ULSAN NATIONAL INSTITUTE OF SCIENCE AND TECHNOLOGY

2022 춘계 KNS 워크샵 발표

무붕산 자율운전 소형원자로용 UO_2-U_3Si_2 복합 핵연료

안 상준*

울산과학기술원 | 원자력공학과 | 핵연료및재료연구실

2022. 5. 18.

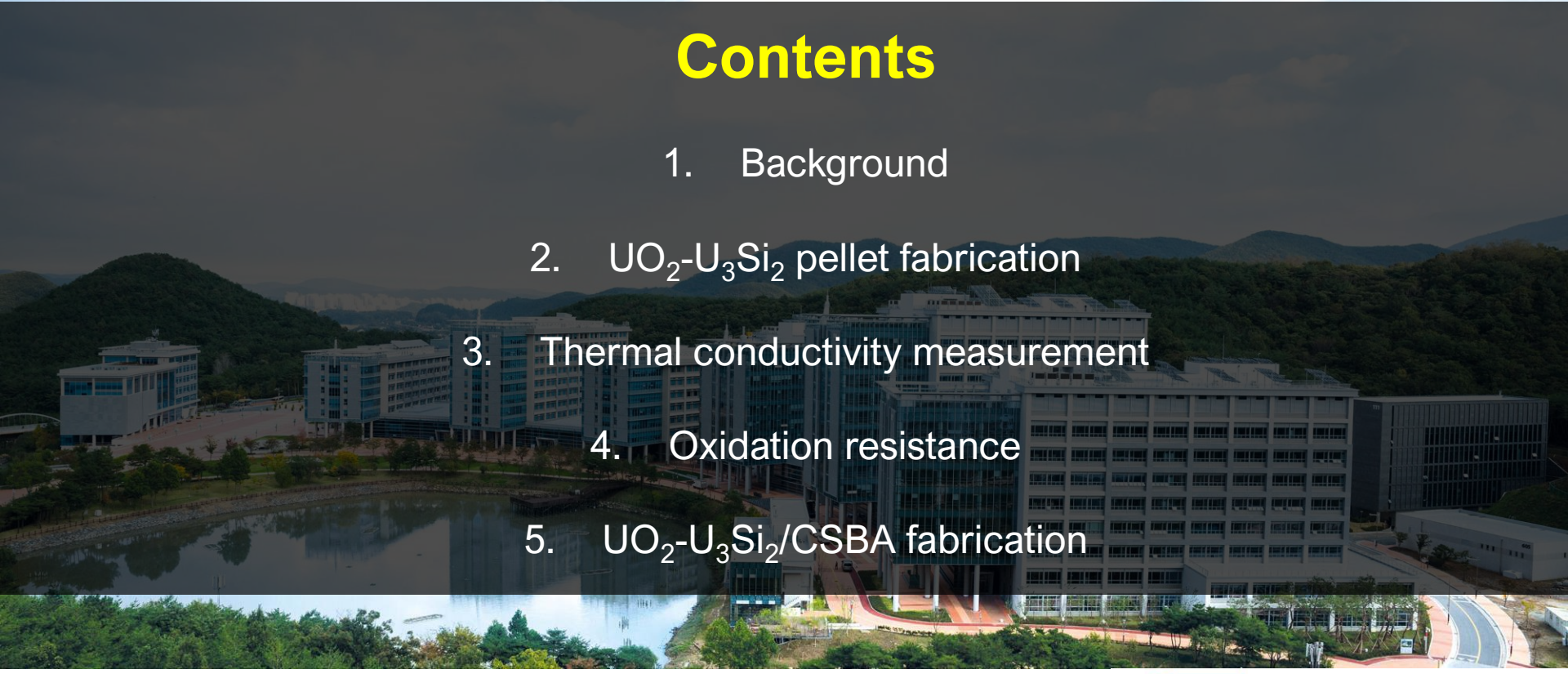
*UNIST RAdioactive NUclear Materials (URANUM) Laboratory

URANUM NUCLEAR FUEL LAB

ULSAN NATIONAL INSTITUTE OF SCIENCE AND TECHNOLOGY

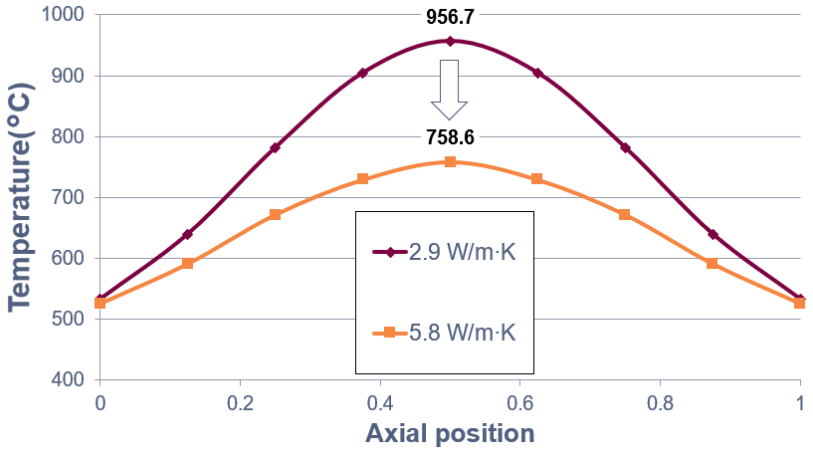
Contents

1. Background
2. UO_2-U_3Si_2 pellet fabrication
3. Thermal conductivity measurement
4. Oxidation resistance
5. UO_2-U_3Si_2/CSBA fabrication

