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The effect of pre-processing with various holding times on hydrogen permeation experiements

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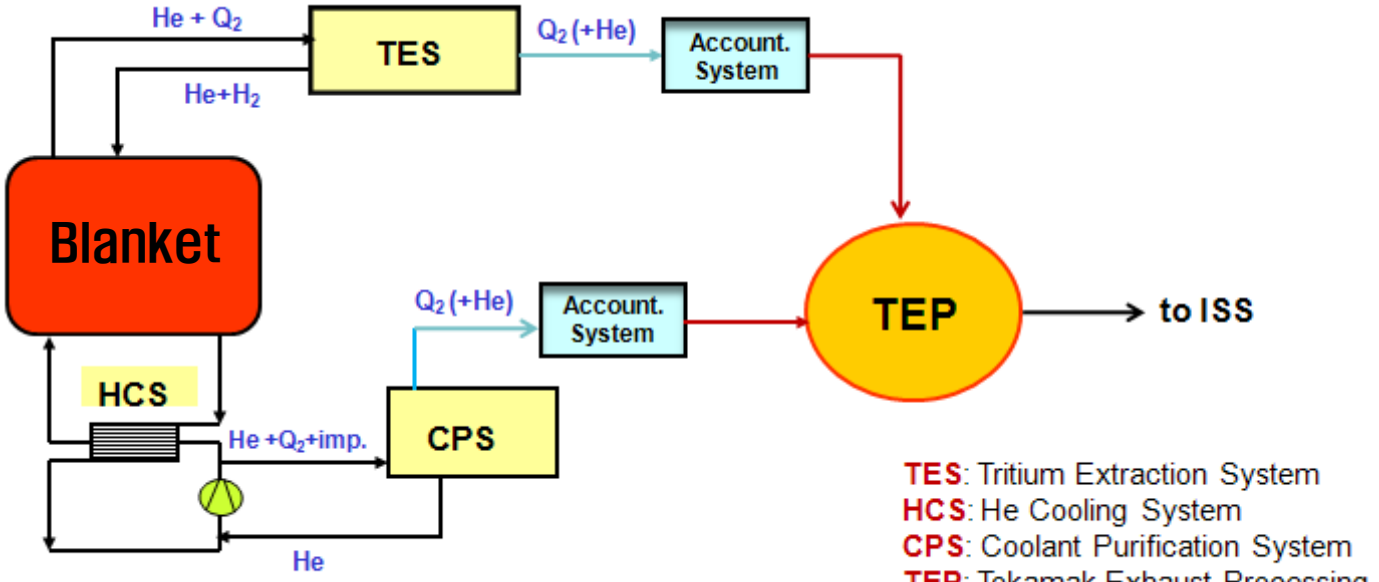
Outline

1. Introduction
2. Experimental Description
3. Experimental Results
4. Summary

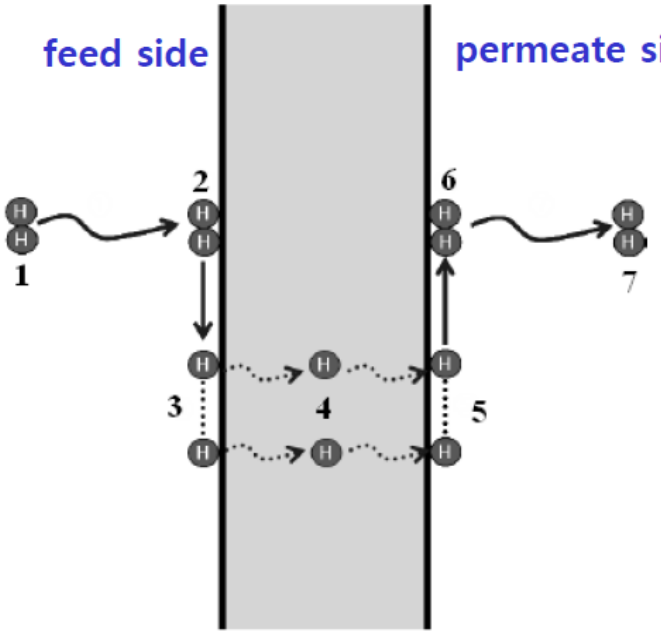
1. Introduction

- Permeation of hydrogen isotopes inevitably occurs due to high-temperature operation in fusion reactor.
- Transport and inventory of hydrogen isotopes are important as safety, fueling and reliability in fusion reactor design.
- One of hydrogen isotopes, tritium is radioactive and has strong permeation through materials.
- To minimize permeation of hydrogen isotopes, permeation properties of structural materials are essential as input data for numerical anlysis.



