

# Permeation characteristics of fresh water in hollow fiber membrane module for pressure retarded osmosis

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## Abstract

Pressure Retarded Osmosis (PRO) is expected to the new power generation system. The performance of the PRO module is closely relating to the permeation characteristics. In this paper, when fresh water flow rate is changed, 4 factors related to the reduction of permeation flow rate are researched with experiment. These 4 factors are concentration of solute in fresh water, salt leakage, concentration polarization, and dissipation of fresh water. The characteristics of the fresh water flow and permeation are studied for the hollow fiber membrane module used in PRO system. It is cleared that the dissipation of fresh water and the concentration polarization in hollow fiber largely influence to the reduction of permeation flow rate in the case of the low fresh water flow rate. Concentration of solute in fresh water, salt leakage influence to the reduction of permeation flow rate in the case of high fresh water flow rate.

**Key words** : Pressure Retarded Osmosis(PRO), Forward Osmosis(FO), Concentration polarization, Hollow fiber, Fresh water flow rate, Hollow fiber membrane module

## 1. Introduction

There are a lot of conversion power generation techniques with sustainable energy like the solar, wind, biomass, and ocean energy. The Pressure Retarded Osmosis (PRO) is also one of these power generations with sustainable energy and is suggested by S.Loeb (Loeb and Norman, 1975). PRO which utilizes the osmosis phenomena is a method to convert the salinity gradient energy to the hydroelectric energy. The fresh water and the pressurized salt water are separated by semi-permeable membranes in membrane module. The fresh water permeates to salt water by osmosis and is mixed with pressurized salt water. In PRO, sea water is used as salt water, and the osmotic pressure of sea water is about 3.0[MPa] that corresponds to 300[m] hydrostatic pressure head. Therefore, pressure of the fresh water is increased without the pump power. This pressurized fresh water generates the electric power. So the permeation flow rate is important to generate power. In 1981, Lee et al. mentioned that PRO is technically feasible, but not economically viable with currently available membrane (Lee et al.,1981). This means that the permeation flow rate is low and the required power to supply the salt water and fresh water is large.

Honda had evaluated performance of interesting PRO system set under the sea level (Honda, 1990). Yasugahira et al. had predicted the generated power with main river in Japan (Yasugahira et al., 2003). Shiono et al. had demonstrated PRO performance with fresh water from river (Shiono et al., 2004). Tan and Ng established prediction model of the power osmotic power generates (Tan and Ng, 2008). Statkraft which is a leading company in hydropower internationally and Europe's largest generator of renewable energy had attempted the PRO prototype (Gerstandt et al., 2008) and obtained a few output from the sea water (Halper, 2010). Saito et al. have developed the PRO system from 2003 and obtained the net output for the concentrated sea water (Saito et al., 2012). These mean the practical system is

not realized yet.

In the field of membrane, two principal problems have been researched to reduce the permeation flow rate. These problems are “Concentration polarization” and “Membrane fouling”. Concentration polarization cause salinity gradient in and on the membrane. This phenomenon leads to reduction of osmotic pressure difference through the membrane. As result, permeation flow rate become low. Sagiv et al. had evaluated the effect of ICP and ECP on permeation performance of FO flat membrane with CFD analysis (Sagiv et al., 2015). Membrane fouling is the phenomenon which is caused by accumulating of solute in and on membrane. This phenomenon refuses to permeate the water molecule. As result, permeation flow rate become low. Many researchers have worked on this phenomenon so far.