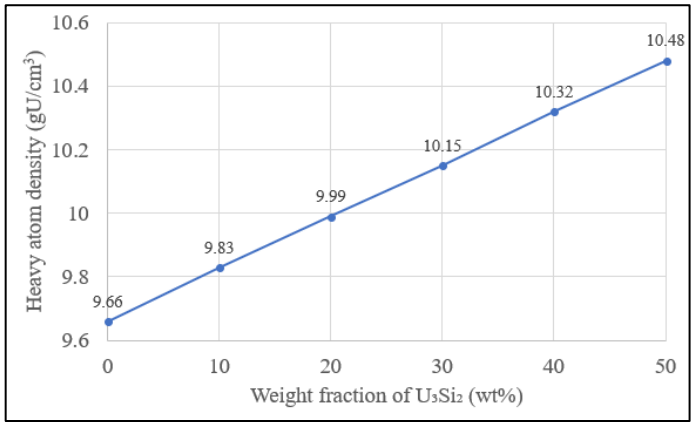


High thermal conductivity fuel for an advanced SMR

- **To achieve autonomous boron-free load-following operation**
  - Low T fuel is required → high thermal conductivity fuel is needed.
- **Strategy to achieve '*high k*' fuel w/o decreasing fissile density & oxidation resistance**
  - UO_2-based *high k* ceramics: UO_2 + U_3Si_2 or UN
    - SPS(Spark Plasma Sintering): High density & fast sintering



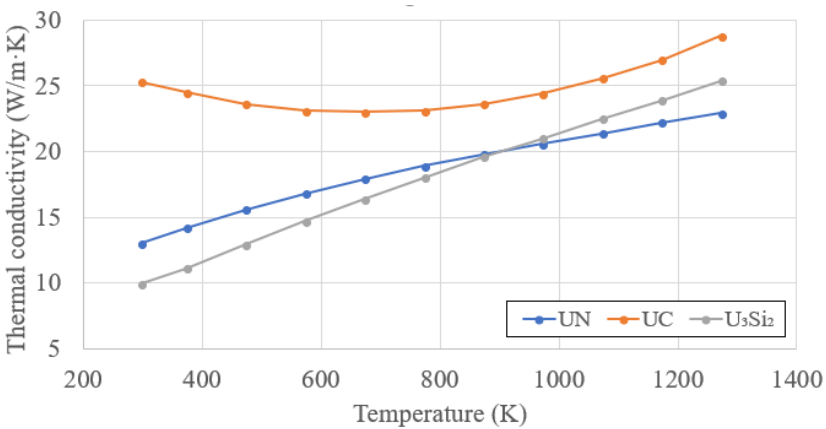
Ab-initio calculation of fuel temperature with fuel thermal conductivity



Fissile density increase with U_3Si_2 composition of UO_2-U_3Si_2 composite pellet

Additives for UO_2-based *high k ceramic*

Fuel	UO_2	UC	UN	U_3Si_2
Density (g/cm^3)	10.96	13.6	14.3	12.2
Heavy atom density (gU/cm^3)	9.66	12.9	13.5	11.31
Melting point (℃)	2865	2350	2850	1665
Thermal conductivity at 1000 ℃ (W/m·K)	2.9	28.8	22.9	25.4
Compatibility with H_2O	Very good	Bad	Bad	Average

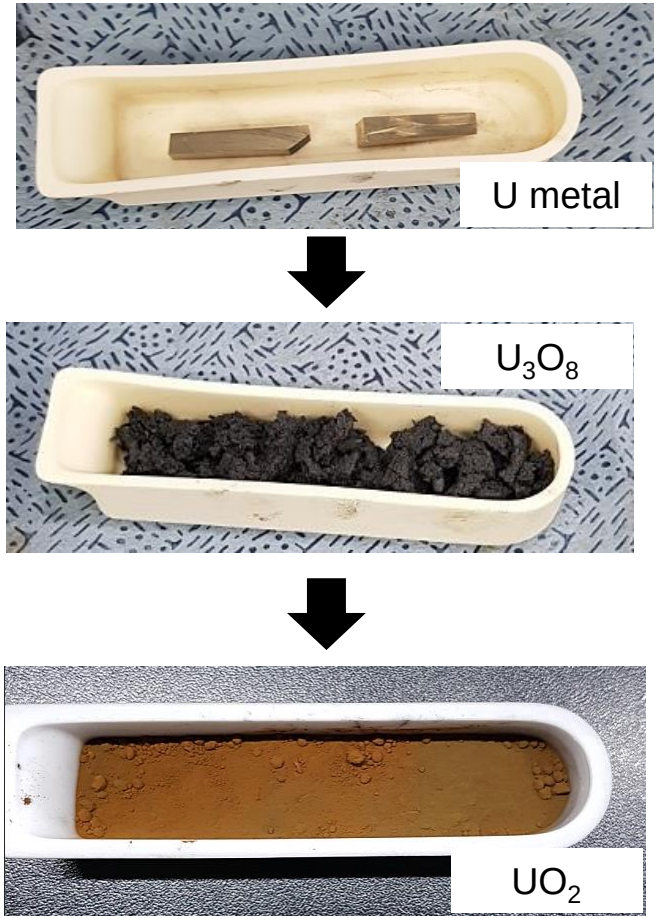


Thermal conductivity of non-oxide ceramic fuel

U_3Si_2 as the material to be tested due to;

1. High thermal conductivity
2. High fissile density
3. Acceptable oxidation resistance
4. Fabrication experience as fuel for research reactor

UO_2 powder preparation



U metal

Oxidization (800 ℃, 4 h, O_2)

U_3O_8

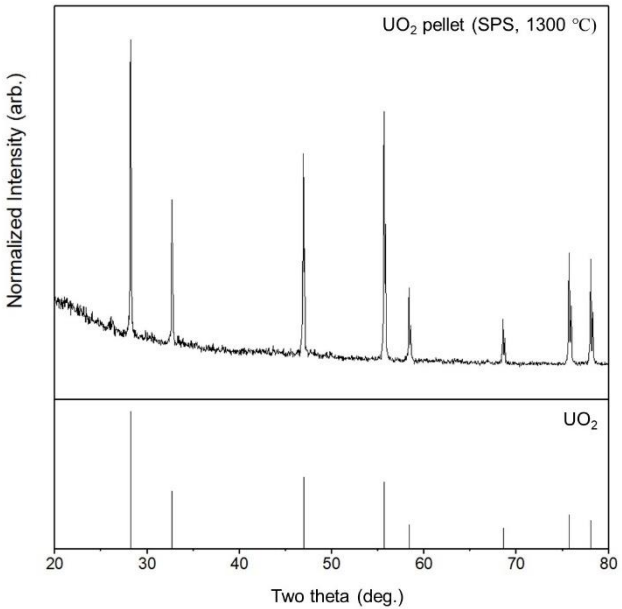
Reduction (800 ℃, 6 h, Ar-4%H_2)

