2012 IPRC

Study on the Feed Form Fabrication for Oxide Reduction Process

29 August 2012

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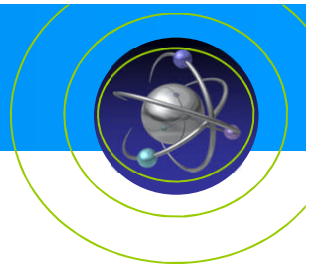
Content

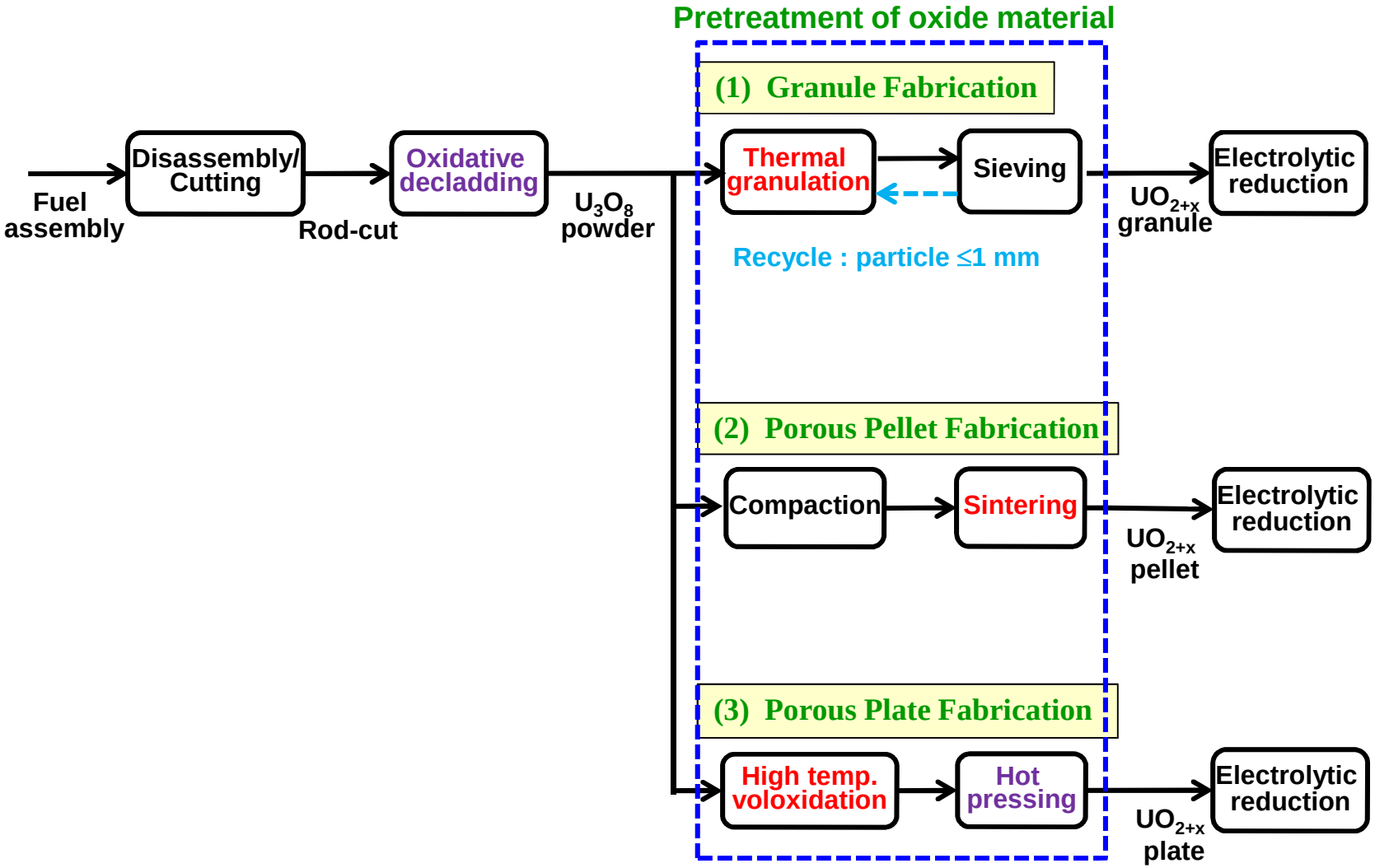


Introduction (I)

