

Innovative Circular Technologies for Harmful Nitrogen Compounds/ To Solve Planetary Boundary Issues

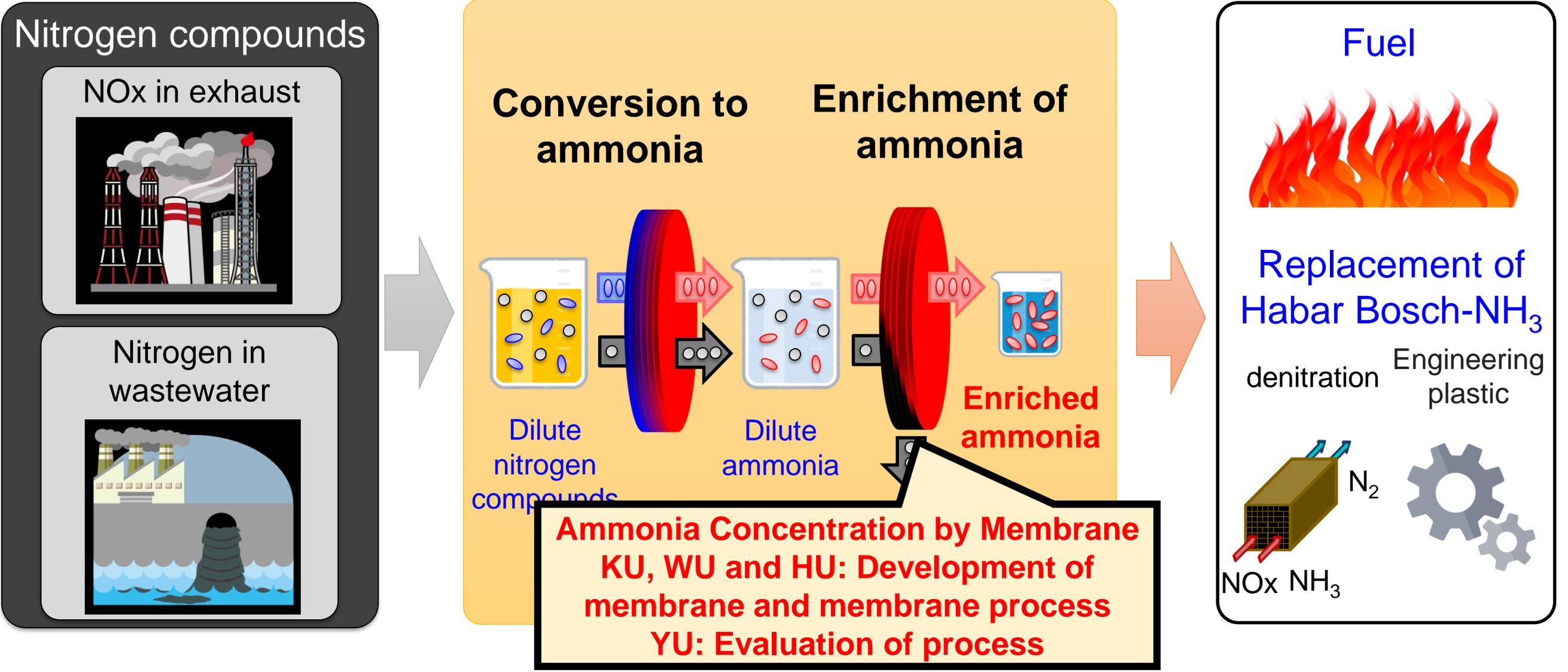
Theme 2. Recycling nitrogen compounds in wastewater to ammonia resource
Theme 2-2. R&D on ammonia recycling by separation and concentration
Development and evaluation of ammonia concentration process
by membrane separation

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Implementing organizations : National Institute of Advanced Industrial Science and Technology (AIST),
The University of Tokyo, Waseda University,
Tokyo University of Agriculture and Technology, Kobe University,
Osaka University, Yamaguchi University, Kyowa, Hakko Bio Co., Ltd.,
ASTOM Corporation, Toyobo Co., Ltd., FUSO Corporation, Ube Industries, Ltd,

Position in the Project

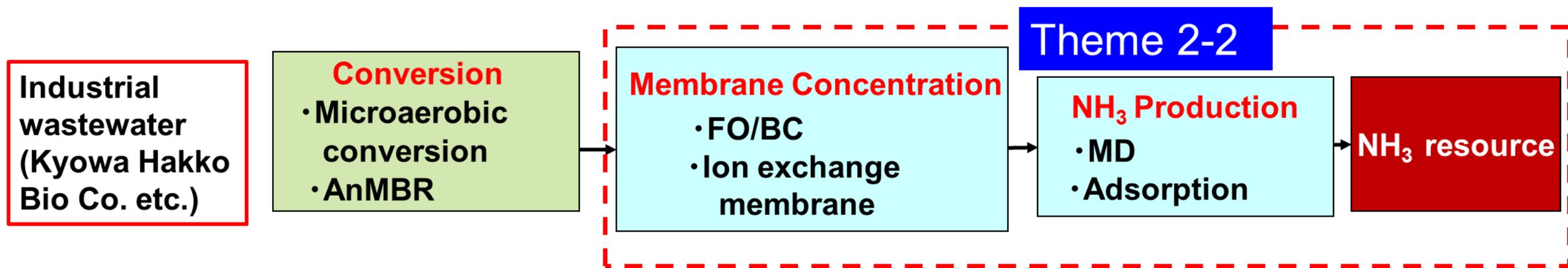


Target of Theme 2 for FY2029 : Demonstration with a pilot plant with and enrichment on a scale of 5~15 m³/d.

Position of Kobe U: Development and evaluation of ammonia concentration process by membrane separation

Target of Kobe U for FY2029:Development of FO process used in pilot plant, Development of DS with performance required for FO process, Establishment of membrane distillation process used in pilot plant.

Concentration systems are developed for the converted NH_4^+ in the Theme 2-1 using various separation membranes and high-performance adsorbents.



【 R&D Items 】

- Development of Forward osmosis (FO) membrane, Brine Concentration (BC) membrane, Ion exchange membrane and Membrane distillation (MD) and Establishment of ammonia wastewater concentration process with low energy consumption by these membrane separations
- Establishment of circular process for Nitrogen Compounds based on development of high-performance adsorbents and adsorption-desorption methods

Membrane

 : Kobe U team

- Development of membrane and membrane process (Kobe U)
- Optimization/modulization of hollow fiber FO (Toyobo Co.)
- Development of Zeolite FO (Waseda U) Subcontract
- NH_4^+ concentration process by IEM (Yamaguchi U)
- Development of IEM and module (ASTOM Co.)
- System/module design of MD (Hiroshima U) Subcontract

Adsorption

- Development of high-performance adsorbents and ammonia recovery process (AIST)
- Ammonia adsorbent using complex reaction (Tokyo U)
- Establishment of ammonia recovery process based on adsorption (FUSO Co.)