1. Introduction

- Stainless steel 316L where well known as structural material was chosen as sample.
- Its permeation properties have been established, but some literatures show the scatterd data.
- Permeation test is affected by experimental procedures and environmental conditions
- Oxidiztion layer or existence hydroxyl impacts the permeability
- To remove them, pre-pocessing was considerd and applied with various holding times.
- Both sides of sample were exposed with hydrogen atmosphere in the pre-processing.



1) diffusion of molecules

2) adsorption

3) dissociation

4) diffusion of atoms

5) recombination

6) desorption

7) diffusion of molecules

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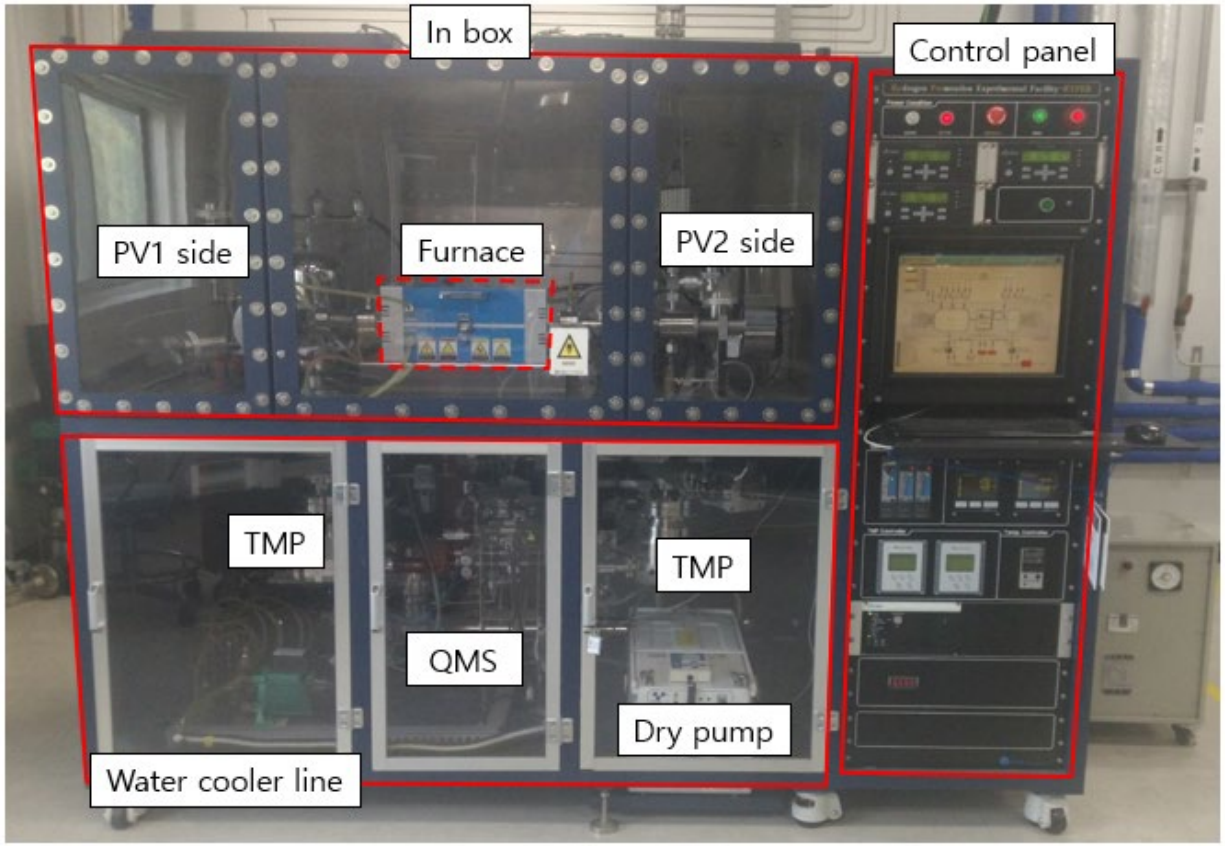
2. Experimental description

