Development of a Simple Reprocessing Process Using Selective Precipitant for Uranyl Ions

- Engineering studies on systems for precipitating and separating -

KAZUHIKO YAMASAKI,[1] TAKAHIRO CHIKAZAWA,[1] YOSHIHISA TAMAKI,[1] TOSHIAKI KIKUCHI,[1] MASATOSHI HANZAWA,[1] MASANOBU NOGAMI,[2] YASUJI MORITA,[3] and YASUHISA IKEDA[4]

[1] Mitsubishi Materials Corporation

[2] Research Laboratory for Nuclear Reactors, Tokyo Institute of Technology

[3] Japan Atomic Energy Research Institute

[4] Institute of Research and Innovation

Background

Key Reaction: $UO_2^{2+} + 2NO_3^{-} + 2NCP \rightarrow UO_2(NO_3)_2(NCP)_2$

FBR reprocessing with NCP

U or U/Pu Precipitation → **Precipitate Separation** → **Processing for Nuclear fuel**

⇒ Mox fuel (U+Pu) & Blanket fuel (U) [DF100] for FBR reprocessing

Our approach for 3years(2002-2004)

Fundamental studies (beaker-scale) ⇒ **Engineering studies**

Fundamental studies (beaker-scale)

- Precipitation mechanism
- Condition for precipitation
- Decontamination factors of FP...etc.
- Resistance to γ-ray radiation

Engineering studies

- Scale-up experiment for
  - precipitation
  - precipitate separation
- Nuclear fuel manufacturing

↓

Evaluation of the system

Outline

Precipitation → (Transfer of slurry) → Separation

Fundamental studies

- High recovery of uranium by precipitation
- Decontamination factor (DF)

Engineering studies (2003-2004)

Design / construction of each apparatus and system

↓

Engineering experiment

Objective of Apparatus Development

- Cost Reduction
  - continuous operation
- Nuclear criticality safety
- Easy Maintenance
  - long-term stability

Precipitator design

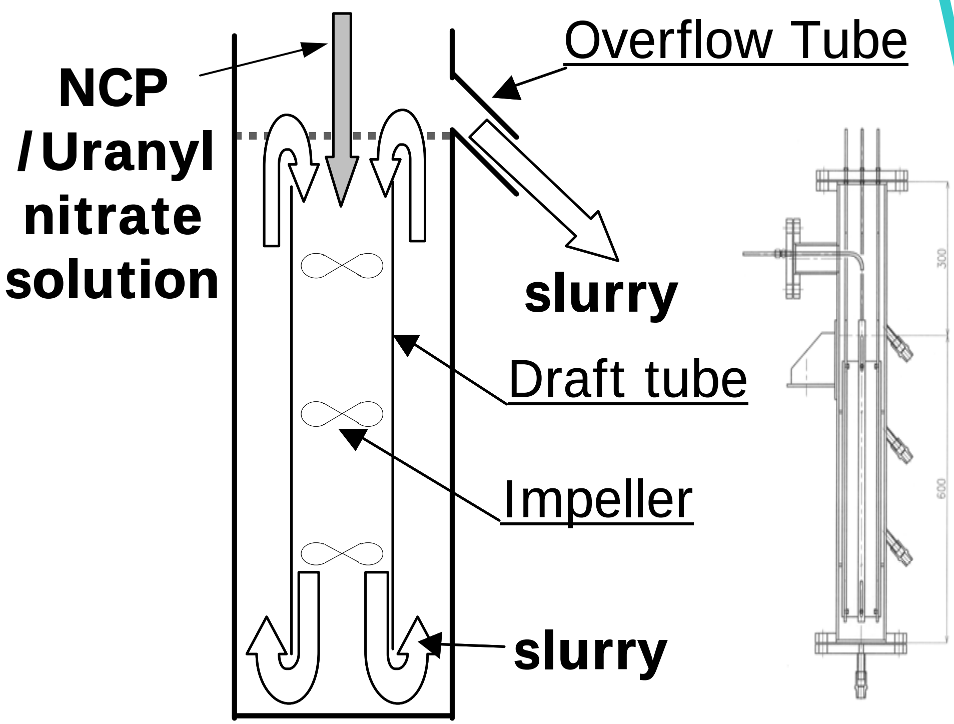
Requirements

- Nuclear criticality safety
- Continuous operation
- Long-term stability
- Sufficient mixing of reactants
- Avoiding NCP short-pass

