nanoporous filtration membranes are directly evident from the present study.

2. Methods

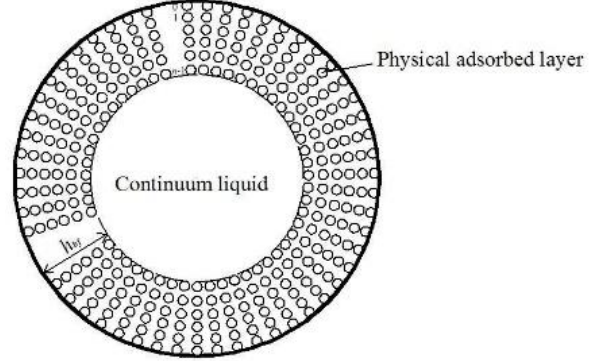
When water flows through the nanopore with the diameter below 1nm such as through biological cellular membranes, the water molecules inside the nanopore behave as non-continuum as shown in Fig.1(a) and their mass flow rate through the nanopore is calculated as (Zhang 2017):

$$q_m = \pi \rho_{bf}^{eff} u_s R^2 + \frac{\pi \rho_{bf}^{eff} S R^4}{4 \eta_{bf}^{eff}} \frac{\partial p}{\partial x} \quad (1)$$

where R is the radius of the nanopore, u_s is the wall slipping velocity, ρ_{bf}^{eff} and η_{bf}^{eff} are respectively the average density and the effective viscosity of the water



(a) The non-continuum flow in a small nanopore (with the diameter on the 1nm scale or less) (Lin and Zhang 2022)



(b) The multiscale flow in the bigger nanopore (with the diameter on the scales of 10nm or 100nm) (Li and Zhang 2021)

Fig. 1 Liquid flow in the nanopores with different sizes

across the pore radius, S is the parameter accounting for the water non-continuum effect, p is the pressure driving the water flow, and x is the coordinate in the axial direction of the nanopore.

Generally, whenever the wall slippage is present or not, the liquid mass flow rate through a small nanopore can be calculated from Eq. (1). When $u_s \neq 0$, there is the wall slipping velocity and the wall slippage occurs. When $u_s = 0$, the wall slipping velocity vanishes and there is no wall slippage. Whether the wall slippage occurs or whether the magnitude of the wall slipping velocity u_s should depend on the adhesive strength of the liquid molecules on the nanopore wall, the magnitude of the exerted external driving pressure, the axial length of the nanopore, and the diameter of the nanopore?

The present analysis is focused on the non-continuum flow occurring in the whole nanopore with a very small diameter as shown in Fig.1(a). For the nanopore with bigger sizes such as with the diameter on the scales of 10nm or 100nm, the liquid (or water) flow in the nanopore may be multiscale consisting of both the non-continuum molecular-scale adsorbed layer flow and the intermediate continuum liquid flow as shown in Fig. 1(b) (Zhang 2020). The analysis for the multiscale flow in the nanopore shown in Fig. 1(b) is actually different from the present analysis (Zhang 2020).

According to the classical hydrodynamic flow theory, which neglects the wall slippage, the mass flow rate through the nanopore in Fig. 1(a) is calculated as:

$$q_{m,conv} = -\frac{\pi \rho R^4}{4 \eta} \frac{\partial p}{\partial x} \quad (2)$$

where ρ and η are respectively the bulk density and the bulk viscosity of the flowing liquid.

Define the flow enhancement factor i.e. the permeation factor as:

$$r_m = \frac{q_m}{q_{m,conv}} \quad (3)$$

Experiments have identified that the values of r_m can be $10^1 \sim 10^7$, depending on the radius of the nanopore, the axial length of the nanopore i.e., the thickness of the membrane and the hydrophobicity of the nanopore wall (Holt *et al.* 2006, Itoh *et al.* 2022, Kannam *et al.* 2013, Majumder *et al.* 2005).