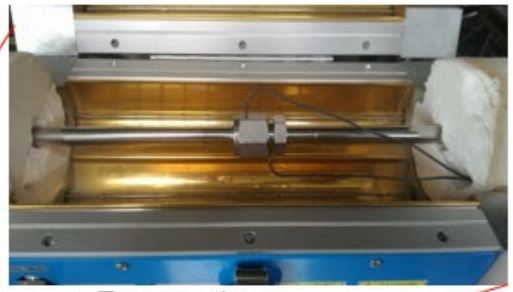
- HYPER was installed and it is in operation at KFE.
  - Hydrogen permeation experimental facility(called HYPER)
  - To investigate permeation properties of structual materials on diversed atmosphere such as H2, D2, etc.
  - For proper permeation procedure, various commissionings are in progress



2. Experimental description

- HYPER was installed and it is in operation at KFE.
  - In-box type for protecting leakage from H2 or D2, contamination by ambient space
  - 4 pressure gauges, 1 thermocouple, at each side
  - 2 thermocouples at test section(to sample)

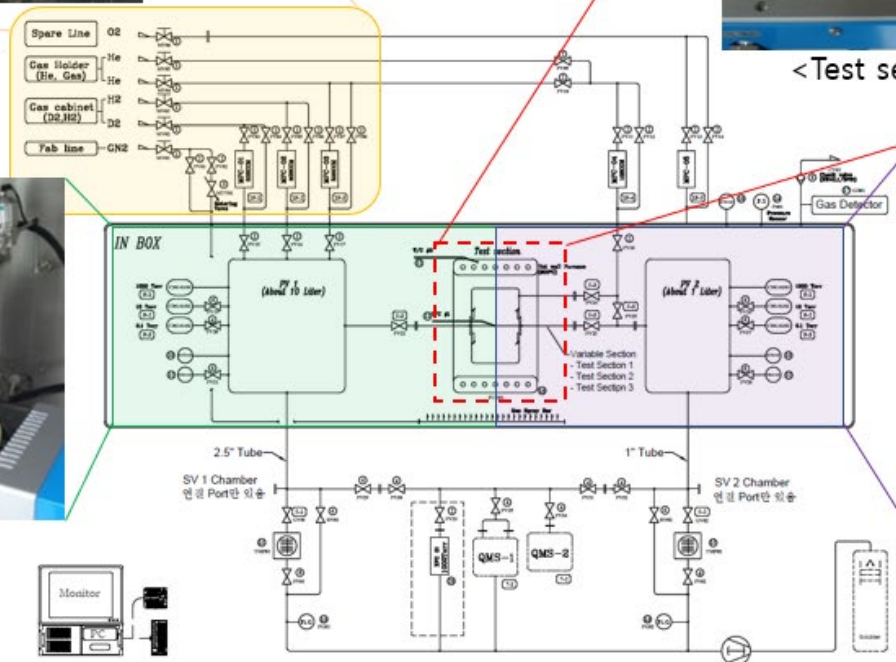
<Gas supply line>



<Test section>

<PV1 side>
Feed side
High pressure side

<PV2 side>
Permeate side
Low pressure side



2. Experimental description

- Equations for permeability, diffusivity and solubility

$$J = \frac{DK}{d}\left(p_h^{\frac{1}{2}} - p_1^{\frac{1}{2}}\right)$$

$$\rightarrow \frac{DK}{d} p_h^{\frac{1}{2}} \qquad \because DK = \Phi$$

$$\rightarrow \frac{\Phi}{d} p_h^{\frac{1}{2}}$$

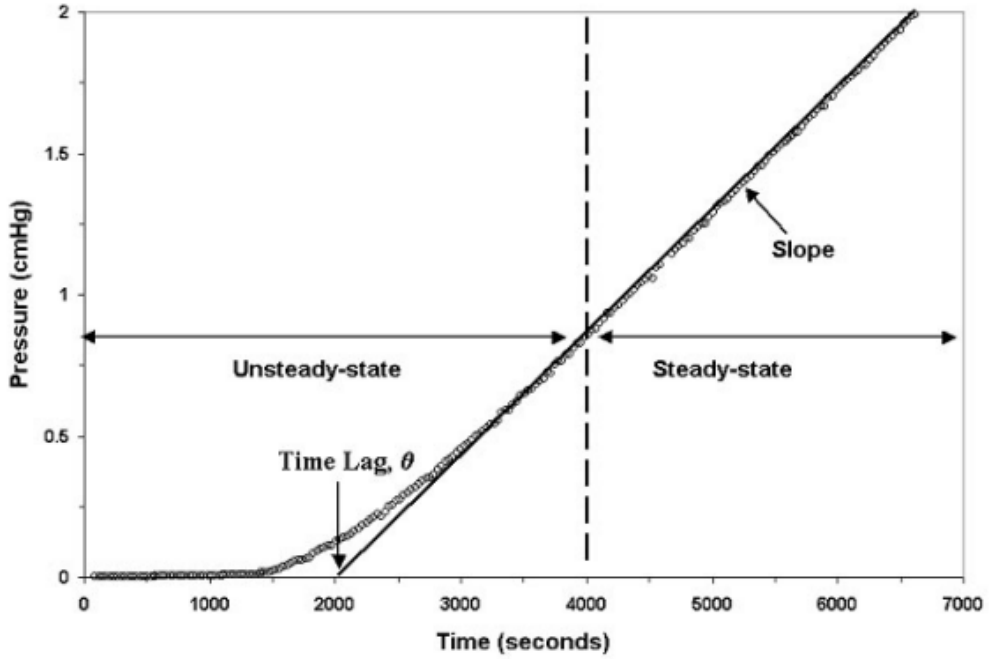
$$t_1 = \frac{d^2}{6D}$$

$$\rightarrow D = \frac{d^2}{6t_1}$$

Symbol	Unit	Description
J	$\frac{mol}{m^2\ s}$	Rate of gas permeation per unit area
d	m	Thickness of material
P_h	Pa	The gas pressure on the high pressure side
P_1	Pa	The gas pressure on the low pressure side ≈ High vacuum, equal to 0
D	$\frac{m^2}{s}$	Diffusion coefficient of material