UO_2

Milling

Sieving

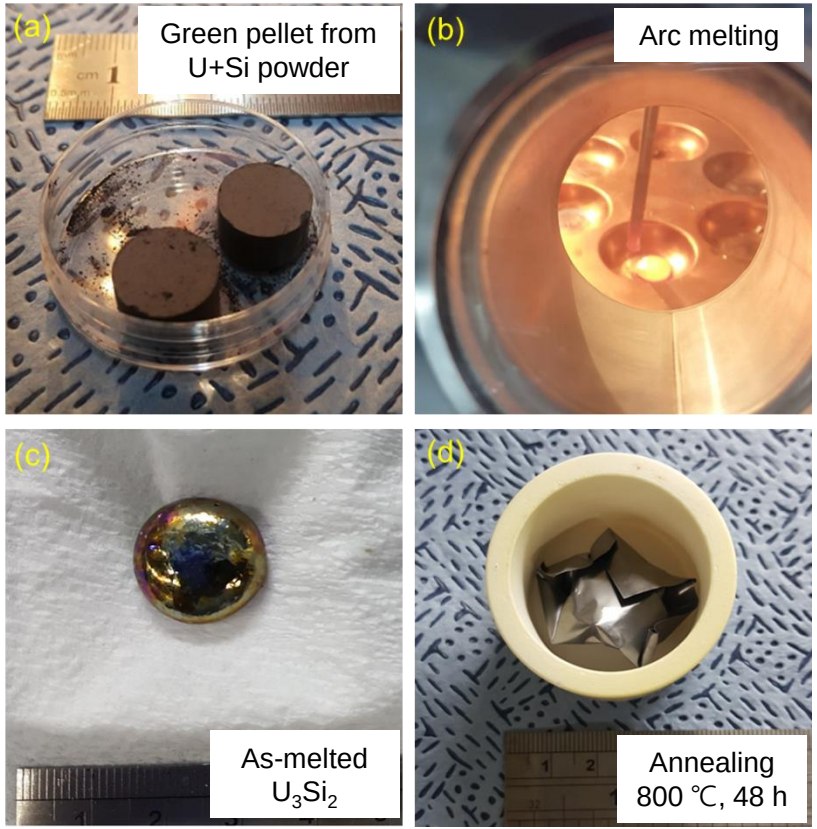
UO_2 fine powder (1~5 μm)