Point of the experiment

- Completely mixed Flow (reaction & withdrawal)
- High reactivity (reaction rate, molar ratio)

Apparatus design



- Column with tall-shape for criticality safe by shape component
- Draft tube installation with effective impeller design

Type : cylindrical-longitudinal
Size: φ100mm×L900mm
Effective volume ：4L
Feed rate ：8L/hr
→ 1/20-scale of 200tHM/y

Precipitate separator design

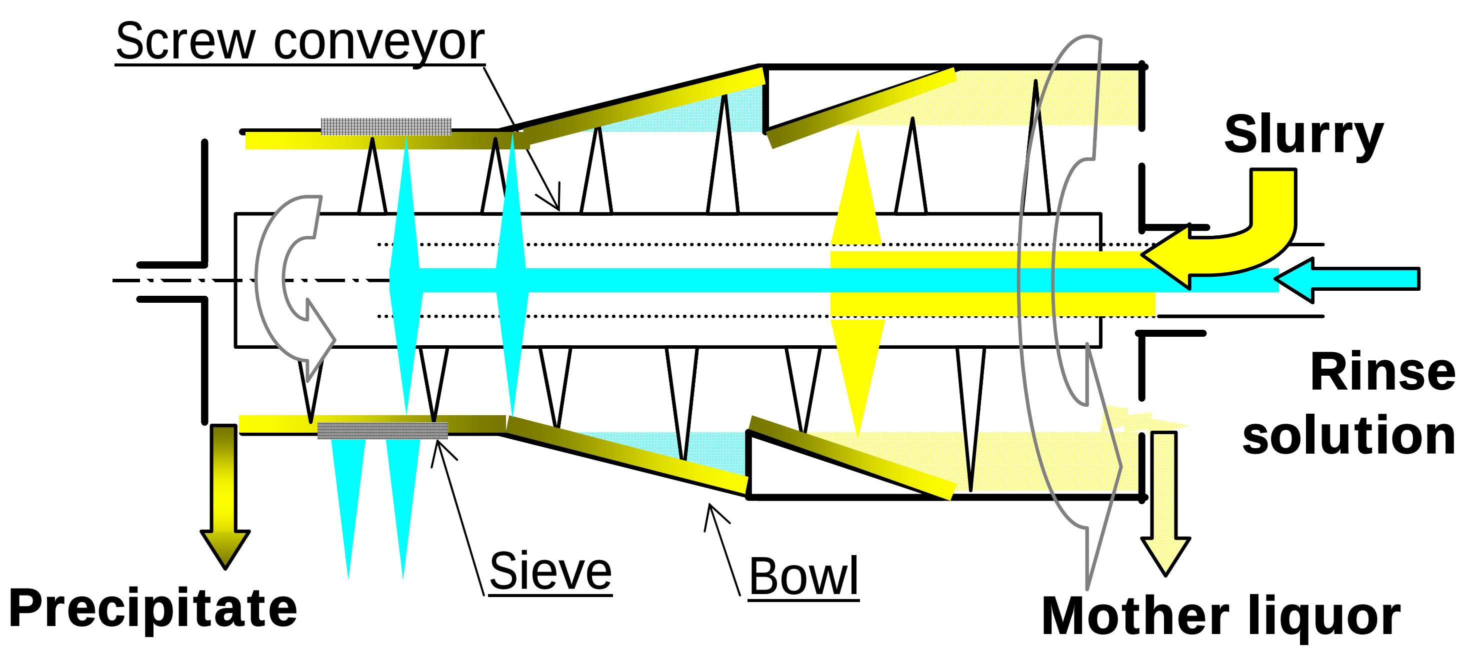
Requirements

- Nuclear criticality safety
- Continuous operation
- Long-term stability
- Rinsing (washing) ability for DF
- Avoiding leakage of solid to soln.