K	$\frac{mol}{m^3\sqrt{Pa}}$	Sieverts' constant of material
Φ	$\frac{mol}{m\ s\sqrt{Pa}}$	permeation coefficient of material
t_1	s	Time lag from pressure-sec curve (slope and intercept)

2. Experimental description

- Equations for permeability, diffusivity and solubility



- Slope : evaluated to permeability

$$\frac{Pa}{s} \rightarrow \frac{mol}{s} \rightarrow J \rightarrow \phi$$

where needed constants for calculating J is ..

⇒ Volume, temperature, permeate area on permeate side

- Time lag : evaluated to diffusivity

Figure 2-10. Typical permeation experiment data analyzed using Time Lag Method.

The permeability coefficient P is calculated from the slope found in Fig.1 within the steady-state region.*

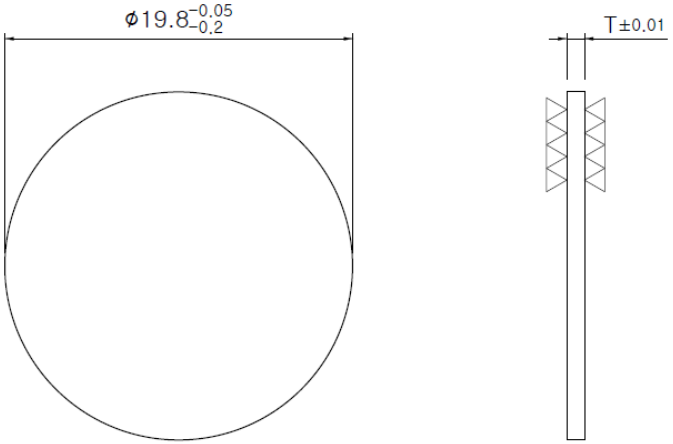
A common error in using this method(time lag) is not allowing enough time to establish steady state.**

* Ref) Pechar T.W. (2004) Fabrication and Characterization of Polyimide-based Mixed Matrix Membranes for Gas Separations, Ph.D. Thesis, Virginia Polytechnic and State University, Blacksburg, VA, 334 pages.

** Ref) Paul D.R. (2016) Time Lag Method for Mass Transport Properties Evaluation in GS., Encyclopedia of Membranes., Berlin, Heidelberg.

2. Experimental description

- Sample information
  - Material : 316L stainless steel
  - Thickness : 0.7mm / Diameter : 19.8 mm
  - Both surfaces were polished as in the following figure.