Nomenclature	
$Q_w$	Permeation flow rate [ $\text{m}^3/\text{s}$ ]
$J_w$	Permeation flux [ $\text{m}^3/(\text{m}^2 \cdot \text{s})$ ]
$A$	Permeation coefficient of water molecules [ $\text{m}/(\text{Pa} \cdot \text{s})$ ]
$\Delta\pi$	Osmotic pressure difference through the membrane [Pa]
$\Delta P$	Hydrodynamic pressure difference through the membrane [Pa]
$P_d$	Pressure of salt water [Pa]
$P_f$	Pressure of fresh water [Pa]
$S_{mem}$	Surface area of membrane [ $\text{m}^2$ ]
$M$	Mole concentration [ $\text{mol}/\text{m}^3$ ]
$R$	Ideal gas constant [ $\text{kg} \cdot \text{m}^2/(\text{K} \cdot \text{mol} \cdot \text{s}^2)$ ]
$T$	Absolute temperature [K]
$J_s$	Permeation flux of NaCl [ $\text{kg}/(\text{m}^2 \cdot \text{s})$ ]
$B$	Permeation coefficient of NaCl [m/s]
$C_{d,m}$	Mass concentration of salt water on membrane surface [ $\text{kg}/\text{m}^3$ ]
$C_{f,i}$	Mass concentration of fresh water at interface between support layer and active layer [ $\text{kg}/\text{m}^3$ ]
$R_{mem}$	Rejection rate of NaCl [-]
$K$	Diffusion resistance [s/m]
$\pi_{d,m}$	Osmotic pressure of salt water on membrane surface [Pa]
$\pi_{f,m}$	Osmotic pressure of fresh water on membrane surface [Pa]
$S$	Structure parameter [m]
$D$	Diffusion coefficient of NaCl [ $\text{m}^2/\text{s}$ ]
$\tau$	Tortuosity of support layer [-]
$t$	Thickness of membrane [m]
$\varepsilon$	Porosity of support layer [-]
$\pi_{d,b}$	Osmotic pressure of salt water in bulk [Pa]
$\pi_{f,b}$	Osmotic pressure of fresh water in bulk [Pa]
$kd$	Mass transfer coefficient [m/s]
$Sh$	Sherwood number [-]
$dh$	Hydrodynamic diameter [m]
$Re$	Reynolds number [-]
$Sc$	Schmidt number [-]
$L$	Length of hollow fiber [m]
$d_i$	Inner diameter of hollow fiber [m]
$d_o$	Outer diameter of hollow fiber [m]
$q$	Fresh water flow rate in a hollow fiber [ $\text{m}^3/\text{s}$ ]
$l$	Length of hollow fiber [m]
$P_0$	Pressure of fresh water at inlet [Pa]
$\nu$	Dynamic viscosity [ $\text{m}^2/\text{s}$ ]
$\rho$	Density [ $\text{kg}/\text{m}^3$ ]
$q_0$	Fresh water flow rate at inlet [ $\text{m}^3/\text{s}$ ]
$P_s$	Pressure of salt water [Pa]

But in fact, membrane module is used in the plant. It's not only membrane. So improving of membrane module performance should be discussed. Then, increase of fresh water concentration, dissipation of fresh water, and deviation flow exist in module as factor for reduction of permeation flow rate. Increase of fresh water concentration is caused by concentration of solute in fresh water, and by leakage of salt. Dissipation of fresh water means that fresh water flow rate is zero. When the length of hollow fiber in module is focused, hollow fiber is so long. So, fresh water in hollow fiber is gradually decreased by permeation. Hollow fiber where dissipated fresh water can't cause osmosis. In this length, permeation flow rate is zero.

But these three factors are not researched yet. So increase of fresh water concentration, dissipation of fresh water, and deviation flow should be discussed with two principal problems : concentration polarization and membrane fouling. In this experiment, test duration is short. Additionally, RO water is used as fresh water in this experiment. So, we assume that effect of fouling on reduction of permeation flow rate is so small.

In this paper, the effect of concentration polarization, increase of fresh water concentration, dissipation of fresh water, and deviation flow on permeation performance is evaluated.

## 2. Hollow fiber and membrane module

In this experiment, we use the hollow fiber as semi-permeable membrane shown in Fig.1. This hollow fiber is polymer membrane made by cellulose tri-acetate (CTA). And the shape of hollow fiber is cylindrical, 60[ $\mu\text{m}$ ] inner diameter and 160[ $\mu\text{m}$ ] outer diameter. The structure of hollow fiber is asymmetric. Outer surface of hollow fiber is active layer with dense pore, and inner layer is support layer which give active layer strength. Permeation flow rate  $Q_W$  is expressed with permeation flux  $J_W$  as eq.(1).

$$Q_W = J_W \cdot S_{mem} = A\{(\pi_{d,b} - \pi_{f,b}) - (P_d - P_f)\}S_{mem} \quad \dots (1)$$

The structure of hollow fiber membrane module is as shown in Fig.2. In slash area, the hollow fibers are wound around center pipe which located at shaft-center. Salt water inflow from right-center, and flow in the center pipe. Some holes are made on the side surface of center pipe. So, salt water can flow out from center pipe to hollow fiber element area. The fresh water inflow from upper-right, and flow to each fiber from end face which is fixed with opening of the hollow fiber. The drain water which don't permeate to salt water is drained at left side. And, the dimension and other parameter value of membrane module and hollow fiber used in this experiment are shown in Table1.