- Benefits of pretreatment(fission products removal)
  - Pt anode protection in OR process
  - Extension of salt replacement cycle due to very low Cs buildup rate in OR process
  - Salt purification by layer crystallization method and salt recycling in OR process
- Removal rate of fission products
  - Increase in diffusivity of fission products in hyperstoichiometric UO_{2+x}
- Review on the appropriate methods to fabricate feed forms using U_3O_8 powder
  - Mixing methods of excessive binder and lubricant for forming of U_3O_8 powder were excluded due to the bad effect on off-gas treatment during pretreatment
  - Selected techniques : (1) thermal granulation, (2) cold pressing and sintering, and (3) hot pressing

Introduction (II)

[Flow sheet of feed form fabrication process]

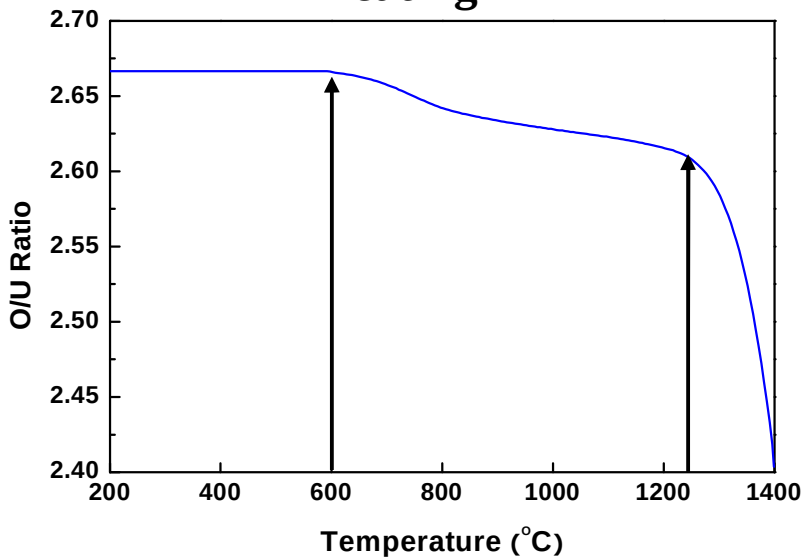


Reduction Behavior of U_3O_8

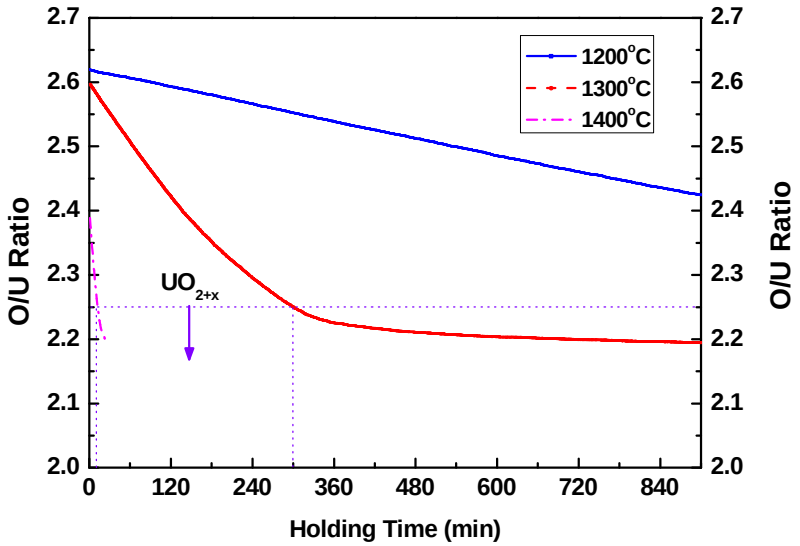
❑ Reduction Behavior of U_3O_8 into UO_{2+x} during Pretreatment

- Analysis using TGA
- Flowing gas
  - Ar : 99.999%(0.001% H_2O)
  - Ar flow rate : 0.1 L/min
- Reduction behavior (TG analysis)
  - ➢ Initiation of thermal decomposition : 600°C ,
  - ➢ Rapid decomposition : 1250°C
  - ➢ U_3O_8 phase converted into UO_{2+x} phase : after 5 h above 1300°C