- Conditions for pre-processing
  - Pressure : 100 kPa on the feed side,
    100 kPa on the permeate side
  - Sample temperature : 500℃
  - Hold time : 0, 1, 2, 3, 4, 6, 12 hr

- Conditions for permeation experiments
  - Pressure : 100 kPa on the feed side,
    ~10^{-6} Pa on the permeate side as minimum
  - Sample temperature : 500℃

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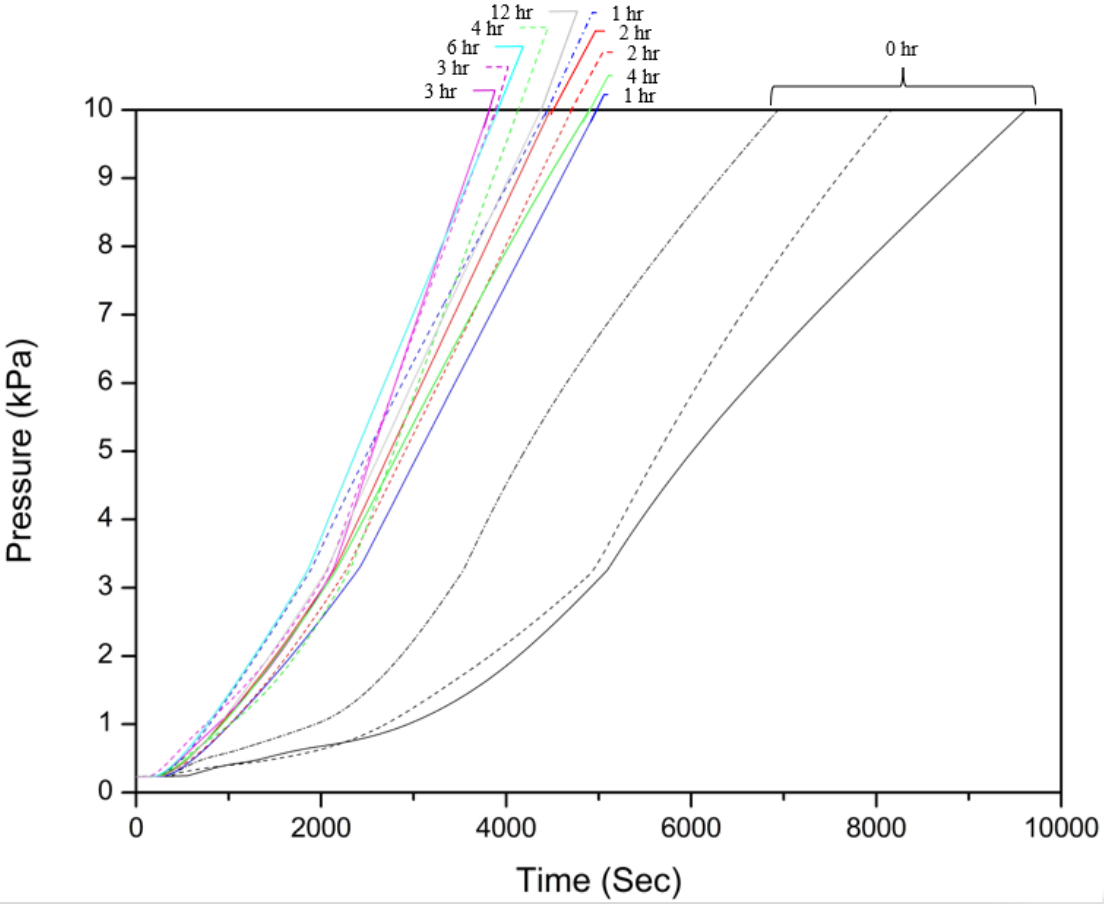
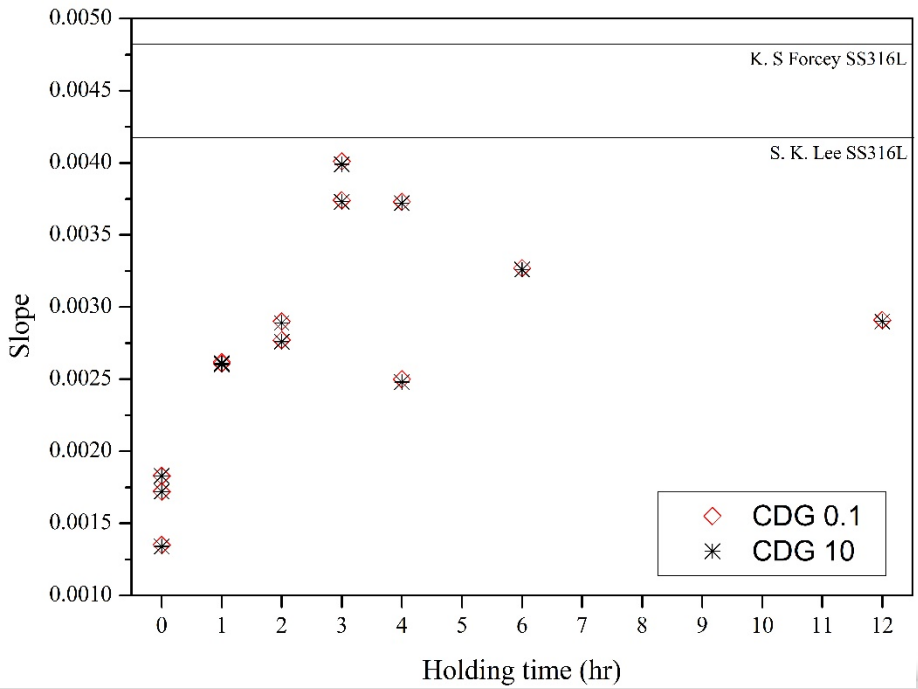
3. Experimental Results