By using the liquid-pore wall interfacial limiting shear strength model for the wall slippage (Zhang 2014), which interprets the wall slippage as the result of the shear stress on the pore wall exceeding the liquid-pore wall interfacial shear strength, the wall slipping velocity is expressed as (Wang and Zhang 2021):

$$u_s = \frac{(POW - POW_{cr})\theta_\tau}{\pi\tau_s l R} \quad (4)$$

where POW is the power cost in the single nanopore, POW_{cr} is the critical power loss on a single nanopore for initiating the wall slippage, τ_s is the liquid-pore wall interfacial shear strength, l is the pore axial length i.e. the membrane thickness, and θ_τ is the correction factor for the liquid-pore wall interfacial shear stress due to the liquid non-continuum effect.

POW_{cr} is equated as (Wang and Zhang 2021):

$$POW_{cr} = -\frac{\pi S}{4\theta_\tau^2 C_y} I^2 \quad (5)$$

where

$$I = \tau_s R \sqrt{\frac{l}{\eta}} \quad (6)$$

and

$$C_y = \frac{\eta_{bf}^{eff}}{\eta} (>1) \quad (7)$$

Lower the values of τ_s , R or l , smaller the value of POW_{cr} ; reducing τ_s and R has the specially significant effect on dropping POW_{cr} as POW_{cr} is directly proportional to τ_s^2 and R^2 ; also the effect of the reduction of τ_s (i.e., the hydrophobicity of the nanopore wall) is the same with the effect of the reduction of R in yielding the low critical power cost for initiating the wall slippage. That is why it is very easy for the wall slippage to occur in very small nanopores (with the radius on the 1nm scale) as found in experiments or molecular dynamics simulations (Holt *et al.* 2006, Kannam *et al.* 2013, Majumder *et al.* 2005, Wang *et al.* 2012).

For the case of the wall slippage, by using the following relation (Wang and Zhang 2021):

$$\frac{\partial p}{\partial x} = -\frac{\tau_s}{R\theta_\tau} \quad (8)$$

substituting Eq. (5) into Eq. (4) and further substituting Eq. (4) into Eq. (1) yields:

$$r_m = \frac{4C_q\theta_\tau^2 POW}{\pi I^2}, \text{ for } POW \geq POW_{cr} \quad (9)$$

where

$$C_q = \frac{\rho_{bf}^{eff}}{\rho} (>1) \quad (10)$$

Eq. (9) shows that for a given nanoporous membrane, the permeation factor r_m of the membrane is directly proportional to the power cost on the membrane; or for a given power cost by the membrane, r_m is inversely proportional to I^2 . It is clearly manifested that reducing τ_s and R has the strongest effect on increasing the permeation through a nanoporous membrane, though the reduction of the thickness (l) of the membrane also increases the permeation.

For hydrophobic nanopores such as in biological cellular membranes, graphene membranes, carbon nanotubes, and fluorinated nanorings, $C_q \approx 1$, $C_y \approx 1$, $\theta_\tau \approx 1$, and $S \approx -1$. For these nanopores, where the wall slippage very easily occurs, it is simply written that:

$$POW_{cr} = \frac{\pi}{4} I^2 \quad (11)$$

and

$$r_m = \frac{POW}{POW_{cr}}, \text{ for } POW \geq POW_{cr} \quad (12)$$

Eqs. (11) and (12) show that the parameter I is the fundamental parameter of a nanoporous filtration membrane, and it determines the permeability of the membrane (for a given POW) or the energy cost on the membrane (for a given r_m) in the case of the wall slippage (i.e., for $POW \geq POW_{cr}$).

3. Characteristics of some typical membranes

It is noteworthy to observe the smooth permeation of water through human cell membranes and through plant cell walls. How much energy cost is for the water permeation through a single biological cell? This is a fundamental question as it relates to maintaining the ordinary life of an animal or a plant. Such an energy cost must be tiny enough, otherwise a creature can not provide sufficient energy for so many cells inside it. Then, what is the relation between the energy cost by a cell and the permeation of water through a cell? Such a question is also very important for artificially manufactured nanoporous filtration membranes, as it addresses on the flux and the energy efficiency of the membrane, which are two critically important issues. Eqs. (11) and (12) give the final mathematical expression for the relation between the permeation factor of a small nanopore and the corresponding power cost by the nanopore in the case of the wall slippage. According to the present analytical results, for addressing the above mentioned questions, we calculated the values of the parameter I for some popular nanoporous membranes and show them in