Table 1 Main dimensions of 5-inch membrane module

Module diameter	130 [mm]
Module length	641.8 [mm]
Total area of hollow fiber membrane	58 [m <sup>2</sup> ]
Number of hollow fiber	180,000
Outer diameter of hollow fiber	160 [ $\mu\text{m}$ ]
Inner diameter of hollow fiber	60 [ $\mu\text{m}$ ]

## 3. Apparatus and conditions

The experimental system is shown in Fig.3. This experimental system is consisted of salt water flow path which inflow the salt water flow side of membrane module, fresh water flow path which inflow the fresh water flow side of membrane module, mixed water flow path which mixed water made by salt water and fresh water as permeation water outflow from membrane module, and drain flow path which fresh water do not permeate outflow from membrane module.

In salt water flow path and fresh water flow path, tank, pump, flow meter, and pressure gauge are installed. In the mixed water flow path, pressure gauge, a flow meter, and an electrical conductivity meter are installed. In drain water flow path, electric conductivity meter is installed. Permeation flow rate is obtained by difference of the mixed water flow rate and salt water flow rate. We washed with tap water to remove salt in each experiment. The experimental conditions are shown in Table 2.

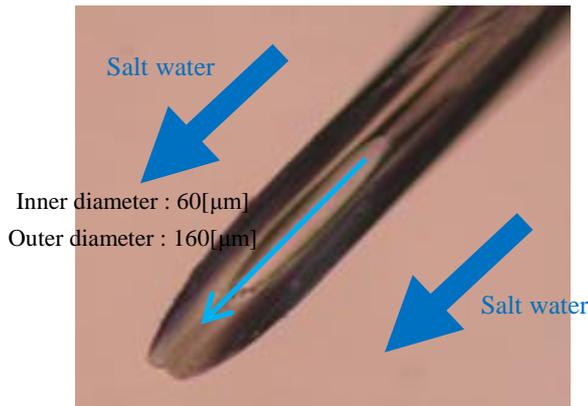


Fig.1 Photograph of hollow fiber membrane

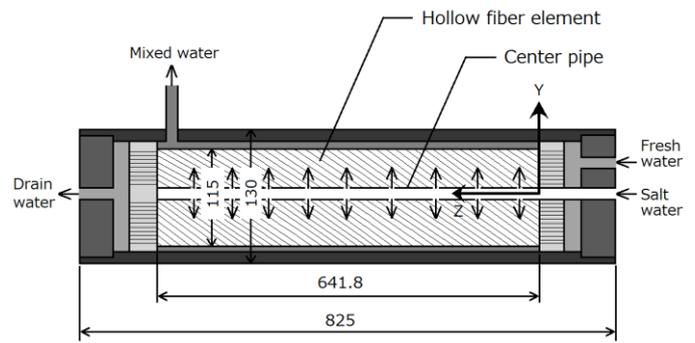


Fig.2 Schematic of cross-section of membrane module

Table 2 Experimental conditions

Salt water flow rate	1.0 [l/min]
Fresh water flow rate	1.0, 2.0, 3.0, 3.5 [l/min]
Salt concentration	0.5 [%]
Solute concentration of fresh water	0.0015 [%]
Salt water pressure	0.08, 0.18, 0.30, 0.38 [MPa]
Fresh water pressure	0.08, 0.18, 0.30, 0.38 [MPa]