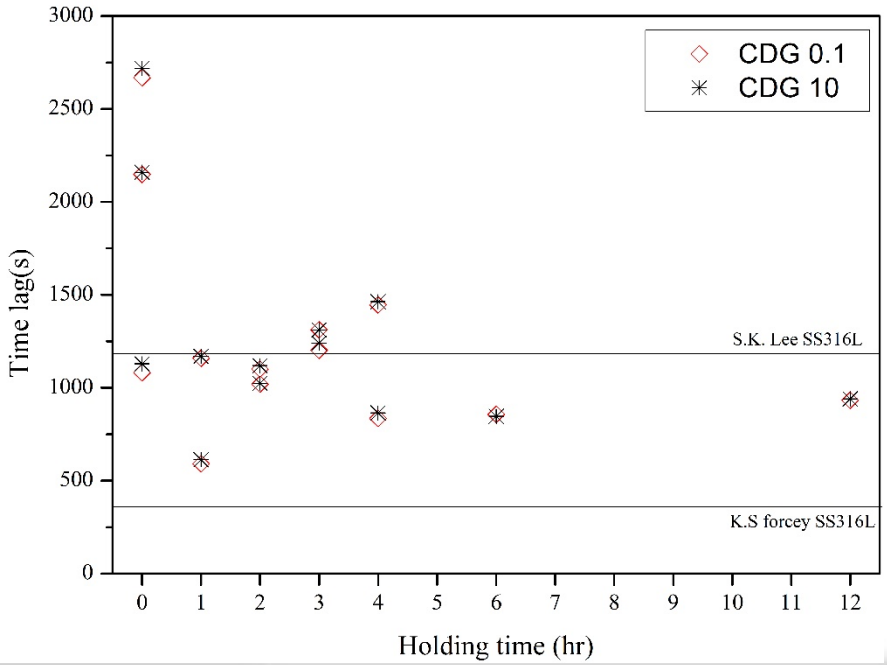
- Pressure rise data
  - Results without the pre-processing show significant differences among the results.
  - Bare SS316L could contain an error in permeability.