Point of the experiment

- High DF (target value, 100)
- Precipitate with low water content
- Low leakage of precipitant to solution

Apparatus design

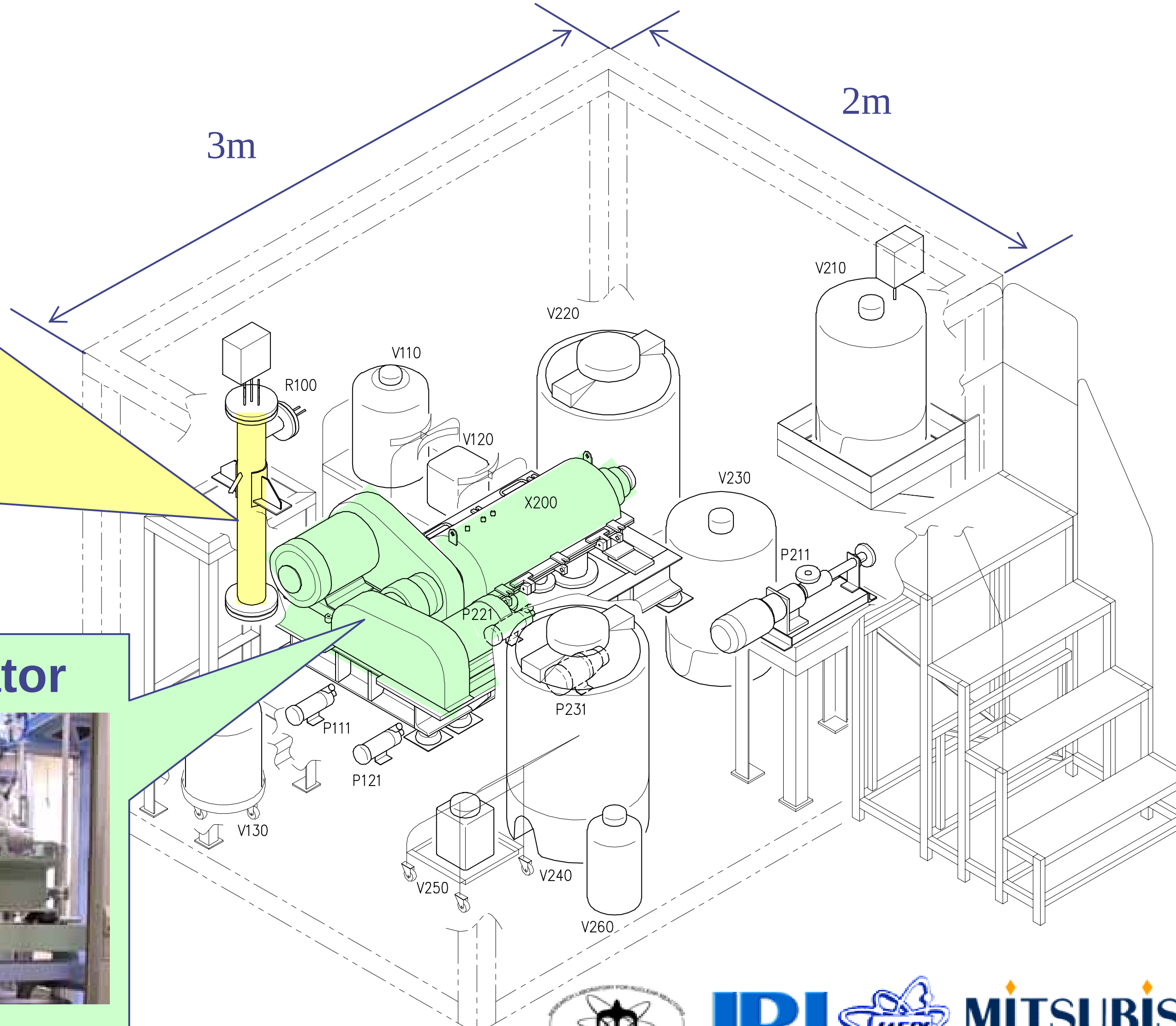


Type:
Centrifuge-type decanter
Size:
L1500×W1000 ×H500
Max Feed rate:
150L/hr
(200tHM/y)