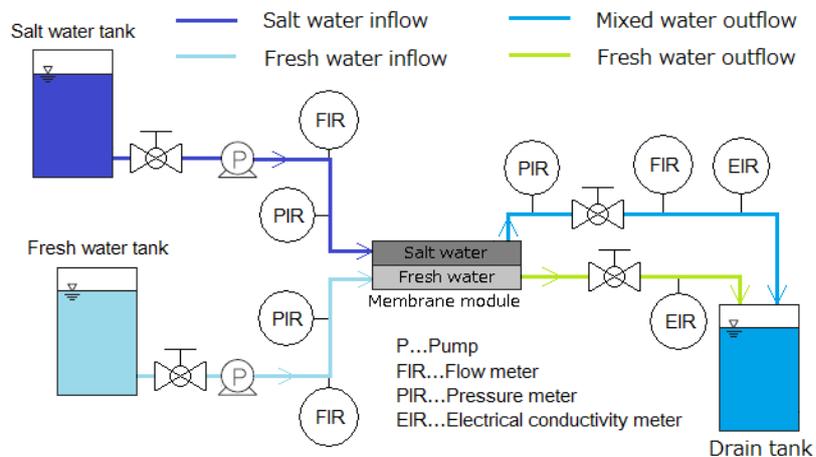


Fig.3 Schematic diagram of experimental system

## 4. Results and discussion

### 4.1. Comparison of permeation flow rate

In this paper, permeation characteristics are investigated at the case of low fresh water flow rate. Permeation flow rate for fresh water is shown in Fig.4. Solid line indicates theoretical value. Theoretical permeation flux  $J_w$  [ $m^3/(m^2 \cdot s)$ ] is expressed by the eq.(1). The osmotic pressure  $\pi$  in eq.(1) can be calculated by the van't Hoff equation (2).

$$\pi = 2MRT \quad \dots (2)$$

where  $M$  [ $mol/m^3$ ] is the molar concentration,  $R$  [ $kg \cdot m^2/(s^2 \cdot kmol \cdot K)$ ] is the gas constant,  $T$  [K] is the absolute temperature.

At the case of 3.5[l/min] fresh water flow rate, the permeation flow rate, the averaged salt concentration in the module, pressure of salt water, pressure of fresh water, and the total area of the hollow fiber membrane are applied to eq.(1). Then, the permeation coefficient of water  $A=2.5E-6$  [m/(s•MPa)] is obtained. The reason for applying this  $A$  value is that flow state at 3.5[l/min] fresh water flow rate is ideal state. The fresh water flow in the hollow fiber is increased. This means that increase of fresh water concentration is sufficiently low, and dissipation of the fresh water in the hollow fiber is not considered. In addition, the difference between the theoretical value and experimental value indicates possibility. This difference can't be evaluated quantitatively. Furthermore, the velocity of fresh water increase, as shown in the section of the "Effect of concentration polarization", the Reynolds number of the fresh water increases, the external concentration polarization is reduced. When we focus on the permeation flow rate at the lower fresh water supply in Fig.4, the difference between permeation flow rate of experiment and calculated permeation flow rate is large. Then, the decrease of permeation flow rate is caused with four factors as follows. 1: Concentration of solute in fresh water, 2: Leakage of salt from salt water to fresh water, 3: Concentration polarization, and 4: Dissipation of fresh water.