Process

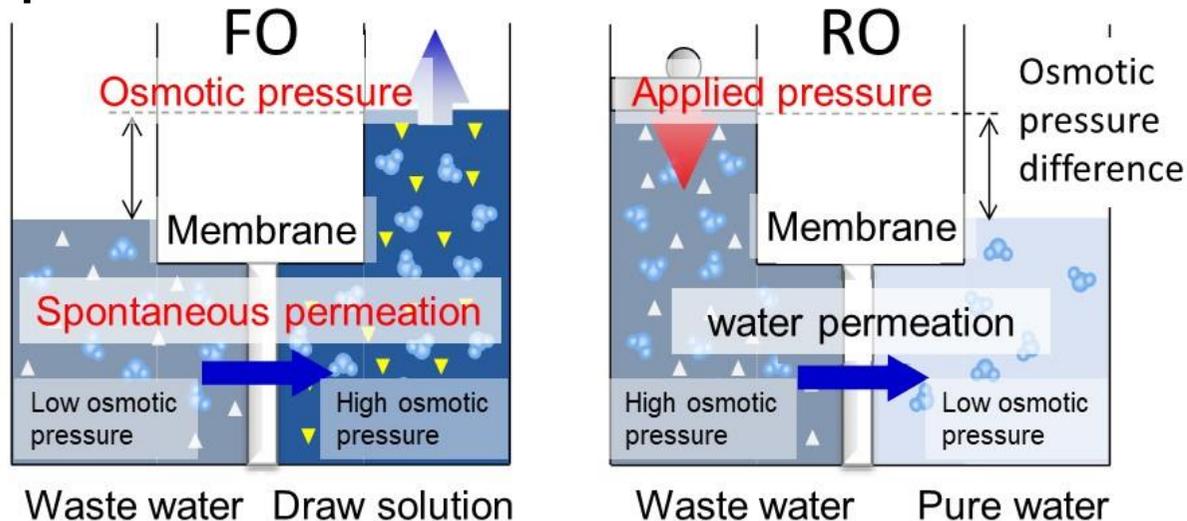
- Evaluation of ammonia concentration process using membrane and adsorption (Yamagata U) Subcontract



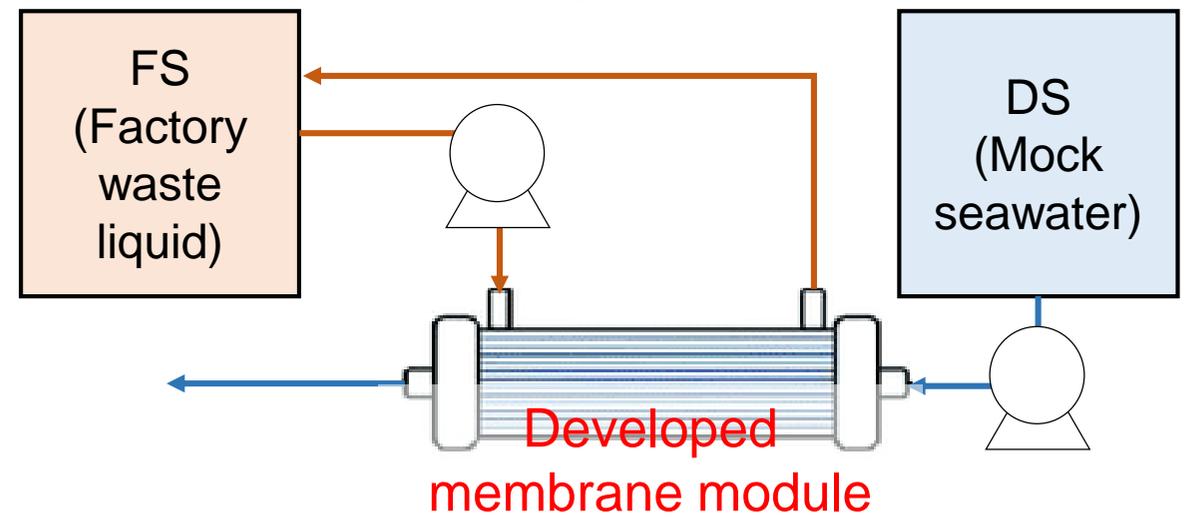
Achievement (1) FO process (Kobe U)

- It was demonstrated that real industrial wastewater A and B could be concentrated by using sea water as DS.
- Achieved the target concentration (T-N 4000 ppm) of real industrial wastewater

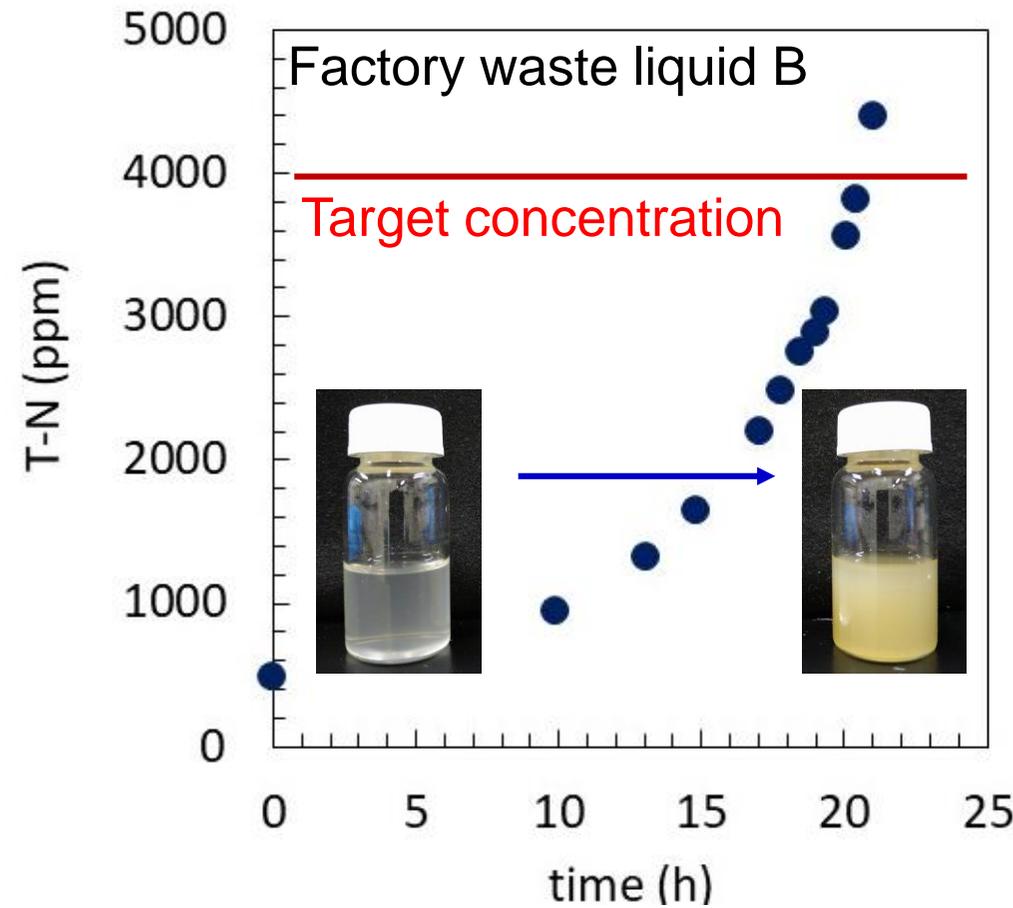
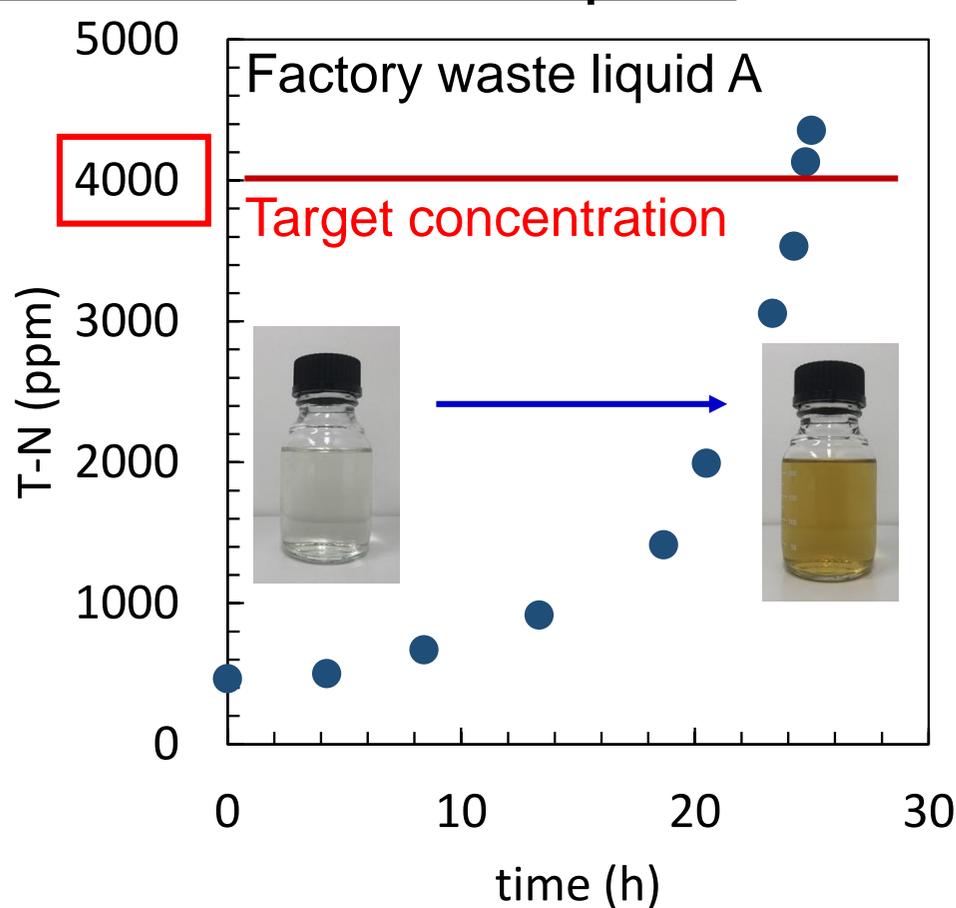
FO process