Table 1 Values of the parameters I and POW_{cr} for some popular membranes for water permeation

Membrane	I ($Watt^{1/2}$)	POW_{cr} (Watt)
Human cell membrane	1.3E-8	1.3E-16
Human cellular connexon	2.0E-8	3.1E-16
Plasmodesmata (plant)	3.3E-8	8.6E-16
Blood capillary wall	3.4E-6	9.1E-12
Mono-layer graphene membrane ($R=0.5nm$)	2.9E-9	6.6E-18
Fluorous nanoring of Itoh <i>et al.</i> (2022)	1.0E-9	7.9E-19
Carbon nanotube of Majumder <i>et al.</i> (2005) ($R=3.5nm$)	5.3E-9	2.2E-17
Carbon nanotube of Kannam <i>et al.</i> (2013) ($R=0.4nm\sim 5nm$)	1.0E-8	7.9E-17
Carbon nanotube of Holt <i>et al.</i> (2003) ($R=0.65nm\sim 1nm$)	1.0E-7	7.9E-15
γ alumina nanoporous filtration membrane (Koklu <i>et al.</i> 2017) ($R=5nm\sim 20nm$, $l = 50\mu m$)	2.5E-4~1.0E-3	4.9E-8~7.9E-7
Silica, silicon nitride or silicon carbonized membranes ($R=0.5nm$, $l = 50\mu m$)	1.0E-5	7.9E-11
Membrane	I ($Watt^{1/2}$)	POW_{cr} (Watt)

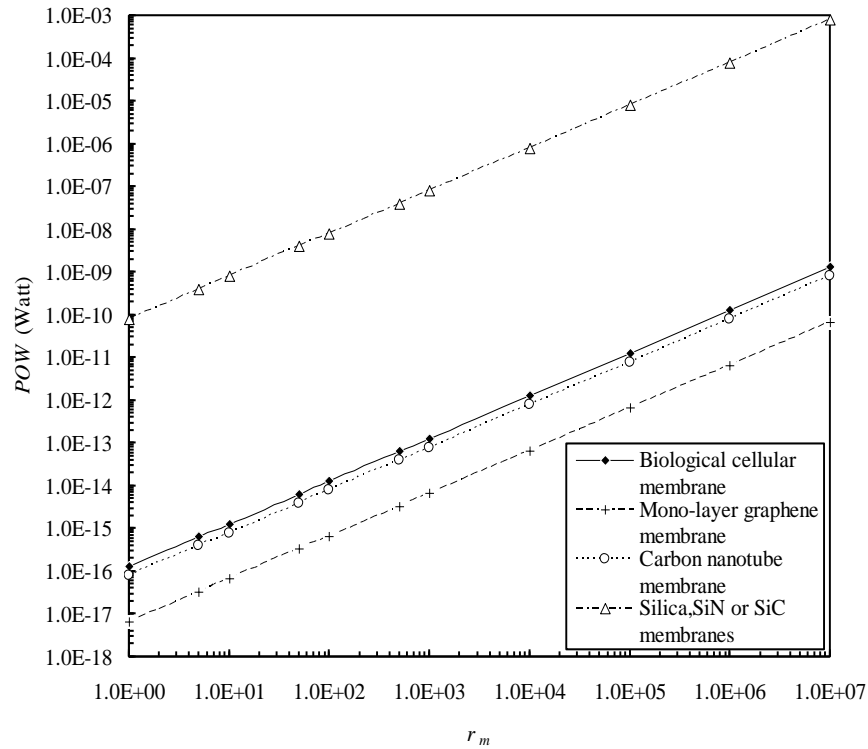
Fig. 2 The POW versus r_m curves for different membranes for water permeation, $R=0.5nm$