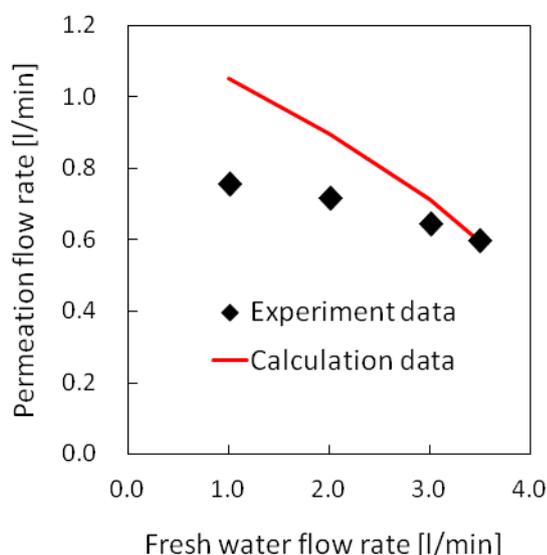


Fig.4 Permeation flow rate of experiment and theoretical value

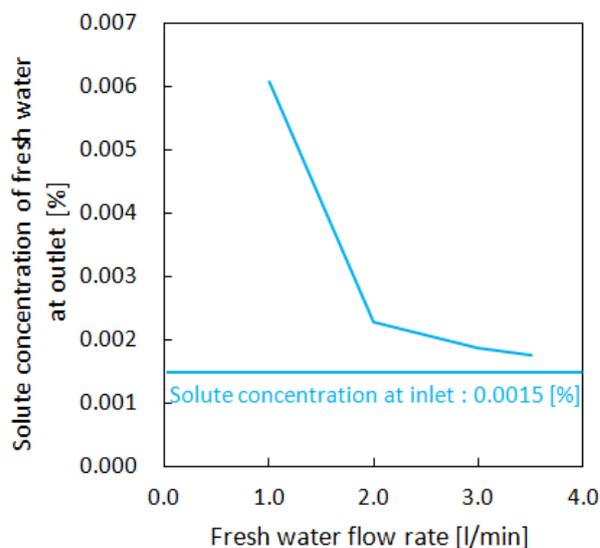


Fig.5 Solute concentration of fresh water at outlet

#### 4.2 Effect of fresh water concentration

In this section, the effect of factor-1, concentration of solute in fresh water, is clarified. The solute concentration of fresh water at outlet is calculated with amount of solute in fresh water at inlet. The solute concentration against fresh water flow rate at outlet of fresh water can be calculated with amount of solute in fresh water at inlet.

The result of this calculation is shown in Fig.5. The horizontal solid line indicates the solute concentration of fresh water at inlet. At the 2.0[l/min] fresh water flow rate, solute concentration of fresh water decrease rapidly. Pressure difference between salt water and fresh water at outlet side of membrane module related to this phenomenon. At over 2.0[l/min] fresh water flow rate, pressure difference at outlet side is large, because pressure drop of fresh water is very large. Then permeation flow rate is decreased as shown eq.(1). This means that solute concentration in fresh water decrease rapidly at 2.0[l/min] fresh water flow rate. Therefore, at the low fresh water flow rate, it is easy to cause concentration by permeation. But this increase of solute concentration is small compared to salt concentration of salt water at inlet. This means that effect on decrease of permeation flow rate is small.

#### 4.3 Effect of salt leakage

In this section, the effect of salt leakage is clarified. The semi-permeable membrane ideally does not permeate the solute. But, a little salt can permeate from salt water to fresh water. The salt flux  $J_s$  [kg/(m<sup>2</sup>•s)] is expressed as below.

$$J_s = B(C_{d,m} - C_{f,i}) \quad \dots (3)$$

where  $B$  [m/s] is permeation coefficient of salt,  $C_{d,m}$  [kg/m<sup>3</sup>] is salt concentration on the membrane surface of salt water side,  $C_{f,i}$  [kg/m<sup>3</sup>] is salt concentration at the interface between support layer and active layer. The permeation coefficient of salt is defined as below.

$$B = \frac{1 - R_{mem}}{R_{mem}} J_w \quad \dots (4)$$

$R_{mem}$  [-] is the salt rejection. We obtained the result shown in Fig.6 with this calculation theory. At the low fresh water flow rate, salt permeation flux  $J_s$  increase. At the low fresh water flow rate, solute concentration of fresh water increase largely. Therefore, at the low fresh water flow rate, it is easy to cause the salt leakage. This permeation flux corresponds to salt concentration  $1.1 \times 10^{-8}$  to  $6.7 \times 10^{-8}$  [%]. This increase of solute concentration is very small compared to salt concentration of salt water at inlet. This means that effect on decrease of permeation flow rate is very small.

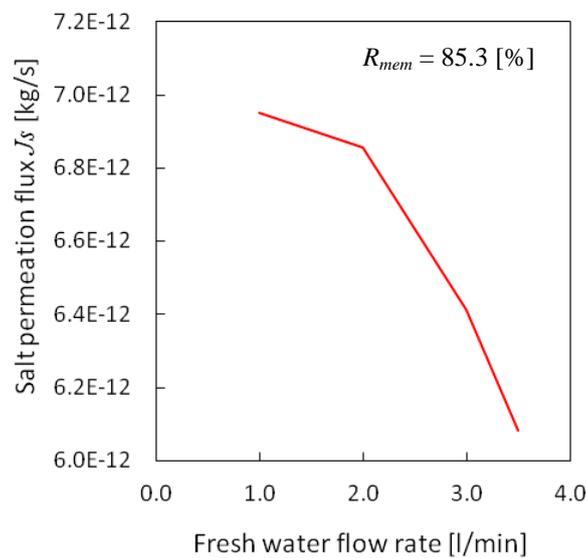


Fig.6 Permeation salt flux against the fresh water flow rate

#### 4.4 Effect of concentration polarization

In this section, the effect of factor-3, concentration polarization, on salt concentration around membrane surface is discussed. McCutcheon and Elimelech derive the theory of external concentration polarization (ECP) which cause at outside of hollow fiber and of internal concentration polarization (ICP) which cause in support layer (McCutcheon and Elimelech, 2006). The permeation flux considered ICP is expressed as eq.(5).