Experimental set up of FO test



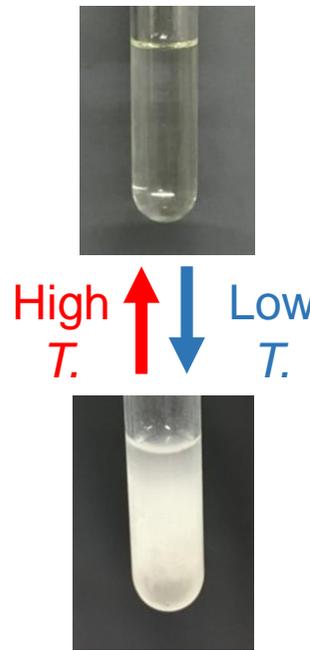
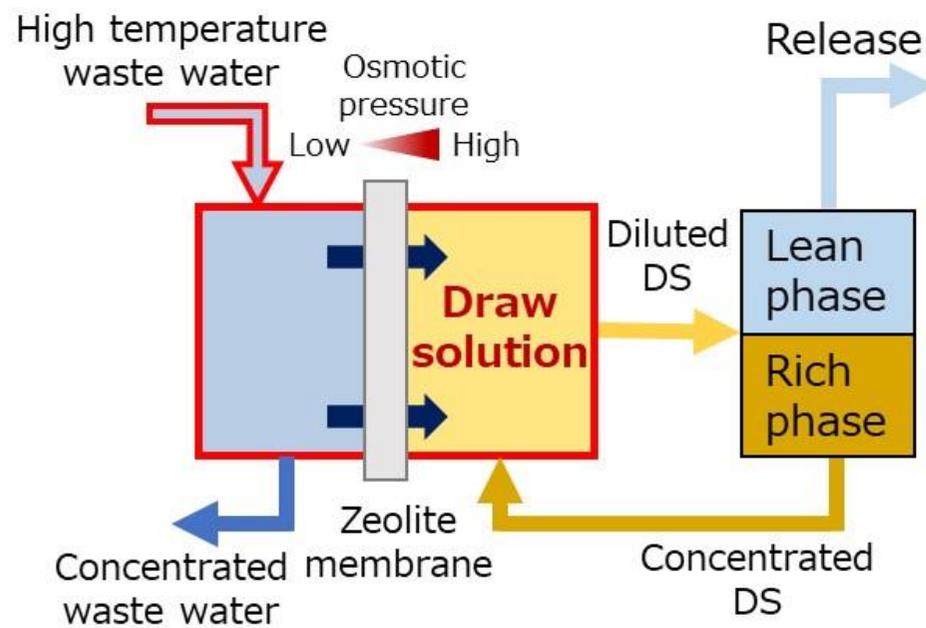
FO test of waste liquids



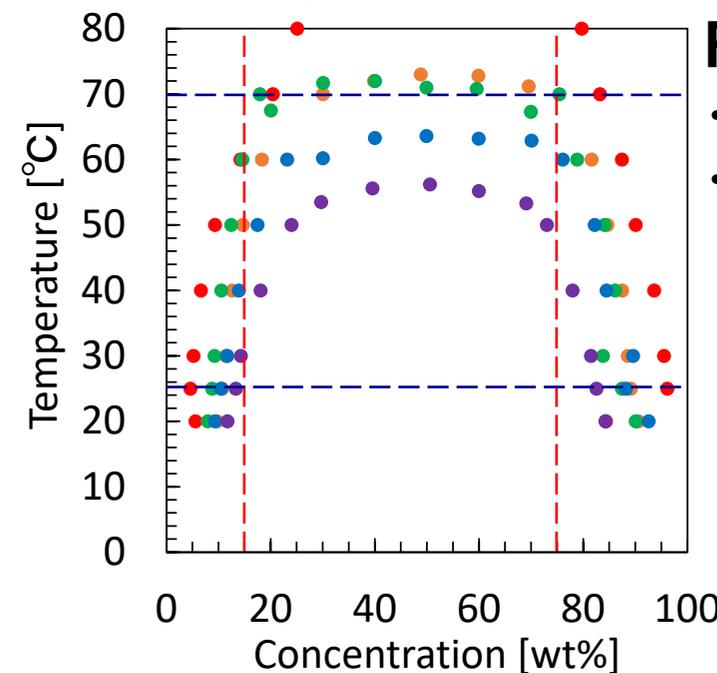
It was demonstrated that both factory waste liquids A and B were able to be concentrated using seawater as the DS.

- Successfully developed the UCST-type ionic liquid (draw solution) that meets the target requirements (Kobe U)
- Demonstrated the high temperature FO operation with a developed zeolite membrane and UCST-type ionic liquid (Waseda U)

High temperature FO Process



Phase diagram of synthesized ionic liquids

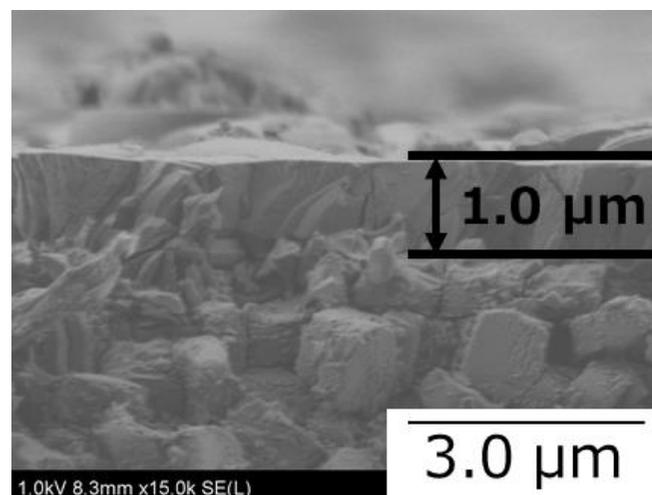
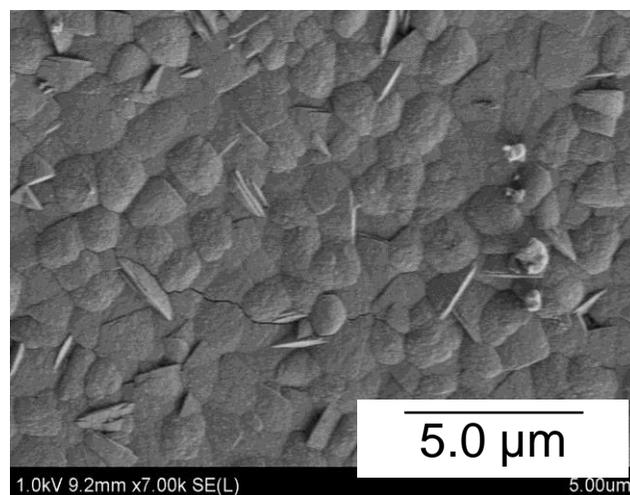