<Heating>



<Holding>



Granule Fabrication (I)

- Design of Granule Fabrication
  - UO_{2+x} granule fabrication from U_3O_8 powder by thermal granulation in Ar using rotary voloxidizer (cylindrical chamber without baffles)
  - Particle ≤1 mm is recycled for thermal granulation

<Rotary voloxidizer>

Granule Fabrication (II)

- Granule(Particle Size > 1mm) Fabrication Using SIMFUEL (No rotation)
  - Agglomeration of U_3O_8 powder by sintering during pretreatment at high temperature
  - Weak bonding of agglomerate ➲ fine powder production during sieving : affected by sieving condition rather than pretreatment time
  - Granule fabrication in Rotating mode was carried

<Agglomerate preparation condition>

SIMFUEL	Sintering conditions	Sieving
UO_2	1200 °C. 10 hrs, (Ar)	15 min (Retsch AS200)
30,000 MWd/tU	1200 °C. 3 hrs, (Ar)	5 min
60,000 MWd/tU	1200 °C. 3 hrs, (Ar)	5 min

<Cumulative particle distribution>

Particle size	Cumulative Fraction (%)		
	UO_2	30,000	60,000
> 4 mm	18	0	0
> 2 mm	20	26	24
> 1 mm	21	35	27
> 75㎛	27	50	43
> 45㎛	39	66	56
< 45㎛	61	34	44

Granule Fabrication (III)

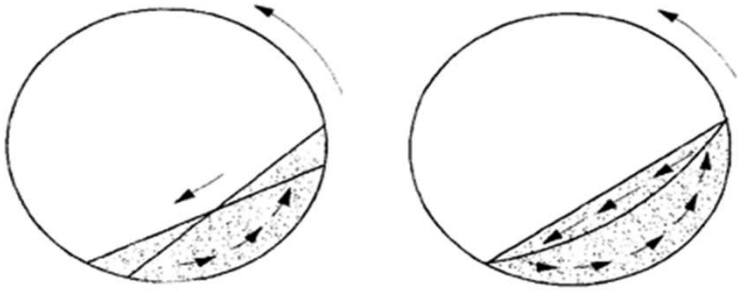
❑ Granule Fabrication

- Fabrication parameters
  - Initial particle bed motion of U_3O_8: slumping, rolling
  - Pretreatment temperature : 1150-1200°C, Time : 5-15 h
  - Rotation speed : slumping, 1-3 rpm
- Fabrication conditions considering properties and recovery rate of granule
  - 1200°C/10 h, 2 rpm (slumping)

<$UO_{2.49}$ Granule Properties>

Density (g/cm^3)	Porosity (%)	Recovery rate of granule(%)	Crushed strength(N)
4.38	55.3	~ 85 %	17

<Particle Bed Motion>



Granule Fabrication (IV)

❑ Granule Fabrication

- Strong and weak point of granule
  - Process simplicity, highly porous
  - Weak mechanical stability ➲ fine powder production during handling
- Granule properties were significantly effected by temperature

  ➲ related to rotary cylinder chamber material
- Requirement of chamber material(alloy) with resistance to high temperature corrosion for density enhancement

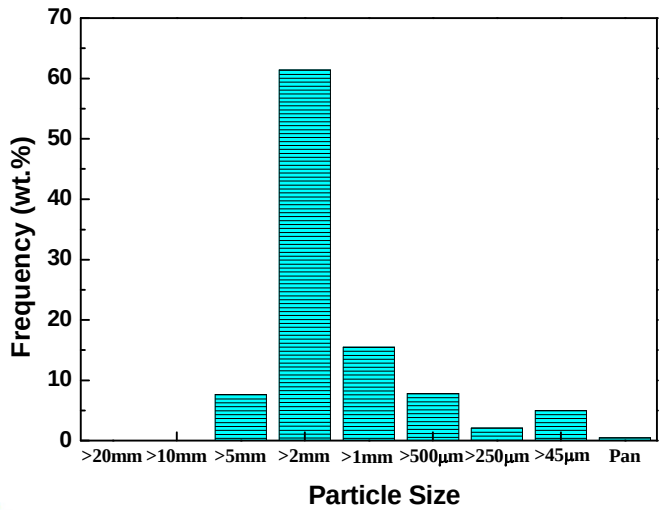
Temp.(°C)/10 hrs	Density (g/cm^3)	Crushed strength (N)	Recovery rate of granule (%)
1150	3.16	5	78
1200	4.38	17	85
Increasing rate	39%	340%	9%