$$J_w = \frac{1}{K} \ln \frac{B - J_w + \pi_{d,m}}{B + A\pi_{f,m}} \quad \dots (5)$$

where  $K$  [s/m] is diffusion resistance in support layer,  $B$  [m/s] is permeation coefficient of salt,  $\pi_{d,m}$  [Pa] is osmotic pressure at salt water side on membrane surface,  $\pi_{f,m}$  [Pa] is osmotic pressure at fresh water side on membrane surface. Additionally, diffusion resistance  $K$  which indicate the level of solute diffusion in support layer is defined as follow eq.(6).

$$K = \frac{S}{D} = \frac{\tau t}{D\varepsilon} \quad \dots (6)$$

Where  $D$  [m<sup>2</sup>/s] is diffusion coefficient,  $S$  [m<sup>2</sup>] is structure parameter for the membrane. This structure parameter  $S$  is represented by porosity of support layer  $\varepsilon$ [-], tortuosity of the membrane  $\tau$ [-], and the thickness of the membrane  $t$  [m].

ECP is caused on salt water side and fresh water side. In fresh water side, solute included in fresh water is concentrated at support layer surface. Then, osmotic pressure is increased on membrane surface. The ratio between osmotic pressure of bulk solution and osmotic pressure on membrane surface is expressed as eq.(7).

$$\frac{\pi_{f,m}}{\pi_{f,b}} = \exp\left(\frac{J_w}{k_d}\right) \dots (7)$$

On salt water side, permeated fresh water dilutes the salt water on membrane surface. Then, osmotic pressure is decreased on membrane surface. The ratio between osmotic pressure of bulk solution and osmotic pressure on membrane surface is expressed as eq.(8).

$$\frac{\pi_{d,m}}{\pi_{d,b}} = \exp\left(-\frac{J_w}{k_d}\right) \dots (8)$$

In this eq.(8), mass transfer coefficient  $k_d$  [m/s] is defined with Sherwood number  $Sh$ , diffusion coefficient  $D$ , and hydraulic diameter  $dh$  as follow eq.(9).

$$k_d = \frac{Sh \times D}{d_h} \dots (9)$$

Sherwood number  $Sh$  is defined as follow at laminar flow condition.

$$Sh = 1.85 \left( Re \cdot Sc \cdot \frac{d_h}{L} \right)^{0.33} \dots (10)$$

Here, Reynolds number of fresh water means flow state in the hollow fiber membrane. Reynolds number of fresh water is under 7. Laminar flow state can be supposed. On the salt water side, Reynolds number means flow state in outer side of hollow fiber membrane. Reynolds number of salt water is under 15. Laminar flow state can be supposed. Thus, for salt water and fresh water, eq.(10) can be utilized. We calculated the osmotic pressure around the membrane surface based on the theory written above.