Equipment for the engineering studies

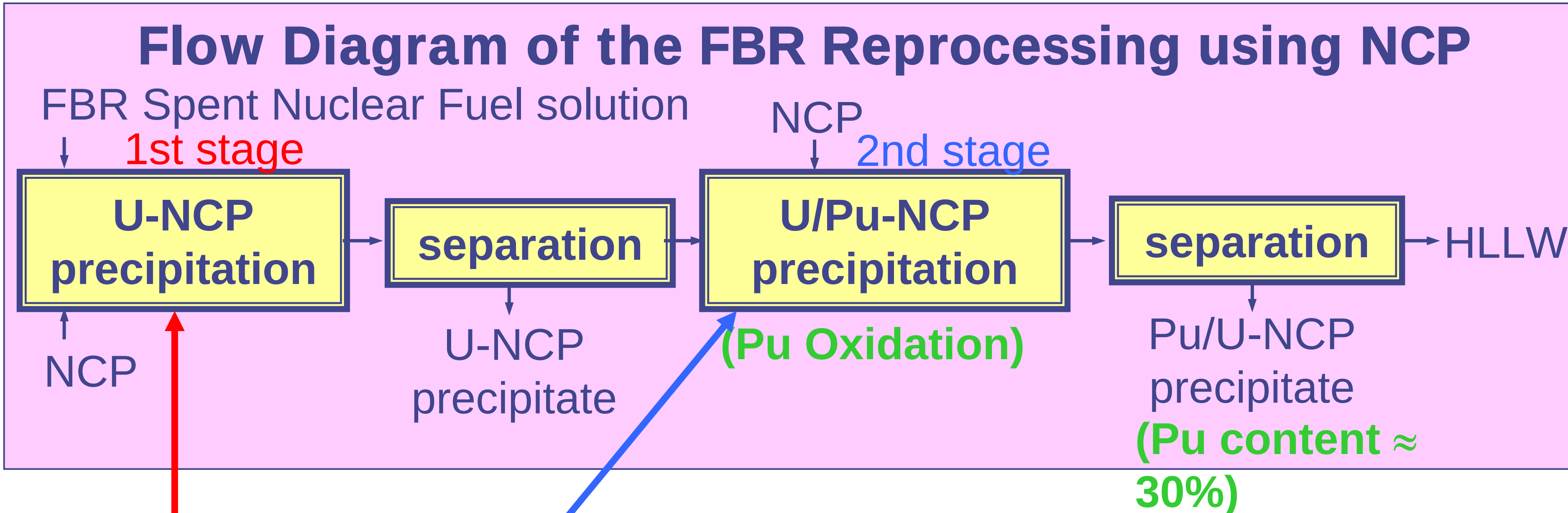
Precipitator

Precipitate Separator



IRI
JAERI
MITSUBISHI
MITSUBISHI MATERIALS

Precipitation experiment (1)



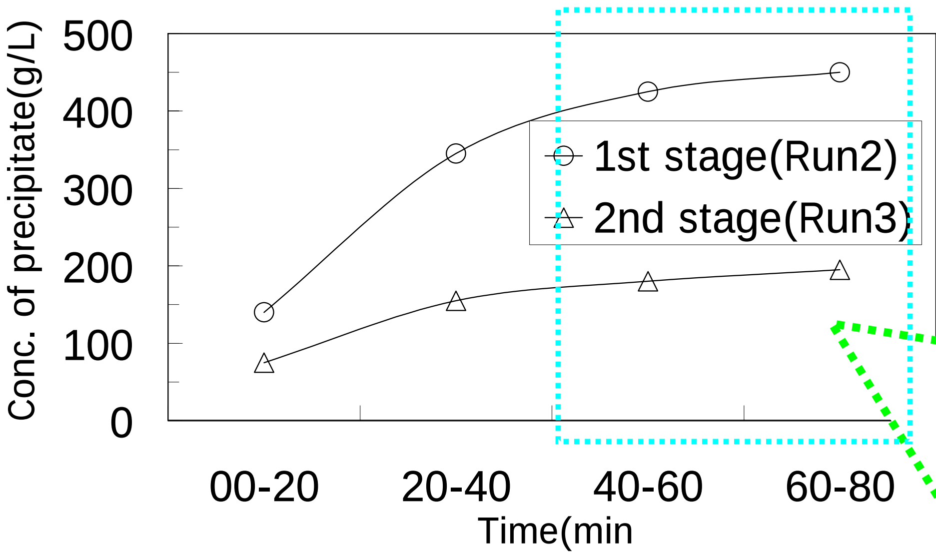
Experimental condition

Run No.	Feed solution: Dissolved solution, U (mol/L)	Feed solution: Dissolved solution, rate (mL/min)	Feed solution: NCP, Feed rate (mL/min)	Equivalent ratio of NCP to U	Residence time (min)
1	**1**	138	31	**0.7**	30
2	**1**	207	48	**0.7**	20
3	**0.3**	200	30	**1.5**	20