Granulation Fabrication (V)

UO_2 Granule Fabrication

- Fabrication conditions
  - ❖ (200g U_3O_8 powder) ➲ Thermal granulation(1200 ^oC/10 hrs/2 rpm in Ar) ➲ Sieving ➲ (Granule) ➲ Reduction treatment of $UO_{2.49}$ granule(1000 ^oC/5 h in 4%H_2-Ar) ➲ (UO_2 Granule)
- Porosity and grain size by reduction treatment of $UO_{2.49}$ into UO_2 : hardly effected
- Mechanical stability by reduction treatment : hardly increased

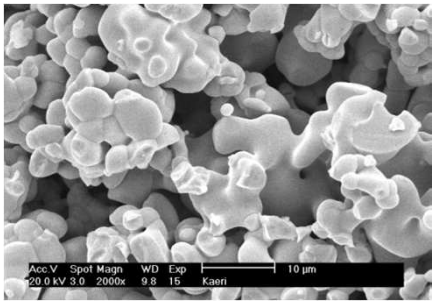
<Particle size distribution>



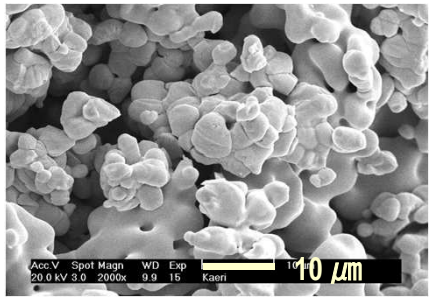
< UO_2 Granule>

Granule size (mm)	Density [% TD]	Crushed strength (N)
1~5	40	17

<GS of $UO_{2.49}$: 2~10 ㎛>



<GS of UO_2 : 2~10 ㎛>

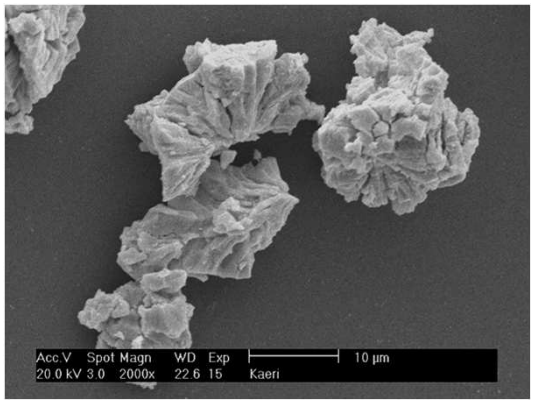


Porous Pellet Fabrication (I)

Design of Porous Pellet Fabrication

- To enhance mechanical stability of feed form
- Porous UO_{2+x} pellet fabrication from U_3O_8 powder by compaction and sintering in Ar and 4%H_2-Ar using ceramic chamber furnace and TMA
- Compaction without milling and lubricant mixing step

[U_3O_8 powder]



[Properties of U_3O_8 powder]