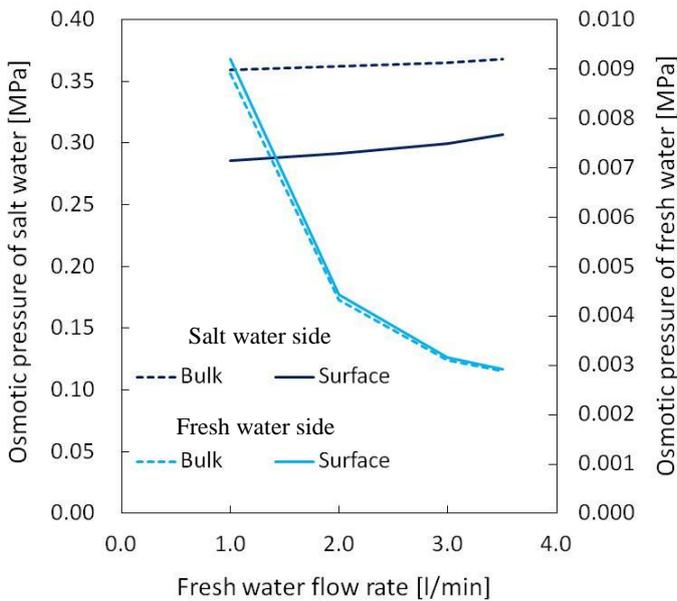


Fig.7 Osmotic pressure of salt water side and fresh water side

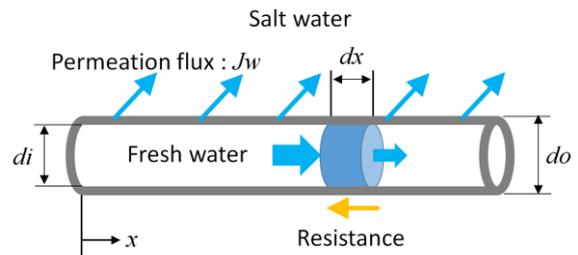


Fig.8 Flow model of fresh water in hollow fiber

Calculated osmotic pressure of fresh water interface and salt water interface are shown in Fig.7 respectively. On the fresh water side, the difference between osmotic pressure on the membrane surface and osmotic pressure in bulk is small. On the salt water side, at the low fresh water flow rate, osmotic pressure on the membrane surface is lower than osmotic pressure in bulk. But this difference of osmotic pressure in bulk and osmotic pressure on membrane surface is constant. Concentration polarization of salt water does not depend on fresh water flow rate.

Therefore, in terms of concentration polarization across the hollow fiber membrane, the larger fresh water supply, concentration polarization of salt water side effects on decrease of permeation flux because the osmotic pressure difference across the membrane is large.

#### 4.5 Effect of fresh water dissipation

In this section, the effect of factor-4, dissipation of fresh water in hollow fiber, is clarified. Firstly, the calculation theory which can calculate the pressure distribution and fresh water flow rate distribution in hollow fiber is derived. In this calculation theory, a model about permeation in hollow fiber as shown in Fig.8 is supposed. Cylinder in Fig.8 shows 1 hollow fiber. In this fiber, fresh water flows. In outer side of this fiber, salt water flows. Here, permeation flow rate is sufficiently low than fresh water flow rate. Thus capillary flow can be supposed. In the micro length  $dx$  of hollow fiber, we derived the relational expression between the rate of change for fresh water flow rate  $dq/dx$  and permeation flow rate  $J_w$  as follow eq.(11).

$$\frac{dq}{dx} = -J_w \pi d_i l \quad \dots (11)$$

Where  $d_i$  [m] is inner diameter of hollow fiber,  $q$  [m<sup>3</sup>/s] is fresh water flow rate in hollow fiber. For pressure gradient in hollow fiber  $dP/dx$ , we obtained by using Darcy-Weisbach equation as follow eq.(12). In here, we suppose that permeation flow rate is sufficiently low than flow rate in hollow fiber, so permeation do not influence on pressure drop in hollow fiber.

$$\frac{dq}{dx} = \frac{128\nu\rho}{\pi d_i^4} q \quad \dots (12)$$

$\nu$ [m<sup>2</sup>/s] is dynamic viscosity,  $\rho$ [kg/m<sup>3</sup>] is density. With these equations, pressure distribution and distribution of fresh water flow rate in hollow fiber against the fiber length  $x$  is derived as follow eq.(13),(14).

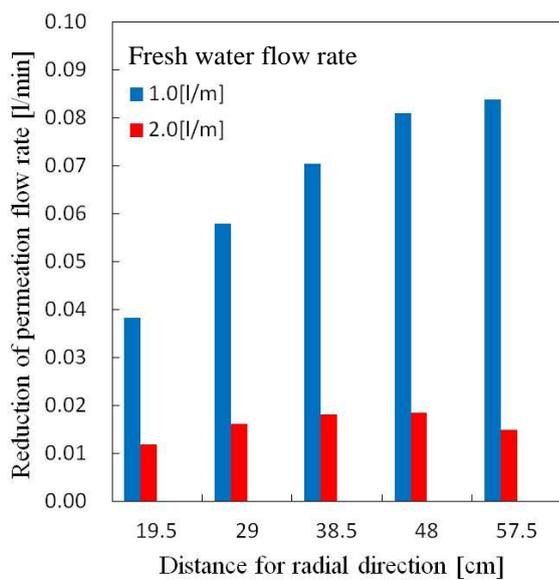


Fig.9 Reduction of permeation flow rate considered effective length for each fresh water flow rate