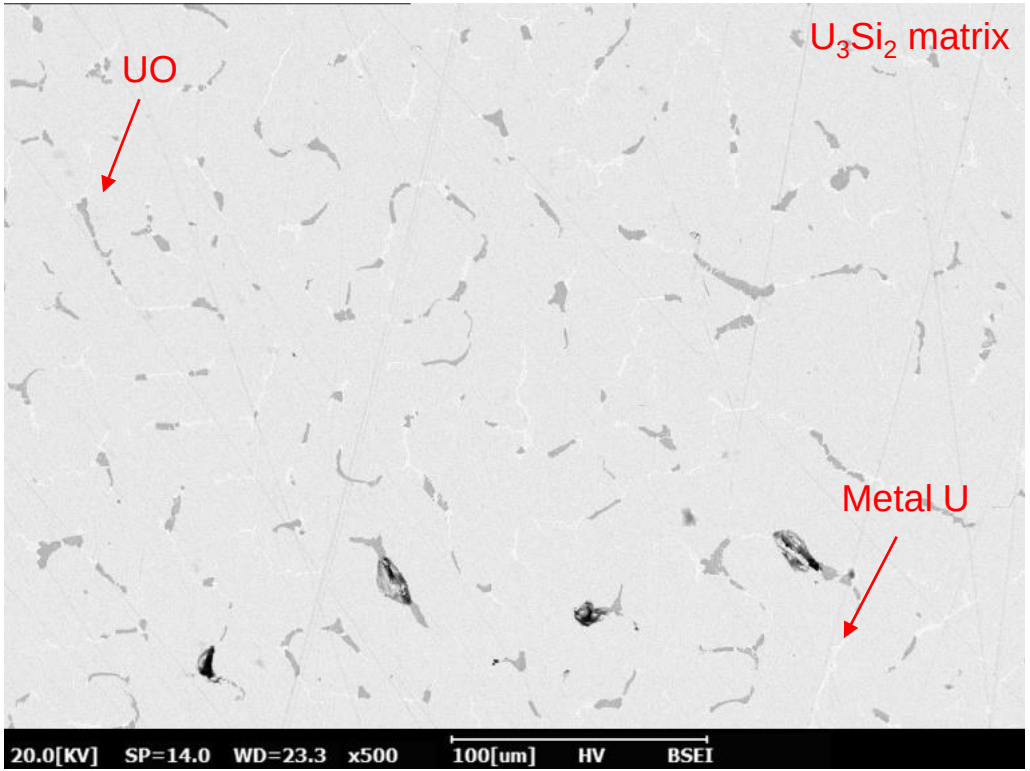


XRD analysis on UO_2 pellet

U_3Si_2 synthesis from U and Si powder

- **Oxidation occurred during process and metal uranium phase identified**

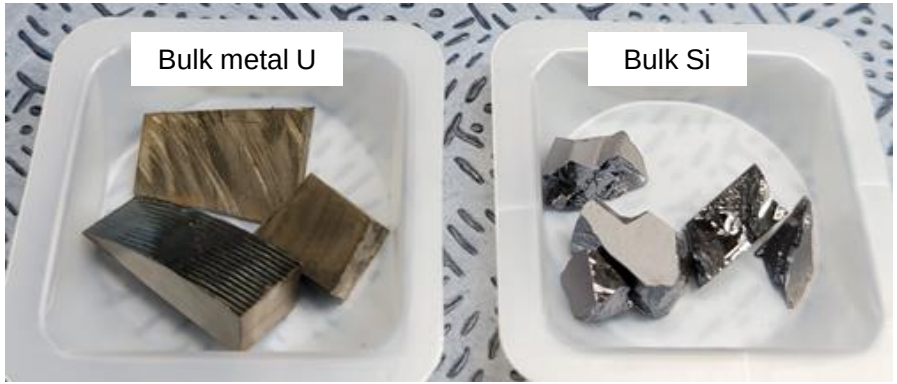


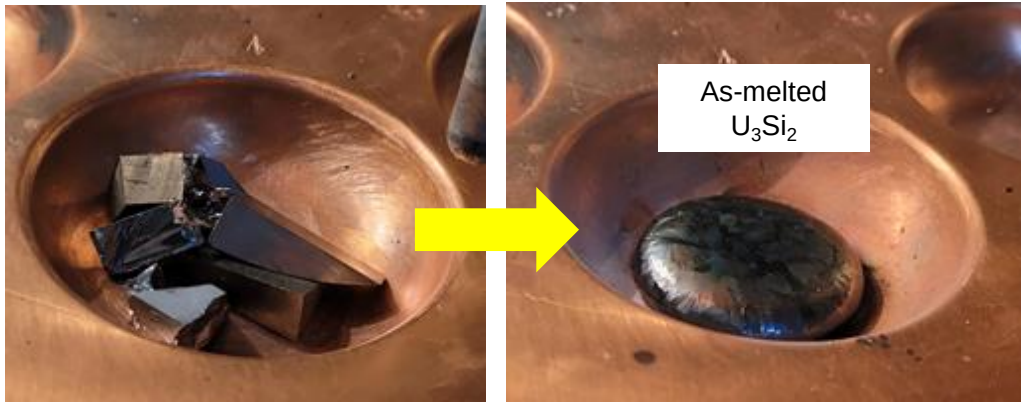


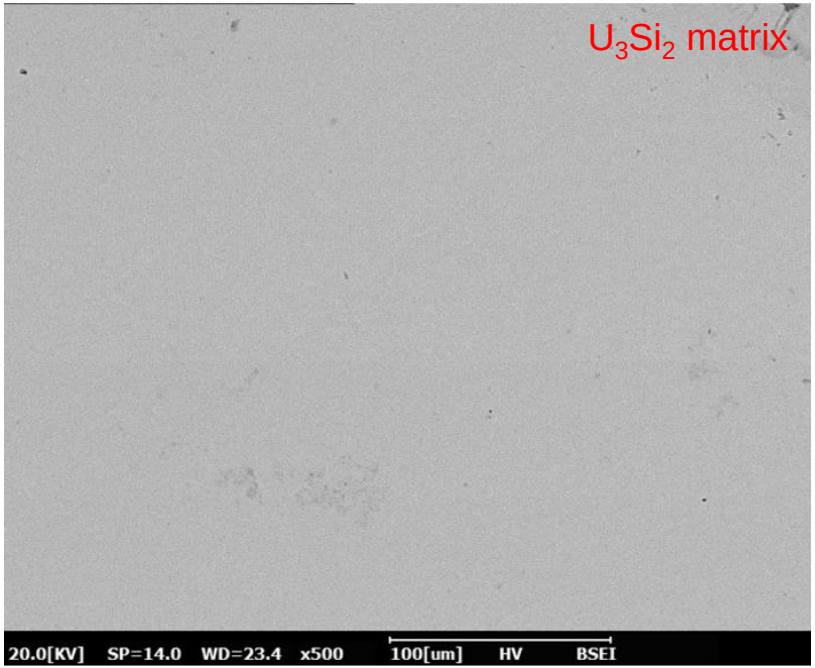
SEM BSE image of as-melted U_3Si_2

U_3Si_2 synthesis from bulk U and Si

- **No oxidation and 2nd phase formation**







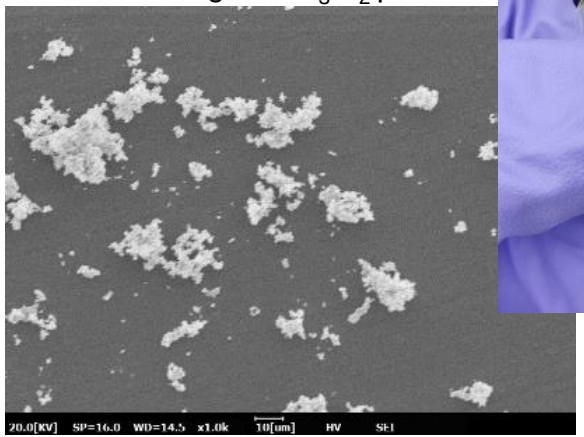
SEM BSE image of as-melted U_3Si_2

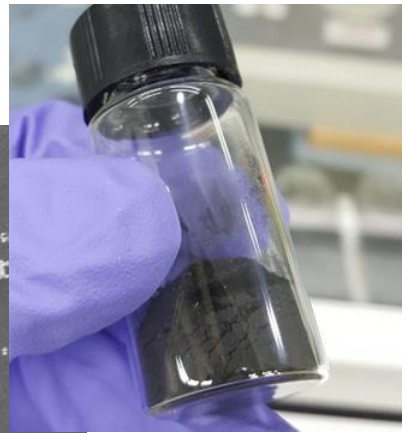
U_3Si_2 pulverization and spark plasma sintering

- **U_3Si_2 pulverization by milling**
  - Milling at 450 rpm, 8 h, Ar atm.
  - Additional annealing to as-milled U_3Si_2 powder to remove accumulated strain during milling
    - Annealing at 800 ℃, 12 h

- **Pellet fabrication through SPS (Spark Plasma Sintering)**
  - SPS: advanced sintering technique using DC pulse current
  - Sintering at 1300 ℃, 10 min, vacuum