Table 1. The values of I for biological cellular membranes are on the scale $1.0E - 8Watt^{1/2}$. The corresponding values of POW_{cr} are on the scale $1.0E - 16Watt$, and should be low enough for maintaining the normal activity of a live cell. Truly, the functions of biological membranes are very good for fast permeation of water just with very limited power cost. The performances of carbon nanotube membranes are very similar with those of biological cellular membranes because of their values of I and POW_{cr} respectively on the same scales. This was also mentioned in the earlier experimental study (Majumder *et al.* 2005). The advanced artificial membranes like mono-layer graphene

nanoporous membrane (Surwade *et al.* 2015) and fluorous nanoring membrane (Itoh *et al.* 2022) have the values of the parameter I on the scale $1.0E - 9Watt^{1/2}$. The functions of these artificial membranes are obviously more advantageous over those of biological cellular membranes. It is thus realizable to produce artificial filtration membranes much better than biological cellular membranes both in the permeability and the energy efficiency by degrading the parameter I . The development of the excellent nanoporous filtration membranes in the future should also be in this direction. However, the currently commonly used membranes made from silica, silicon

nitride or silicon carbonized appear far inferior to biological cellular membranes in both the permeability and the energy efficiency, as their values of I are on the scale $1.0E - 5Watt^{1/2}$. The γ alumina nanoporous filtration membrane is even worse in the performance because its value of the parameter I is on the scales $1.0E - 4Watt^{1/2} \sim 1.0E - 3Watt^{1/2}$. It can be quantitatively evaluated that the water flow rates through these hydrophilic nanoporous membranes are 6-10 orders smaller than those through biological cellular membranes.

Fig. 2 shows the relations between the power cost POW by a single nanopore and the flow enhancement factor r_m for different nanoporous membranes for water permeation ($R=0.5nm$) if the wall slippage occurs. The POW versus r_m curve for biological cellular membrane is very close to that for carbon nanotube membrane. This indicates the close performances of these two membranes in both the permeability and the energy efficiency. The mono-layer graphene membrane has a better performance because of its POW versus r_m curve located on the bottom. The silica, SiN or SiC membranes are the worst in the performance because of their POW versus r_m curves located on the top.

The present study provides the method to evaluate the performance and technical merit of nanoporous membranes for liquid permeation. Naturally, biological cellular membranes are appropriate for water permeation because of its value of the parameter I on the scale $1.0E - 8Watt^{1/2}$. Most current commercial membranes are far inferior to biological membranes because of their values of I several orders higher. The artificial membrane made from carbon nanotubes can replace biological cellular membrane because of their close I values. The current most advanced artificial membranes give the value of I on the scale $1.0E - 9Watt^{1/2}$. Further progresses should be possible in artificial membrane development by more lowering the parameter I (mainly by reducing τ_s).

4. Conclusions

The paper directly relates the flow enhancement factor r_m i.e., the permeation factor of nanoporous membranes to the power cost POW by a single nanopore by using the fundamental parameter I which is defined as $I = \tau_s R \sqrt{l/\eta}$ (in the unit $Watt^{1/2}$), where τ_s is the shear strength of the liquid-pore wall interface, R is the radius of the nanopore, l is the membrane thickness, and η is the bulk dynamic viscosity of the flowing liquid. The relation equation reads:

$$r_m = \frac{POW}{POW_{cr}}, \text{ for } POW \geq POW_{cr} \quad (12)$$

where POW_{cr} is the critical power loss on a single nanopore for initiating the wall slippage and for hydrophobic nanopores $POW_{cr} = \pi I^2/4$. Eq. (12) ascribes the flow enhancement in hydrophobic nanoporous membranes as experimentally observed to the wall slippage; the flow enhancement factor is determined only by the two parameters POW and I , the latter being the intrinsic fundamental parameter of the membrane.

Both the permeability and the energy efficiency of

nanoporous membranes are evaluated by Eq. (12) if the wall slippage occurs. They are essentially determined by the parameter I . Lower the value of I , better the overall performance of the membrane. The future direction of developing nanoporous filtration membranes should be further lowering the value of I by fabricating super hydrophobic nanopores.

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