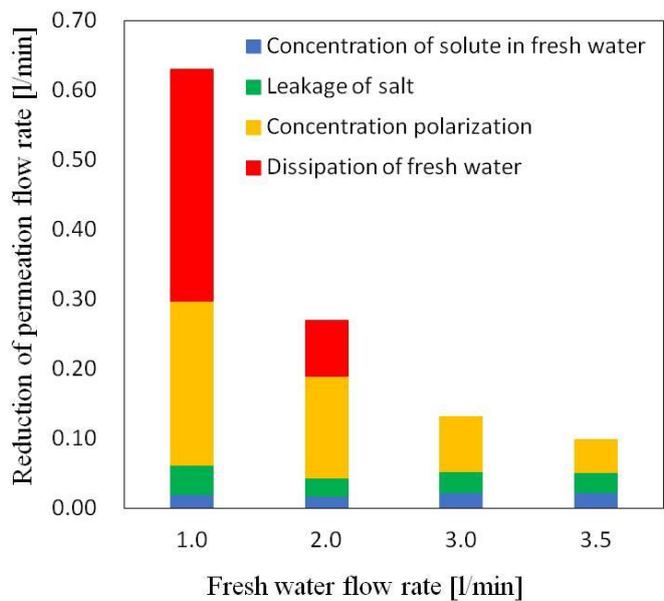


Fig.10 Decrease of permeation flow rate for every effect

$$P = P_0 + \sqrt{\frac{128\nu\rho}{A\pi^2 d_i^5}} \left\{ 1 - \exp\left(\sqrt{\frac{128\nu\rho A}{d_i^3}} x\right) \right\} q_0 \quad \dots (13)$$

$$q = q_0 \exp\left(\sqrt{\frac{128\nu\rho A}{d_i^3}} x\right) - A\pi d_i \left\{ P_0 + (\Delta\pi - P_s) + \sqrt{\frac{128\nu\rho}{A\pi^2 d_i^5}} q_0 \right\} x \quad \dots (14)$$

We can consider the effect of dissipation of fresh water in hollow fiber on decrease of permeation flow rate by using these eq.(13),(14), not eq.(1). Additionally, in terms of osmotic pressure and hollow fiber length in this calculation, we considered that osmotic pressure and hollow fiber length change for radial direction of membrane module.

Decrement of permeation flow rate obtained by this calculation for radial direction is shown in Fig.9. According to this result, decrease of permeation flow rate exists at low fresh water supply. Fresh water in hollow fiber exists over the entire length of hollow fiber at high fresh water supply. In the outer diameter area of hollow fiber element, the larger decrement of permeation flow rate cause because hollow fiber length is long in outer diameter area of hollow fiber element.

#### 4.6 Comparison of each factor

According to 4 factors written above, we quantitatively evaluated the effect on the decrease of permeation flow rate against fresh water flow rate. Results of summarized for 4 factors are shown in Fig.10. According to this result, it is clear that dominant factor of effect on decrease of permeation flow rate change at each fresh water flow rate. In this experiment and calculation, dissipation of fresh water and concentration polarization mainly effect on the decrease of permeation flow rate at 1.0, 2.0[l/min] fresh water flow rate. On the other hand, concentration of fresh water and leakage of salt mainly effect on the decrease of permeation flow rate at 3.0, 3.5[l/min] fresh water flow rate.

#### 5. Conclusions

In this research, we investigated about the 4 factors which effect on the decrease of permeation flow rate with experiment and theory in the case of changing the fresh water flow rate, and obtained conclusions as follows.

1. If fresh water flow rate is large, the main factors for decrease of permeation flow rate are concentration of fresh water and leakage of salt. But this effect is small.
2. If fresh water flow rate is low, the main factors for decrease of permeation flow rate are dissipation of fresh water and concentration polarization. And this effect is large.
3. According to conclusion [1], [2], the optimal fresh water flow exist.

#### Acknowledgment

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