SEM-BSE image on U_3Si_2 powder

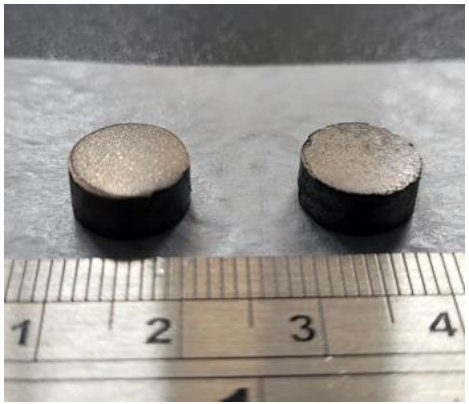




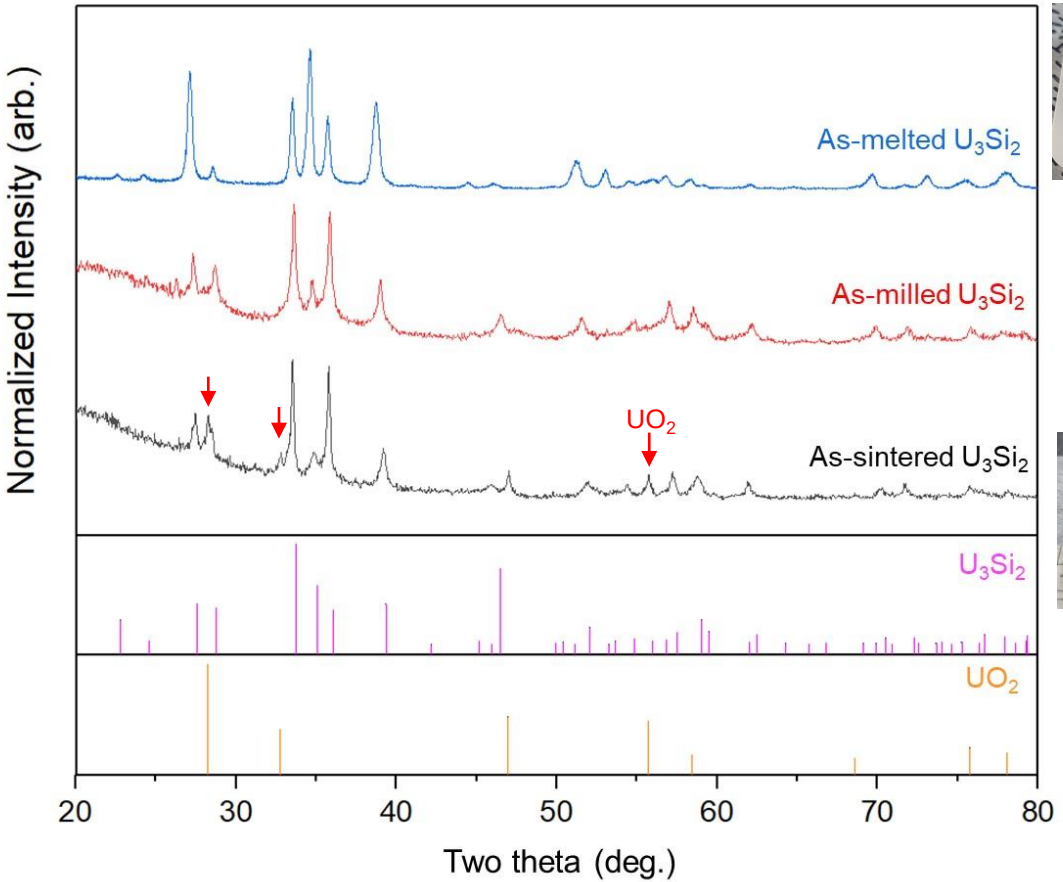
U_3Si_2 powder

Spark plasma sintering on U_3Si_2 powder

U_3Si_2 pellet (93–95%TD)

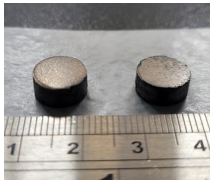


XRD analysis on each powder metallurgy process



Bulk U_3Si_2

Powder U_3Si_2



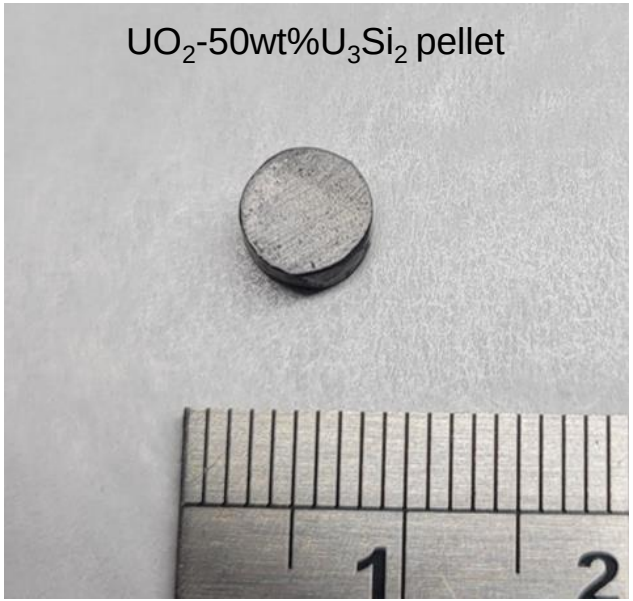
U_3Si_2 pellet

UO_2-U_3Si_2 composite pellet fabrication using SPS

- **Blending UO_2 and U_3Si_2 powder using 3d mixer.**
- **SPS with various silicide composition and sintering temperature.**
  - Sintering pressure (20 MPa) and dwell time (10 min) are fixed.