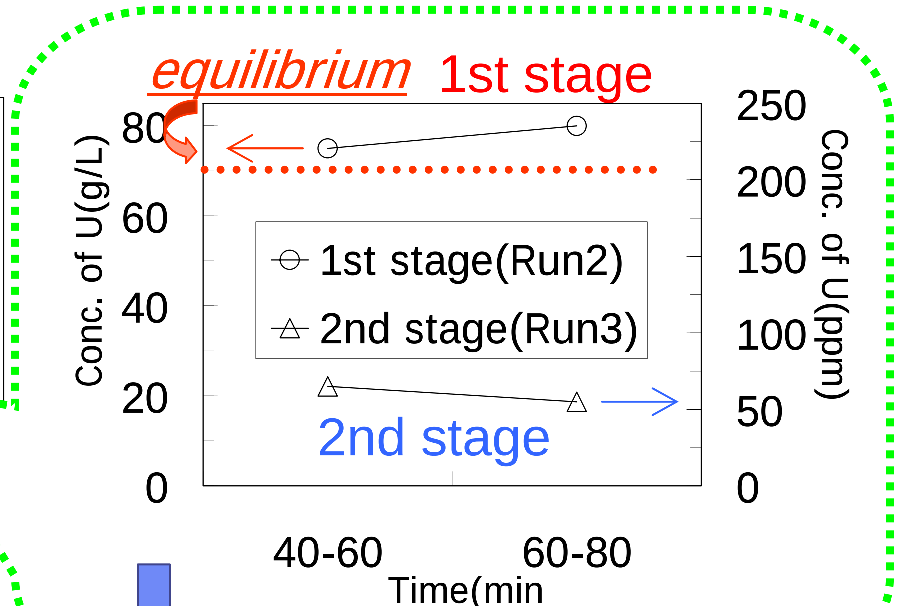
Precipitation experiment (2)

Results:

1. Stable withdrawals of slurry (mixed flow of precipitate and solution) were maintained during operation time (4 times longer than the stipulated residence time).



(1) Conc. of precipitate



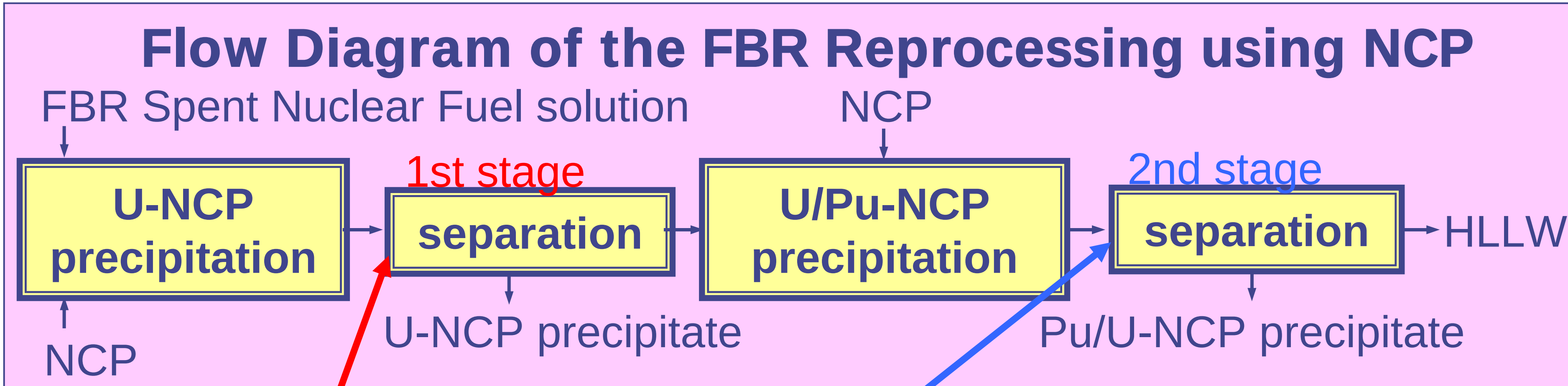
(2) Conc. of U

2. Sufficient mixing of reactants was achieved.

- 95% of NCP was used for formation of UO_2^{2+}-NCP precipitate in 1st stage.
- Over 99% of U was recovered as UO_2^{2+}-NCP precipitate in 2nd stage.