Requirements

- UCST < 70°C
- Concentration @ 25°C
- Rich phase > 75 wt%
- Lean phase < 15 wt%

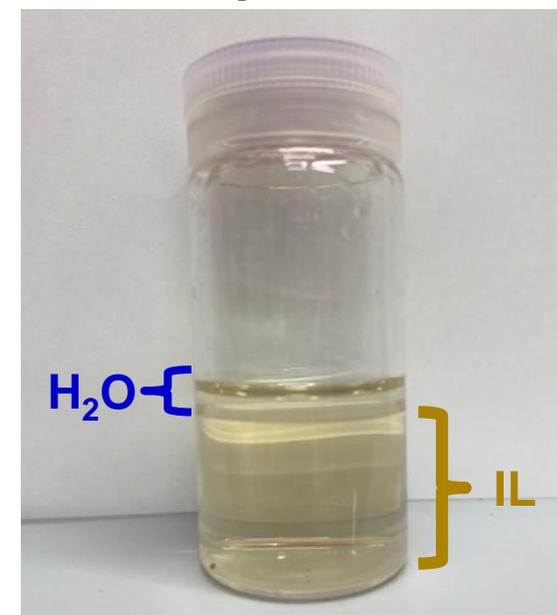
10 types of ionic liquids were synthesized, and **2 of them achieved the target performance.**

Development of zeolite membranes



It was found that a **hydrophilic** zeolite membrane **without a cation exchange site** was required for NH_4^+ concentration.

Ionic liquid after FO



Conditions

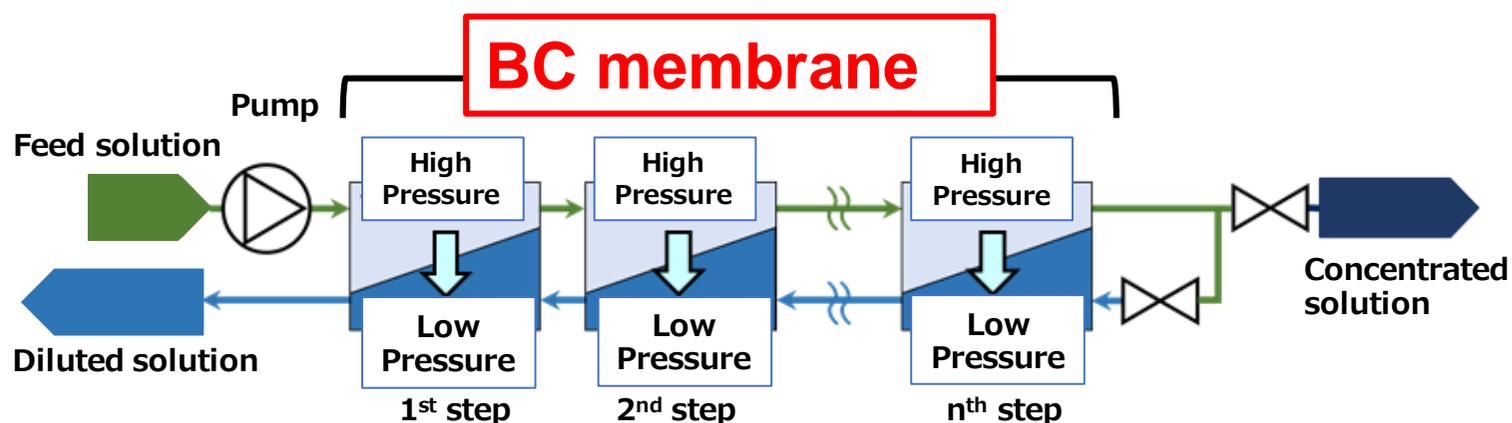
DS: Ionic liquid
FS: H_2O
Temperature: 343 K

- Water permeation through the zeolite membrane was confirmed
- DS rich phase and lean phase were successfully separated

Achievement (2) Brine Concentration (Kobe U)



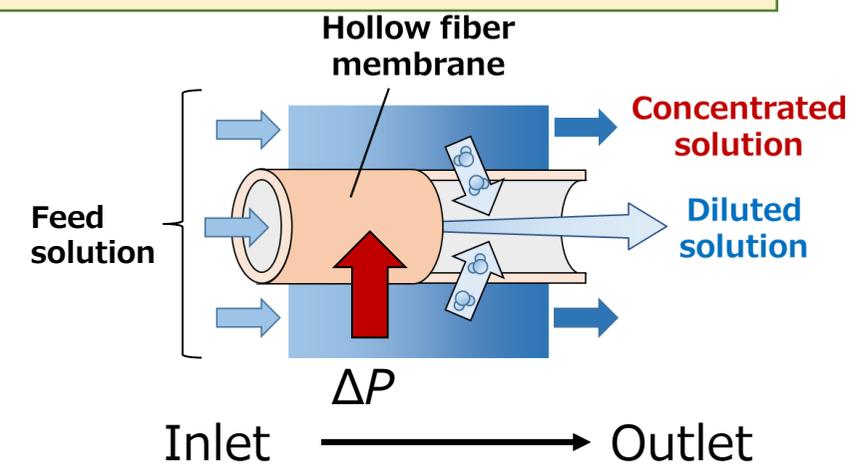
- Optimization of BC operating conditions (Flow rate, Applied pressure, Reflux ratio)
- Achievement of 10 times NH_4^+ concentration using NaCl coexistence model wastewater



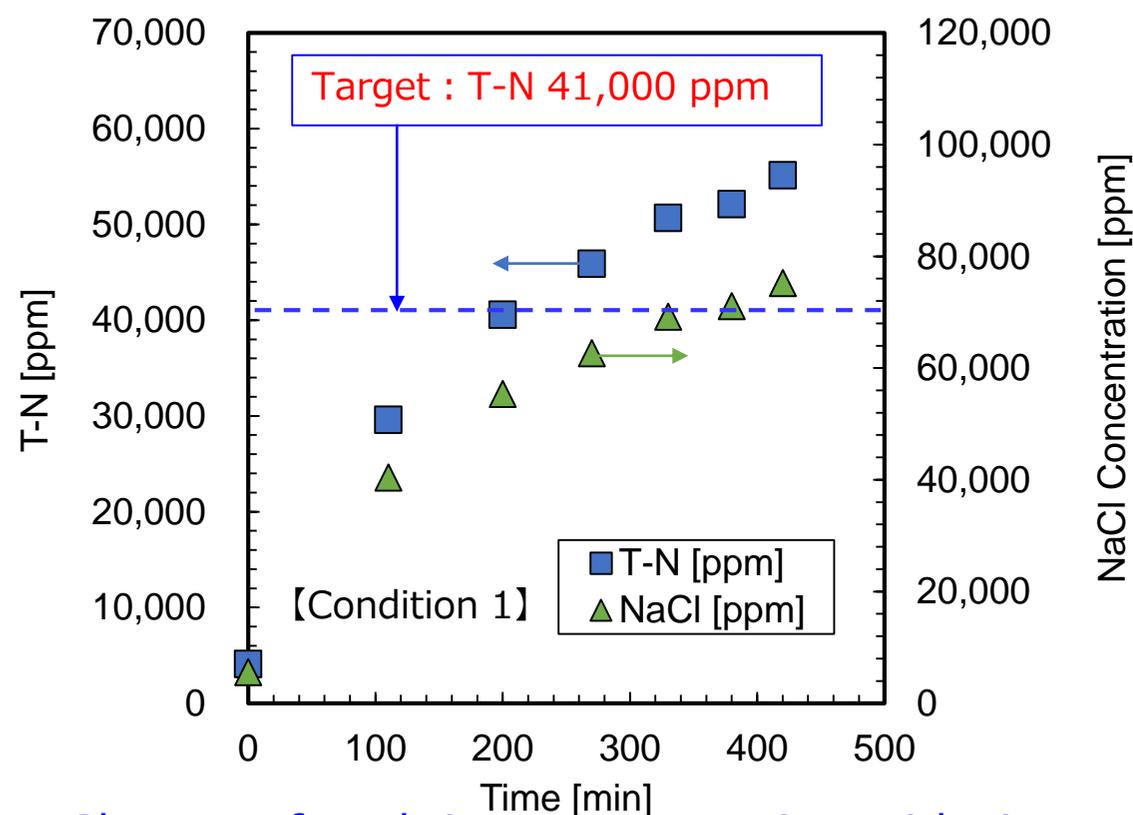
Principle of Brine Concentration (BC) method