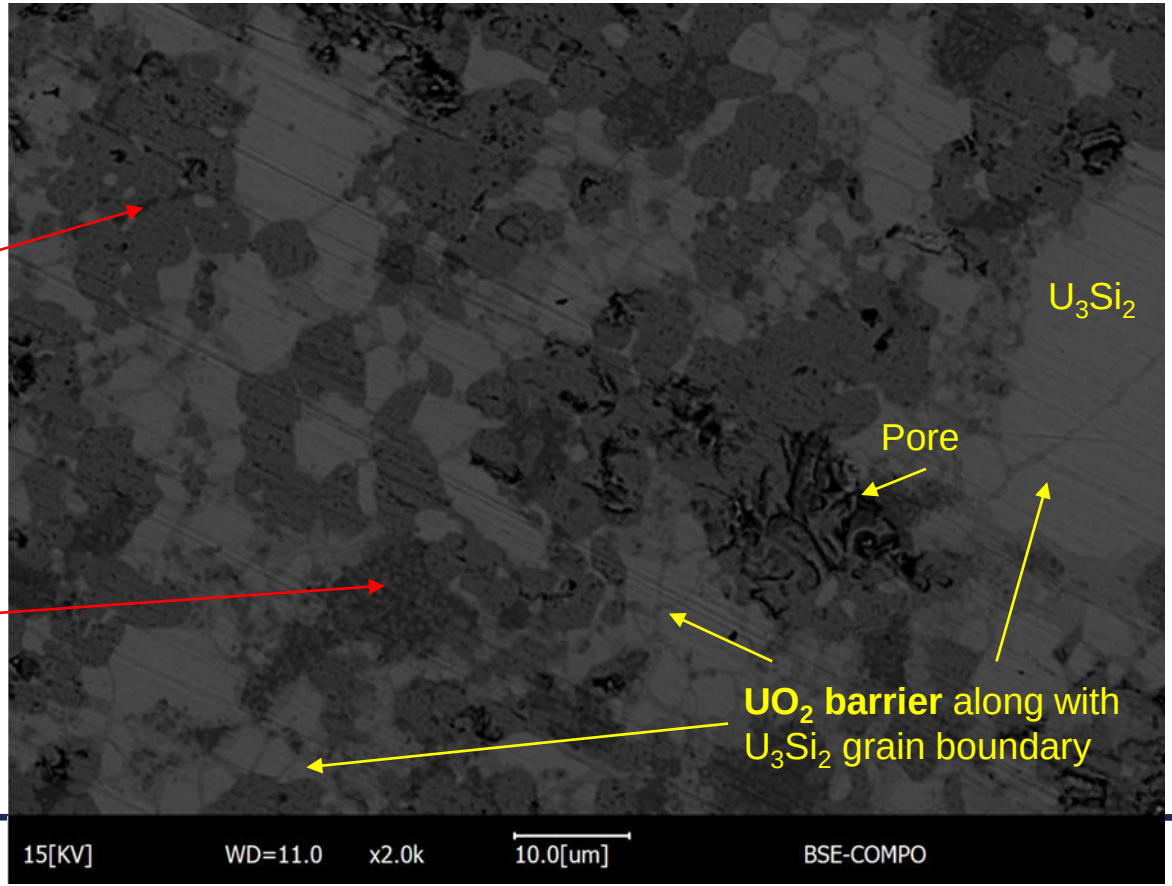
Sample	U_3Si_2 composition (wt%)	Sintering temperature (℃)	Sintered density (g/cm^3)
1	10	1400	9.62
2	30	1400	10.08
3	50	1400	10.18
4	50	1200	9.53
5	50	1000	7.27

*Theoretical density of UO_2: 10.96 g/cm^3
U_3Si_2: 11.32 g/cm^3



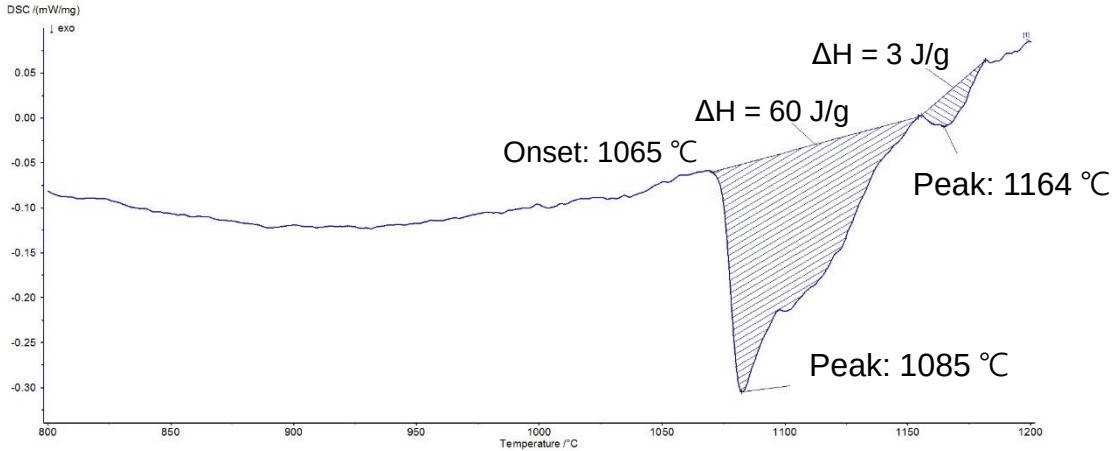
Micrograph analysis on UO_2-50wt%U_3Si_2 pellet

- **UO phase formation during sintering.**
- **UO_2 barrier was formed along with U_3Si_2 grain boundary.**

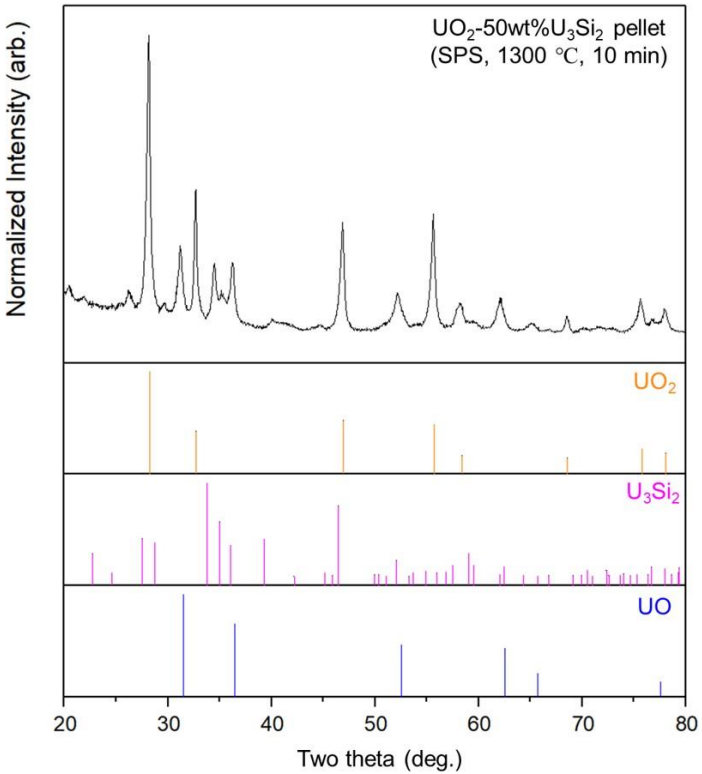


2nd phase formation during SPS of UO_2-U_3Si_2

- **XRD analysis on UO_2-50wt%U_3Si_2 sintered pellet**
  - Showed secondary UO phase peak.
- **DSC analysis on UO_2 and U_3Si_2 powder**
  - Revealed exothermal reaction occurred.
    - From 1050 ℃ to 1200 ℃.