IRI JAERI MITSUBISHI MITSUBISHI MATERIALS

Precipitate separation experiment (1)



Experimental condition

Run No.	Feed slurry make-up condition			Feed rate		
	U (mol/L)	Equivalent ratio of NCP to U	Sr^{2+},Ce^{3+} (g/L)	Slurry (L/hr)	Rinse solution L/hr	Rinse solution Ratio*
1	**1**	0.7	1	30	**0**	—
2	**1**	0.7	1	30	**24**	**2**
3	**0.3**	1.5	1	30	**10**	**2**

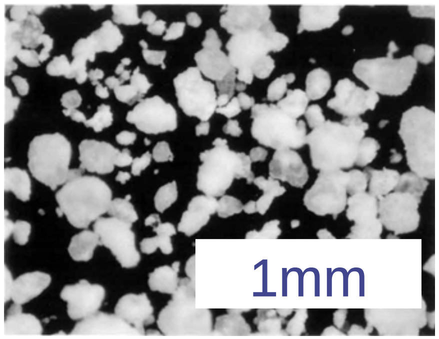
*Volume ratio of [rinse solution] / [precipitate]

Precipitate separation experiment (2)

Results:

1.high DF ,Low water content

Precipitate



Water content : 10-15%

Decrease in U conc. raises DF value.

Run No.	Condition for slurry	DF Sr	DF Ce	Rinse soln. (Ratio*)
1	1st stage (1mol-U/L)	8.2	9.7	–
2	1st stage (1mol-U/L)	33	31	2
3	2nd stage (0.3mol-U/L)	130	120	2

*Volume ratio of [rinse solution] / [precipitate]

The rinsing operation is effective for increasing decontamination of FP.

In progress for over DF100 at the 1st stage

2.Low leakage of precipitant to solution

Amount of precipitate in mother liquor was almost negligible.

Conclusion

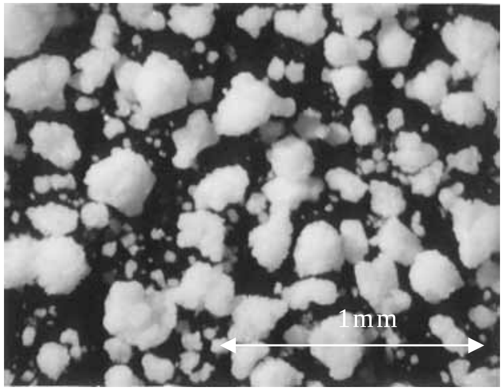
1. Apparatus and system for precipitation and precipitate separation, which enable continuous operation and nuclear criticality safety, was designed.
2. In the precipitation experiment, sufficient mixing of reactants in precipitator and stable withdrawal of slurry were confirmed.
3. In the precipitate separation experiment, effectiveness of the rinsing operation in increasing DF of FP and low leakage of precipitant to mother liquor were confirmed.

Future remarks

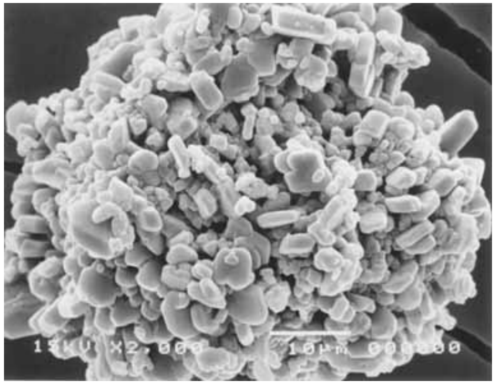
1. Confirmation of long-term stability for equipment.
2. Adjusting the operational condition to achieve over 100 of DF in precipitate separation.

Image of NCP precipitate

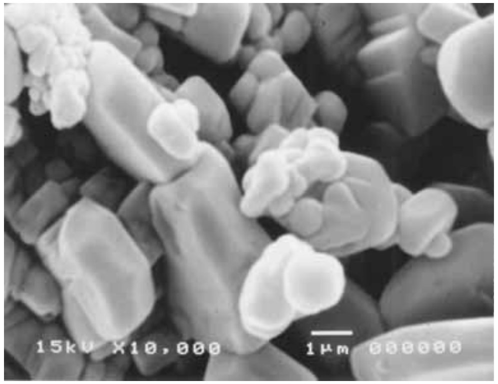
a. Outlook



b. Microscope image



c. SEM image(x2000)



d. SEM image(x10000)