3. Experimental Results

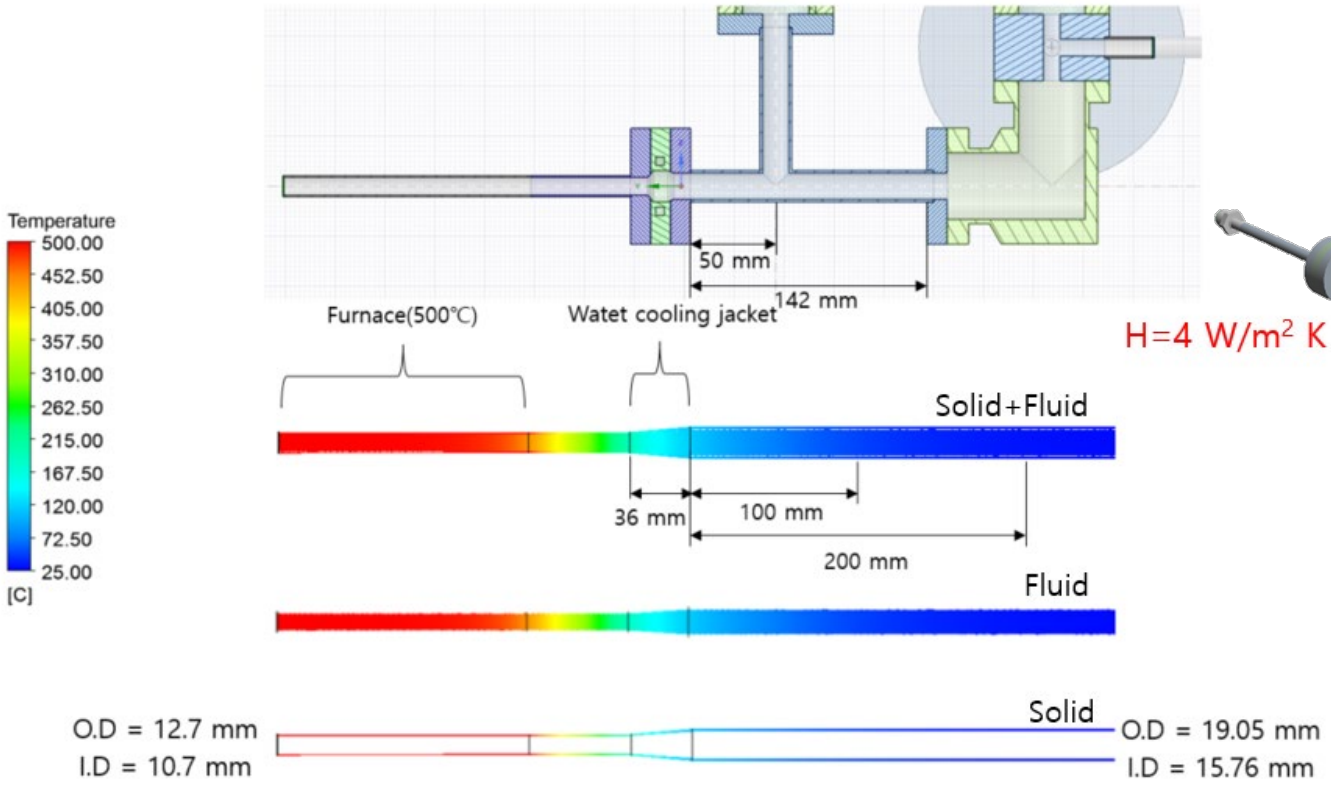
- Slope and time-lag data
  - Results without pre-processing show significantly differences among the results.
  - The slopes, indicating permeation rates, increase as the holding time increase up to 3 hours and then they are saturated.
  - The discrepancy of the time lags at each holding time tends to become small.
  - It suggests that the pre-processing more than 2 hours is necessary to obtain consistent permeation data.