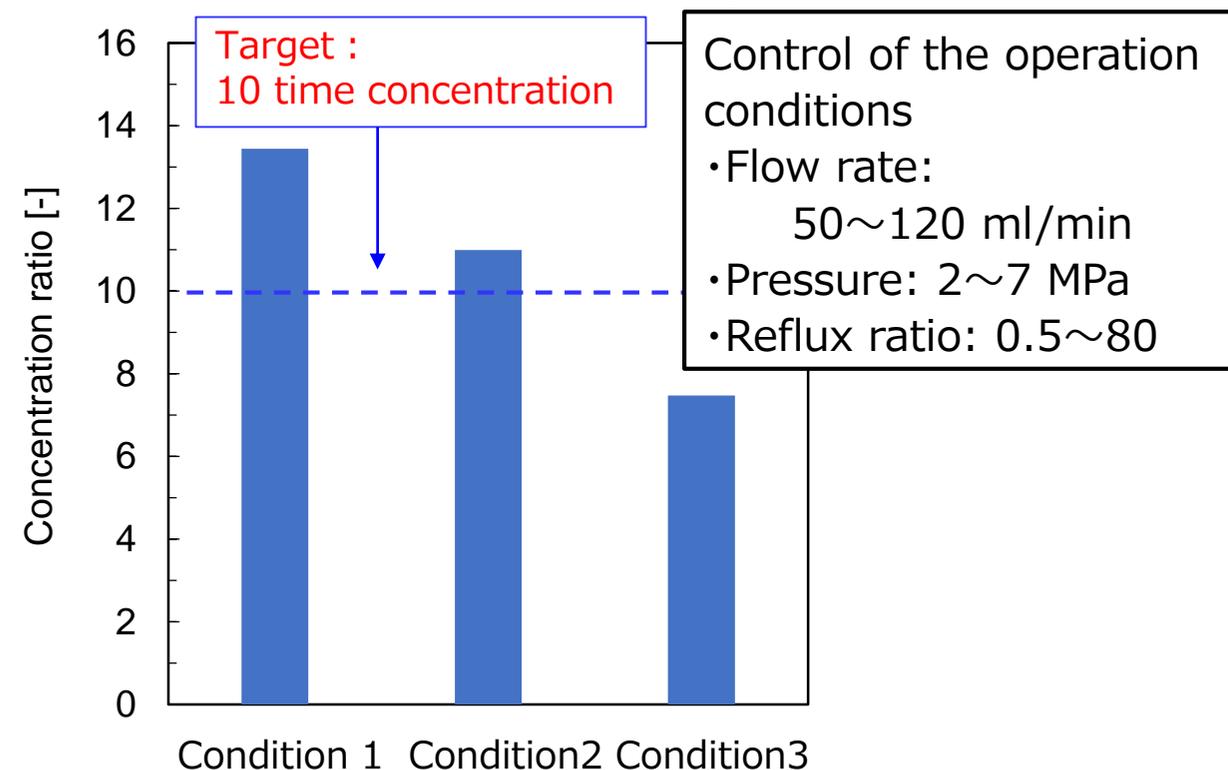
BC membrane module



Concentration using HF membrane



Change of each ion concentration with time in NaCl coexistence model waste liquid



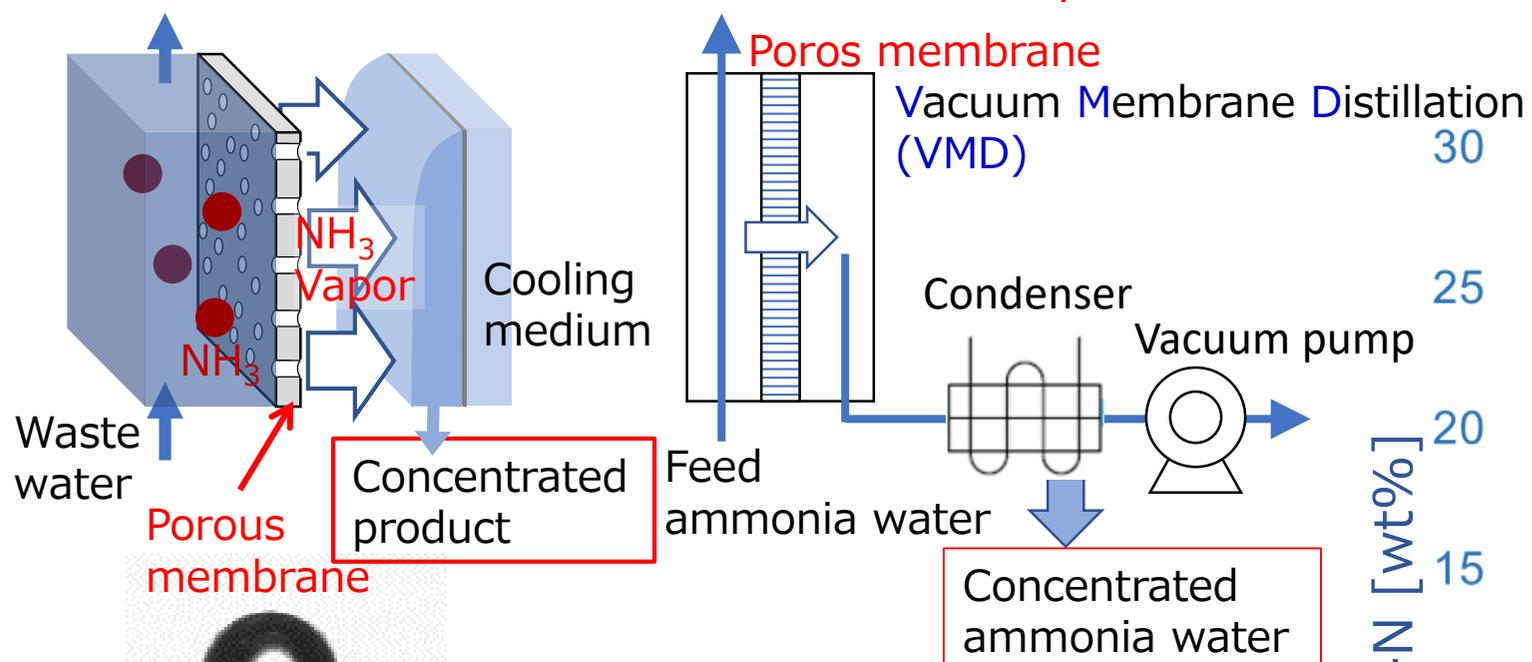
Concentration ratio for each condition

Successful NH_4^+ concentration with the target value (over 10 times) using model waste liquid with NaCl coexistence (T-N 4,000 ppm)