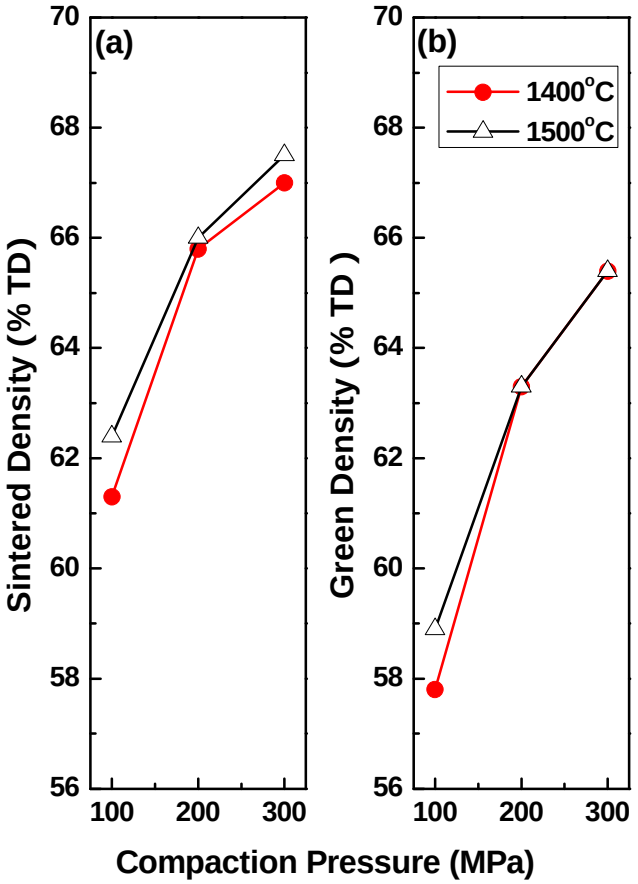
Powder	Average Particle Size(µm)	Specific surface area(m^2/g)	Bulk density		Tap density	
			(g/cm^3)	(% TD)	(g/cm^3)	(% TD)
U_3O_8	10	0.65	1.80	21.4	2.99	35.6

Porous Pellet Fabrication (II)

❑ Porous Pellet Fabrication

- Density control parameters
  - Compaction pressure of U_3O_8 powder : 100-300 MPa,
  - Pretreatment temp. of U_3O_8 green pellets: 1200-1500°C
- Time and atmospheric gas for pretreatment (sintering) : 10 h/Ar
  - ∗ Longer pretreatment time : Cs removal
- Conditions for reduction treatment of UO_{2+x} to UO_2 : 1200-1500°C, 2 h, 4%H_2-Ar
- Porous pellets were obtained

<Sintered density with compaction pressure>



❑ Compaction Pressure Effect

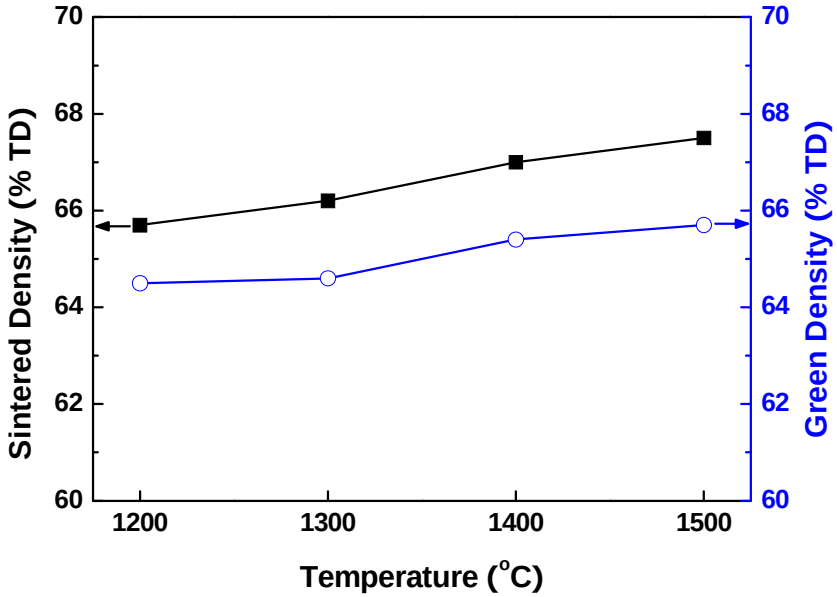
- Pretreatment temperature : 1400 & 1500°C
- Green densities of U_3O_8 pellets : 57.8 – 65.4 % TD(U_3O_8)
- Sintered densities of UO_2 pellets : 61.3 – 67.5 % TD (UO_2)

Porous Pellet Fabrication (III)