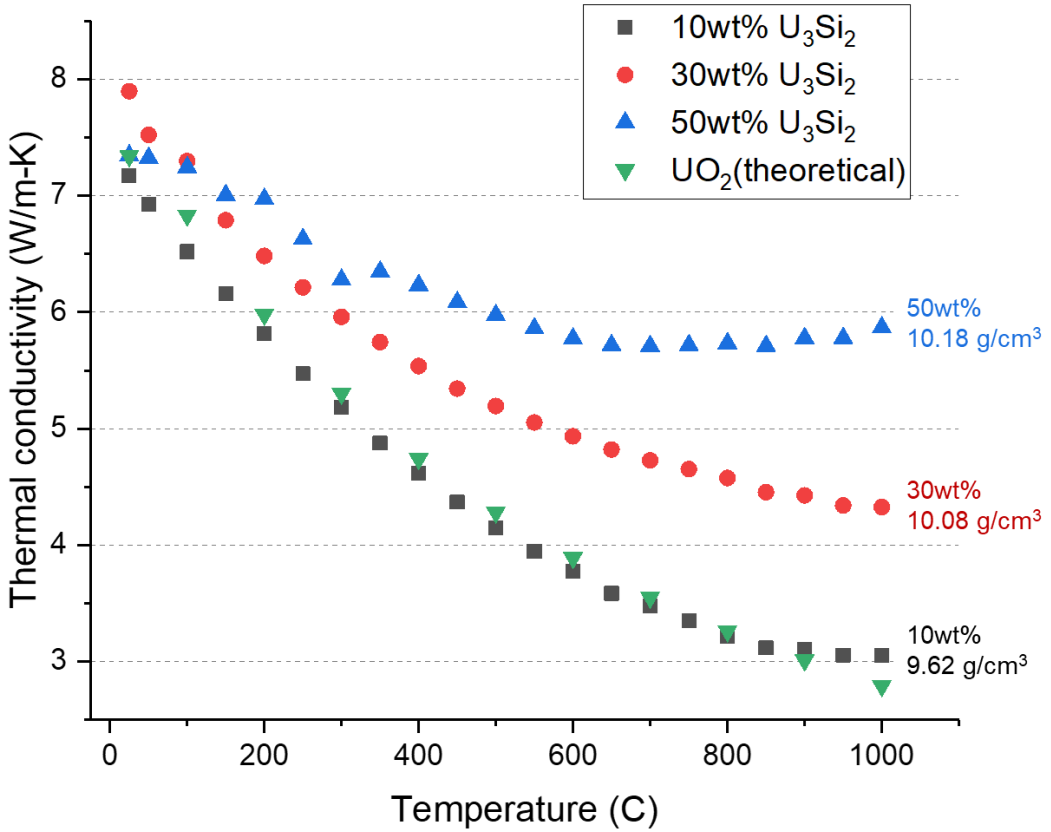


DSC analysis on UO_2-U_3Si_2 mixed powder up to 1200 ℃

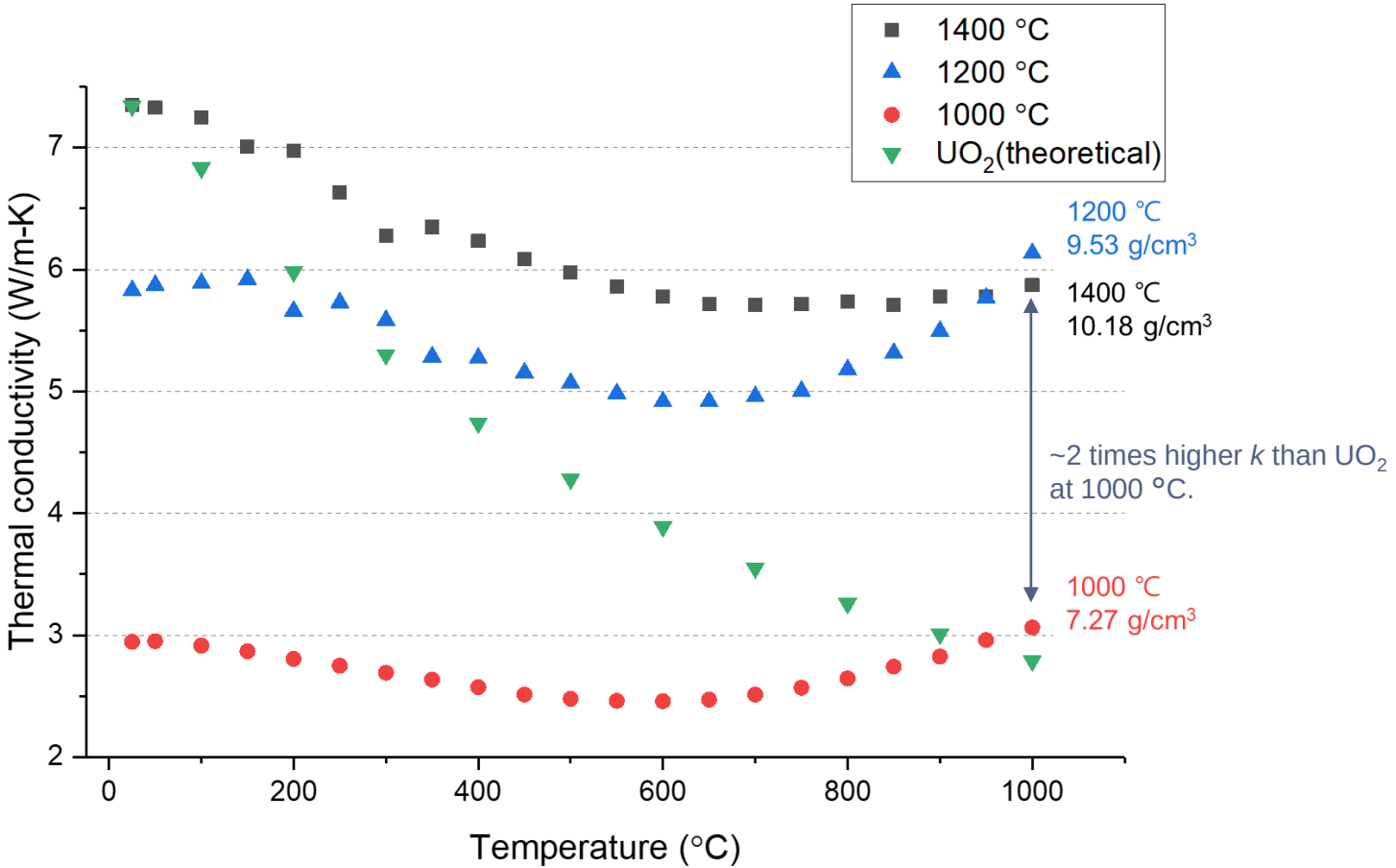


XRD analysis on UO_2-50wt%U_3Si_2 pellet SPSed at 1400 ℃ for 10 min

k of UO_2-U_3Si_2 pellet as U_3Si_2 composition

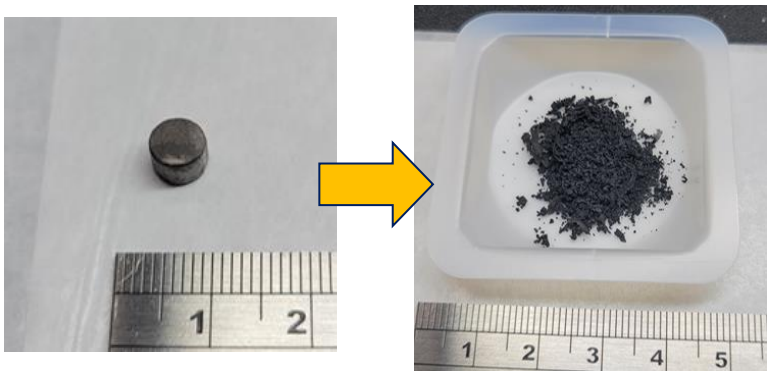


k of UO_2-U_3Si_2 pellet as sintering temperature

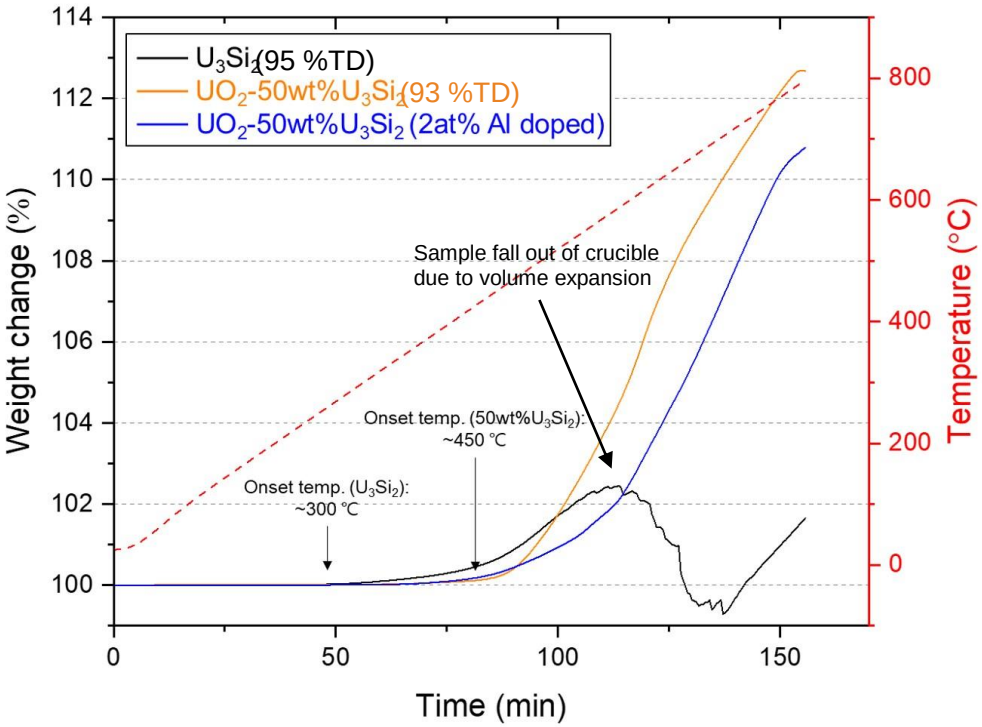


Oxidation resistance of UO_2-U_3Si_2 pellet (air)

- **Ramped temperature oxidation**
  - 5 ℃/min, up to 800 ℃, air
- **UO_2-U_3Si_2 composite pellet**
  - Increased onset temperature of breakaway oxidation than U_3Si_2.