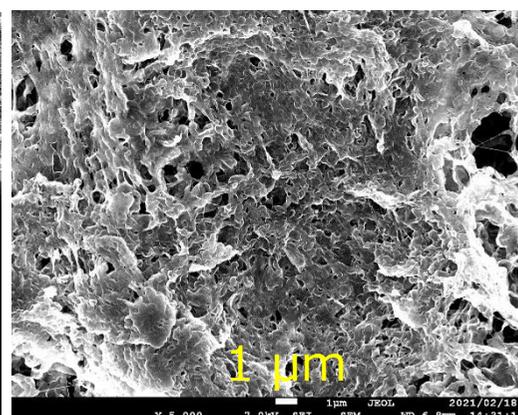
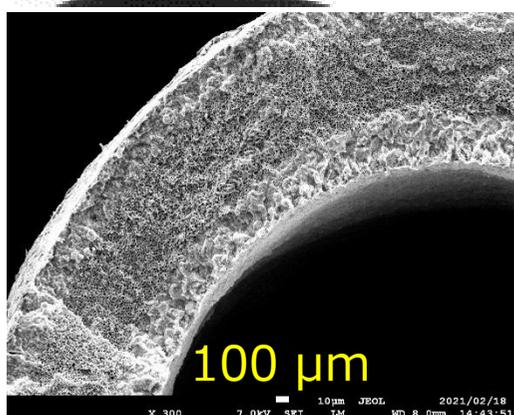
- Hydrophobic hollow fiber membranes that could concentrate ammonia water by membrane distillation method were developed.
- Target concentration with high recovery rate was successfully achieved by flow-type vacuum membrane distillation (VMD) operation.

Membrane Distillation (MD) method

A solution is supplied to one side of the hydrophobic porous membrane, and highly volatile ammonia molecules are preferentially evaporated, permeated, and condensed on the other side of the membrane to obtain a concentrated ammonia water on the permeation side.



Repulsive to ammonia water

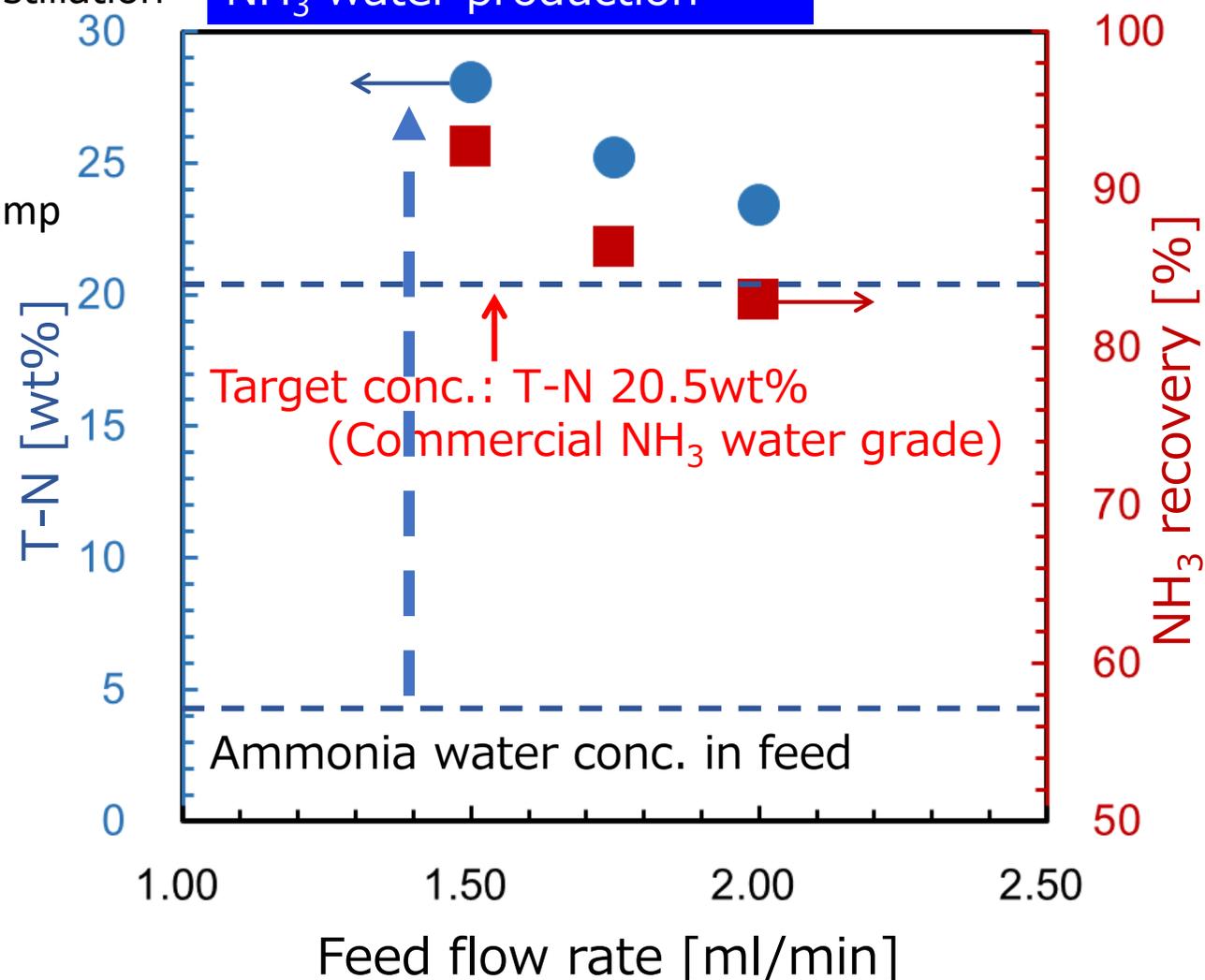


Cross section

Outer surface

Succeeded in preparing hydrophobic porous hollow fiber membrane

Achieved T-N 25wt% for NH₃ water production



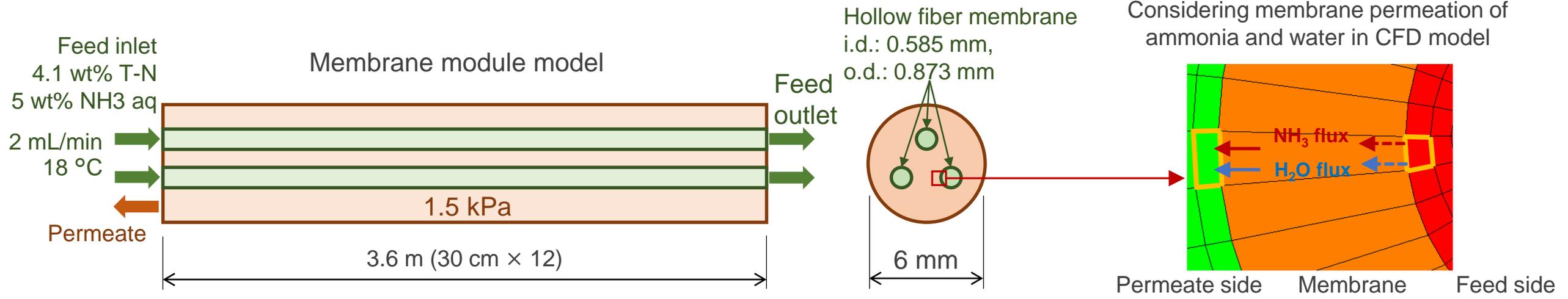
Ammonia water can be concentrated with a high recovery rate.