Pretreatment Temperature Effect

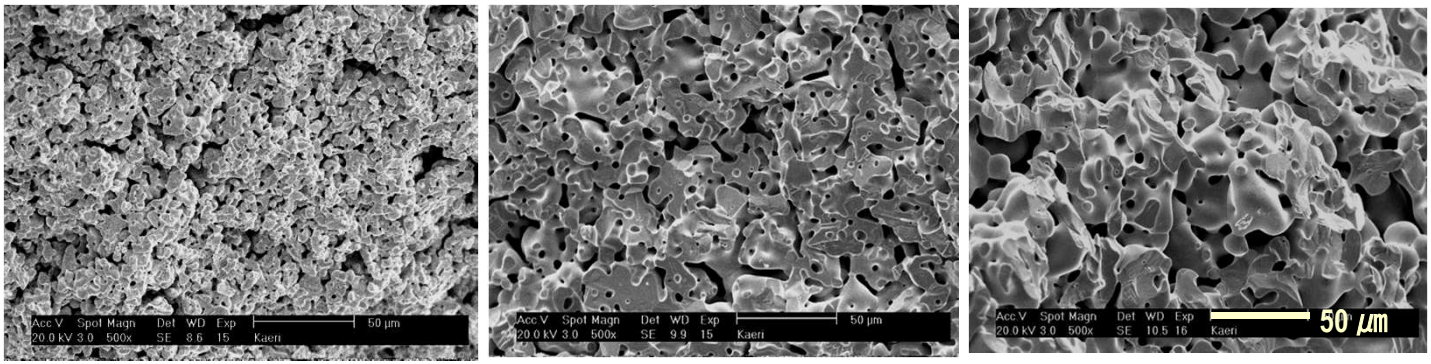
- Sintered densities : 65.7- 67.5 % TD(UO_2)
- SEM microstructure :
  - Grain growth : from 7.5 to 12.5 μm
  - Pore : growth and round shape

<Sintered density with temperature>



<SEM microstructure>

[1200°C] [1300°C] [1500°C]

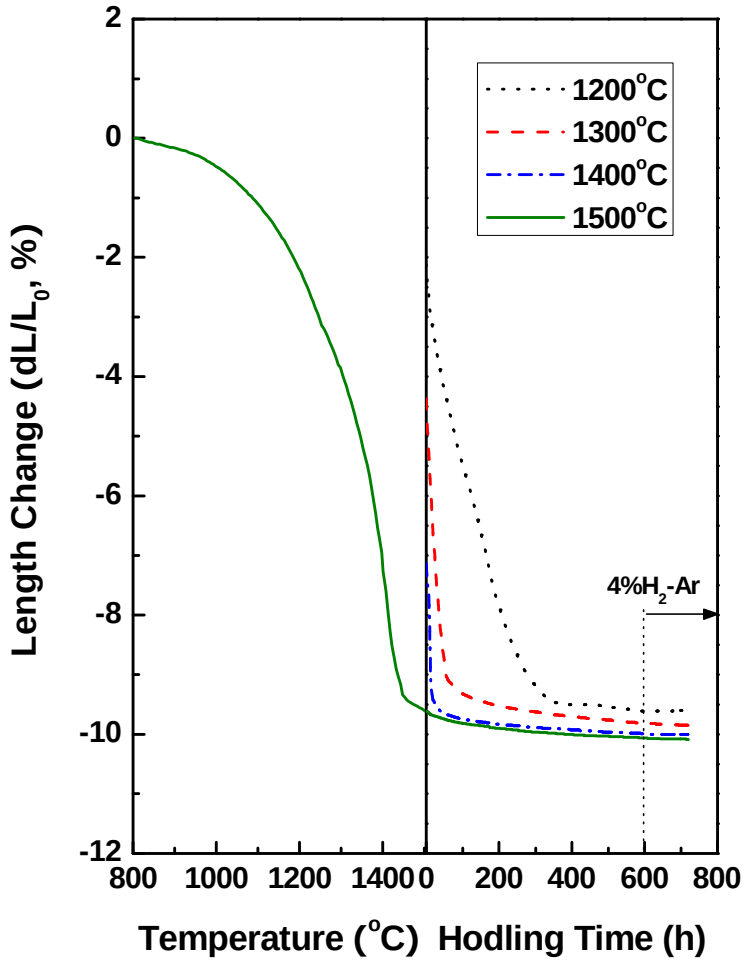


Porous Pellet Fabrication (IV)

Densification Behavior of U_3O_8 Green Pellet

- Analysis using TMA(Thermo Mechanical analyzer)
- Length change at isothermal condition:
  - Low temperature : continuously increase with time
  - High temperature : very slightly increase with time
- Reduction treatment : no effect of densification