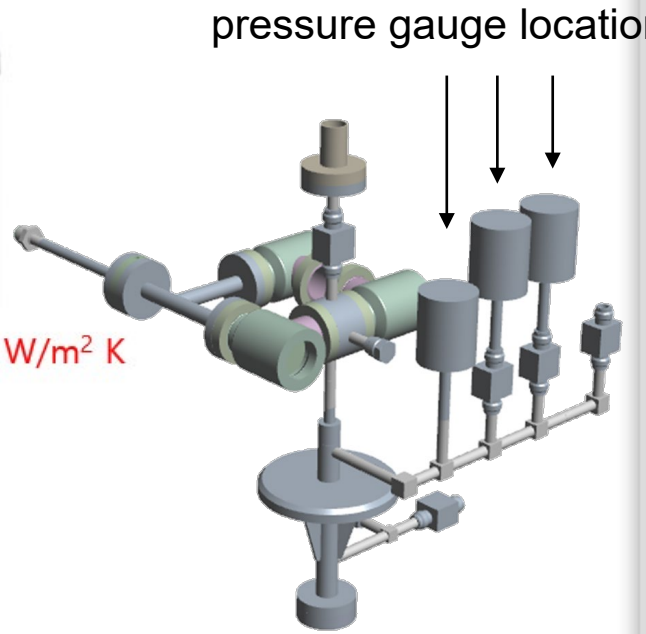




3. Experimental Results

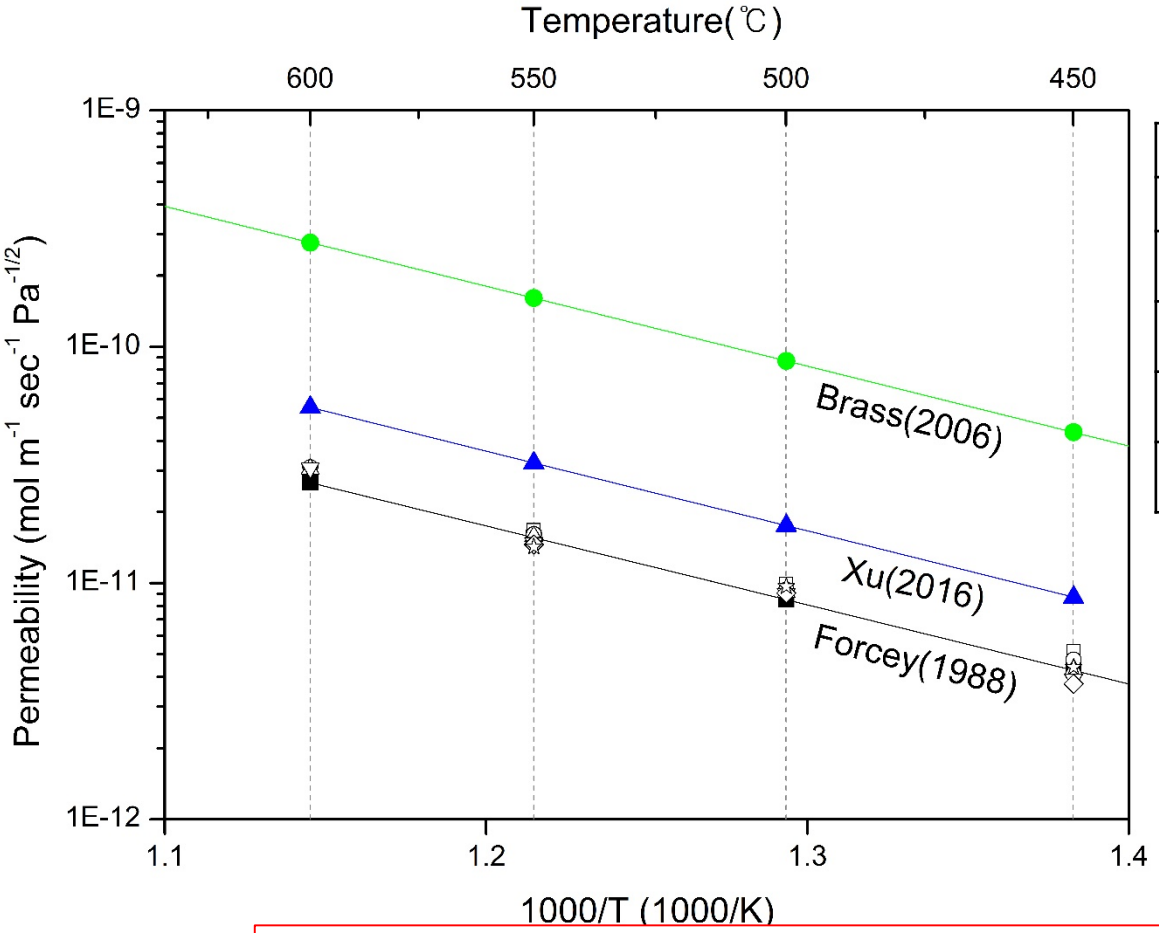
- Checking permeation parameter - Temperature on permeate side
  - thermal anlysis result : Avg. volume temp ; 45.8℃ (on permeate side volume)
  - Thermocouple value(actual) : ≈ room temperature





3. Experimental Results

- Permeability, holding time : 3 hr
  - Results show well agreement to reference data



Permeability, Φ (mol m^{-1} sec^{-1} $Pa^{-1/2}$)		
Temp	Avg.*	Forcey
450	4.381E-12	4.2692E-12
500	9.365E-12	8.5005E-12
550	1.536E-11	1.5567E-11
600**	3.068E-11	2.6599E-11

* 6 point at each temperature

** 3 points

$E_{\Phi,\ avg.}$ = 64.3 kJ/mol ; ref 60~72 kJ/mol, Forcey : 64.0 kJ/mol

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- Permeation test facility(HYPER) is in operation at the KFE.
- The pre-processing for permeation experiment were conducted using HYPER.
- Permeation test without pre-processing could contain an error in properties of permeation.
- Sufficient holding time is necessary to reduce discrepancy in permeation tests.
  - As the results, 3 hours of holding time have been decided
- Further study is expected to elaborate on reliability improvement for diffusivity and solubility using HYPER
- Also, this procedure will be tested to advanced reduced-activation alloy(ARAA) which is under developing for HCCR TBM application

Thank you for your attention !