Achievement (3) Membrane distillation (Hiroshima U)

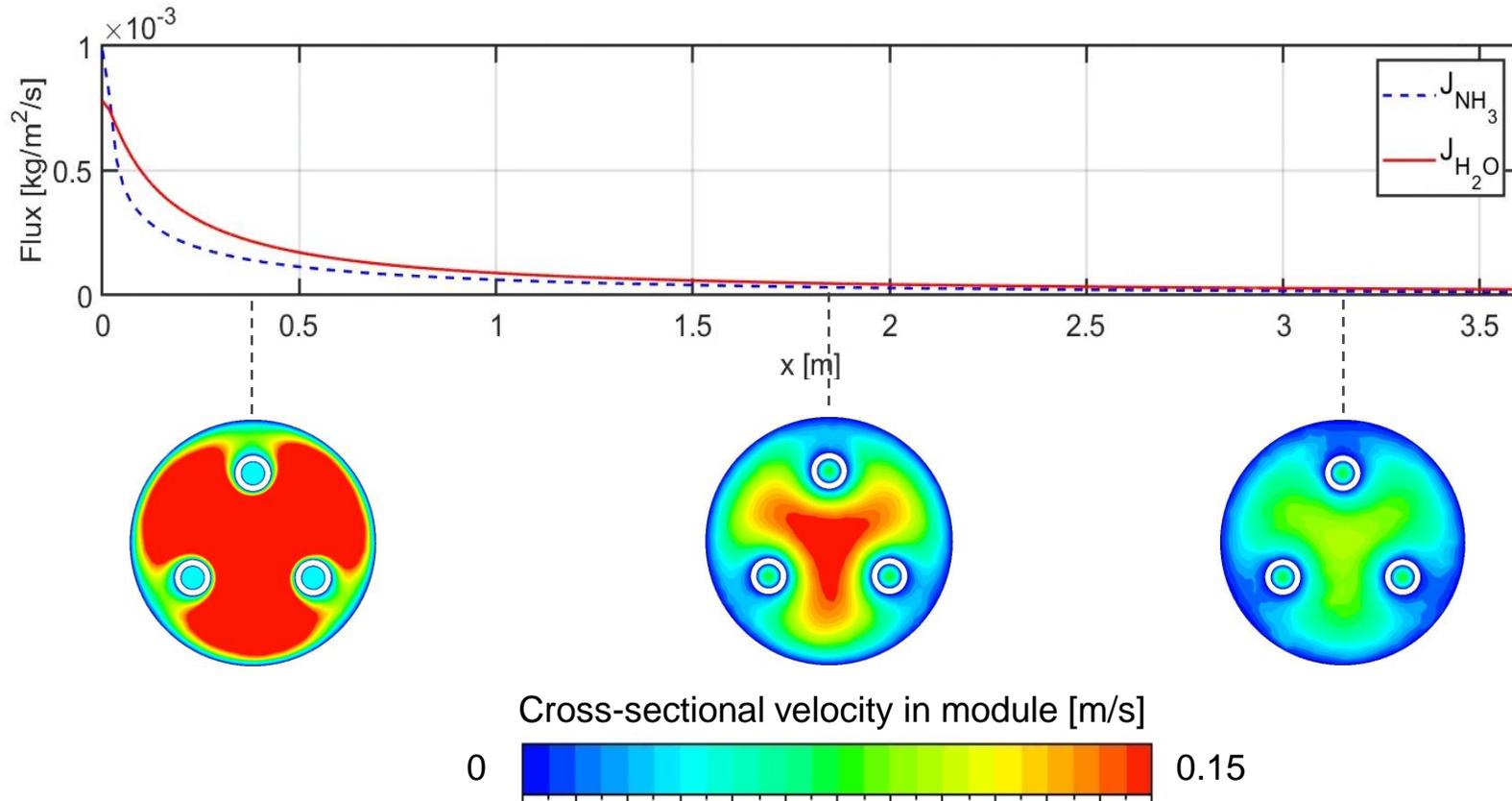


- A computational fluid dynamics (CFD) model for membrane distillation module was developed.
- This simulation model can quantitatively predict the experimental results.

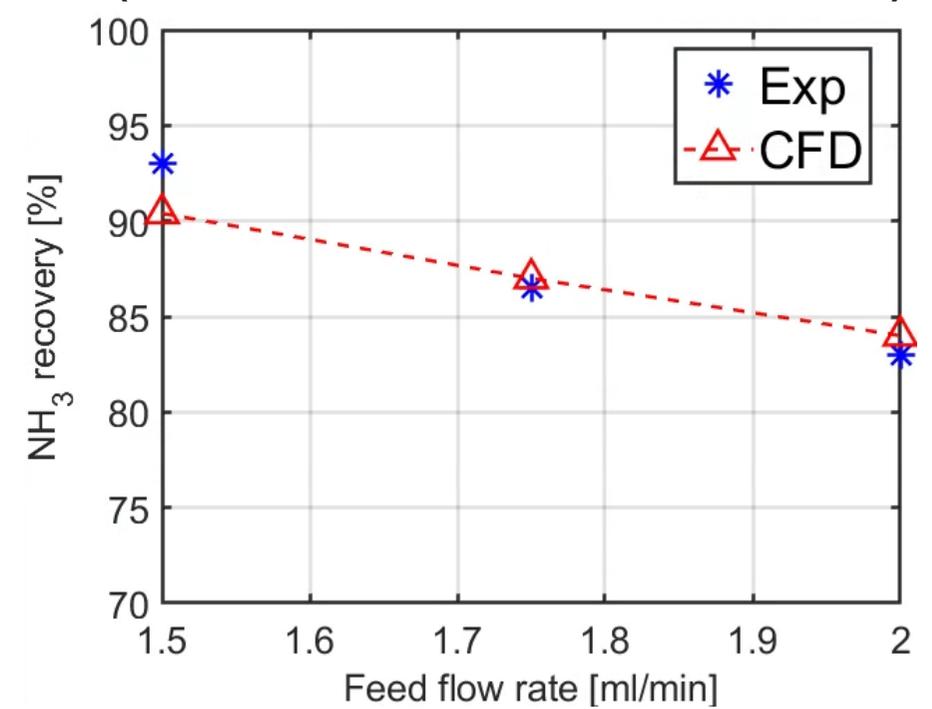
CFD modeling based on experiments of membrane distillation (Kobe Univ.)



Permeation flux distribution of ammonia and water in the module



Comparison with experimental results (Effect of the flow rate of feed solution)

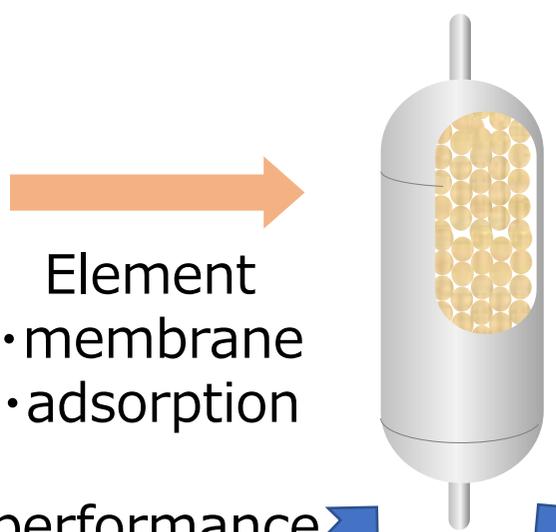


This simulation model can quantitatively predict the experimental results.⁹

Reasonably describe membrane permeation phenomena and velocity distribution in the module

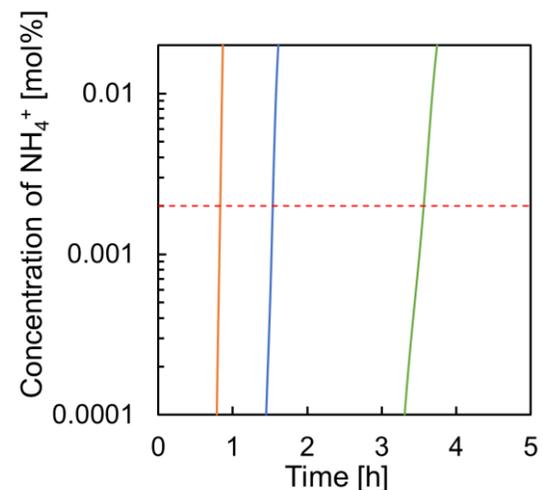
- Separation process for liquid-phase were successfully modeled.
- Operation and design methodologies were developed from the simulation.