<Length change of U_3O_8>



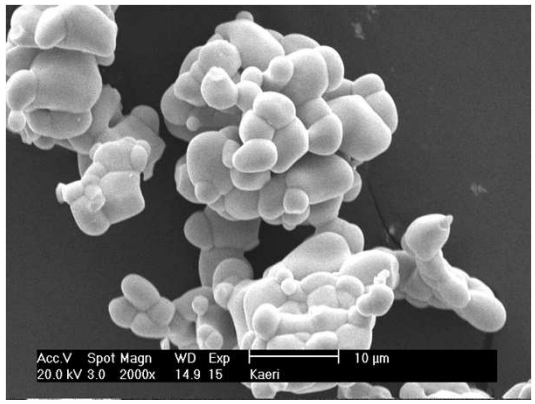
Pretreatment Conditions for Porous Pellet Fabrication

- Sintered densities of porous pellets : very small difference with temperature
- Removal conditions of fission products : determine the pretreatment conditions
- Reduction treatment temperature : drop

Porous Plate Fabrication (I)

- Design of Porous Plate Fabrication
  - To enhance the compactibility and sinterability of the thermally aggregated UO_{2+x} particles produced by high temperature voloxidation (pretreatment)
  - ∗ Green pellet from the thermally aggregated UO_{2+x} particles could not be obtained by cold pressing without a mixing of lubricant or binder
  - ∗ Low specific surface area : 0.13 m^2/g
  - Hot pressing method : introduction to improve the compactibility and sinterability

[Particles produced by pretreatment : 1200°C for10 h in Ar]

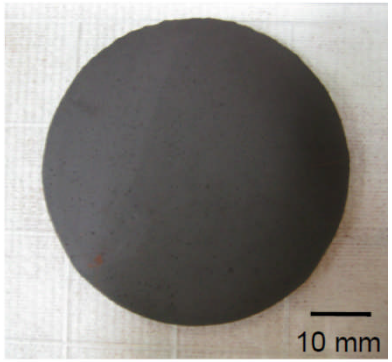


Porous Plate Fabrication (II)

❑ Porous Plate Fabrication

- Weight of $UO_{2.49}$ loaded in a graphite mold of 50-mm diameter : 50 g
- Density control
  - Hot pressing pressure : 20 MPa,
  - Hot pressing temp. & time : 1200°C/2h
- Atmospheric gas for sintering : Ar
- Porous plate was obtained

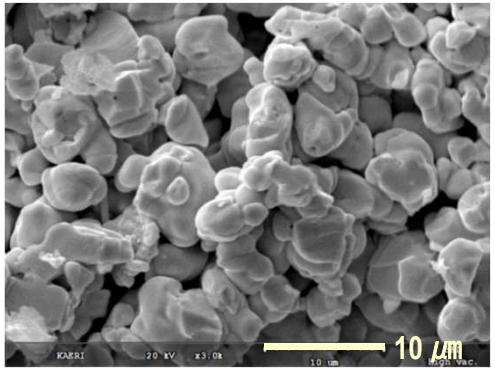
<Porous plate >



❑ Porous Plate

- Sintered density : 69% TD

< SEM microstructure >



❑ Requirements for Porous Plate Fabrication

- Sieving and grinding process with an increase in pretreatment temperature
- Special mold and punch for remote operation
- Press of high throughput due to batch-wise operation

Summary

❑ Feed Form Characteristics

Feed form	Density control	Size control	Packing density	Feed handling
Granule	limited	limited	Poor	- Fine particle production - Good flowability
Pellet	easy	easy	Medium	- Preservation of form - Good flowability
Plate	easy	easy	[1]	- Preservation of form - Poor flowability

❑ Oxide Reduction Characteristics (Test Results of OR Team)

Feed form	Feed form		Oxide reduction rate	Processing capacity	Salt carryover	Form stability
	Density (%TD)	Size (mm)				
Granule	40	1-5	Fast	Poor	High	Poor
Pellet	55-80	ϕ8x7H-ϕ9x8H	Medium	Excellent	Low	Excellent
Plate	69	crushed	Medium	[1]	NA	Excellent

[1] The processing capacity of the plate can be affected by its configuration in the basket and the shape of its pieces because the plate is horizontally large, flat and thin.

Thank you for your attention

Clean Energy! Clean Korea!