Liquid phase separation



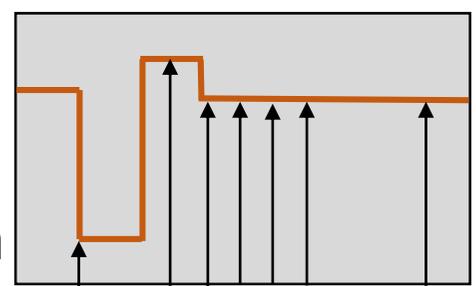
Element
• membrane
• adsorption

Ex. Results of adsorption



- adsorption
- breakthrough
- time
- etc

Process models



Analysis, Evaluation

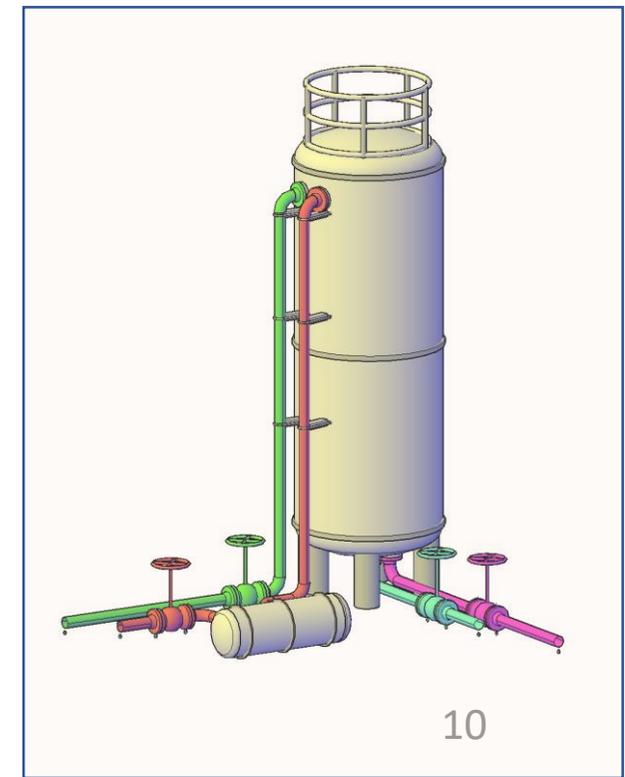
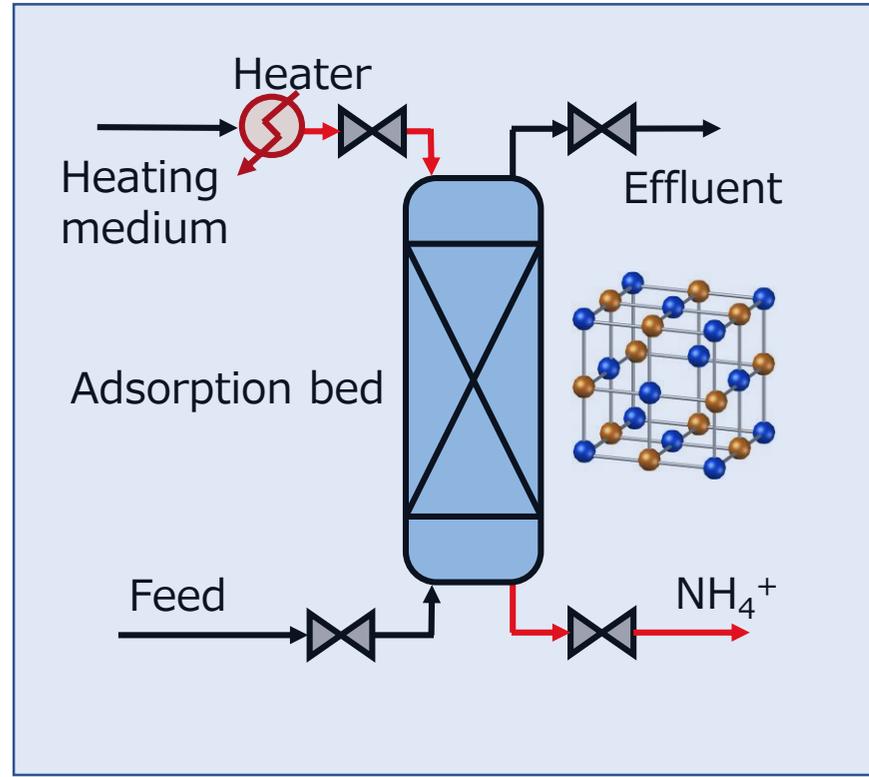
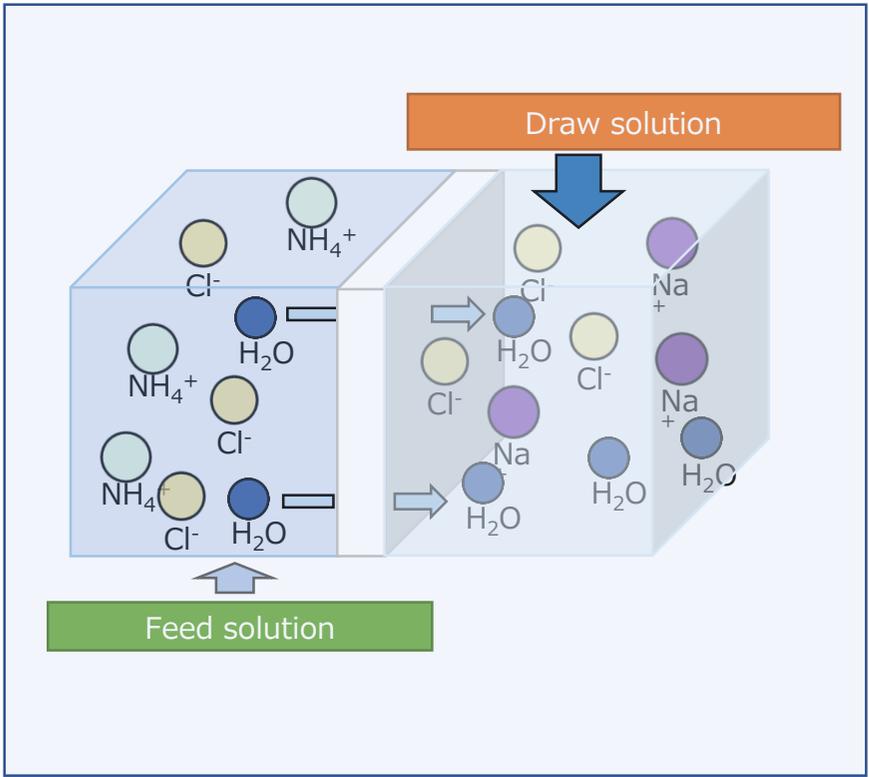
Separation performance
Materials performance

Model performances
calculation results

- Sizing
- Energy consumption
- CO₂ emission

Ex. membrane (FO) modelling & simulation

Ex. adsorption (PB)



Position in the project

Development and evaluation of ammonia concentration process by membrane separation

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R&D items

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- System and module design of MD (Hiroshima U)
- Evaluation of ammonia concentration process using membrane and adsorption (Yamagata U)

Achievement

- 10 times concentration of real wastewater by FO (Kobe U/Waseda U)
- 10 times concentration with T-N40,000ppm by BC (Kobe U)
- Production of 25wt% ammonia solution by MD (Kobe U/Hiroshima U)
- Model of each elemental separation process for ammonia concentration (Yamagata U)