- **Al doping also enhanced oxidation resistance.**



Pulverization of U_3Si_2 during oxidation test



Thermogravimetric analysis (TGA) on UO_2, UO_2-U_3Si_2, and UO_2-U_3Si_2-Al pellets

Corrosion resistance of UO_2-U_3Si_2 pellet (pressurized water)

- **Enhanced corrosion resistance of UO_2-U_3Si_2 than U_3Si_2**
- **Corrosion test at 300 ℃ for 24 h under 100 bar H_2O**
  - UO_2: no corrosion occurred
  - U_3Si_2: U_4O_9 formation and dissolution
  - UO_2-50wt%U_3Si_2: no corrosion occurred

Reaction vessel

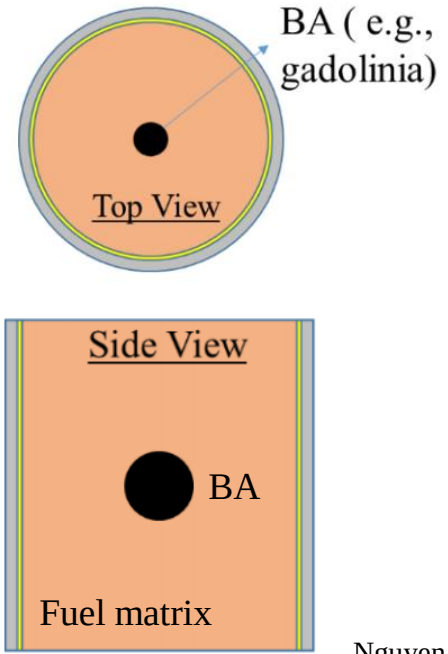
UO_2

U_3Si_2

UO_2-50wt%U_3Si_2

Central Shielded Burnable Absorber (CSBA)

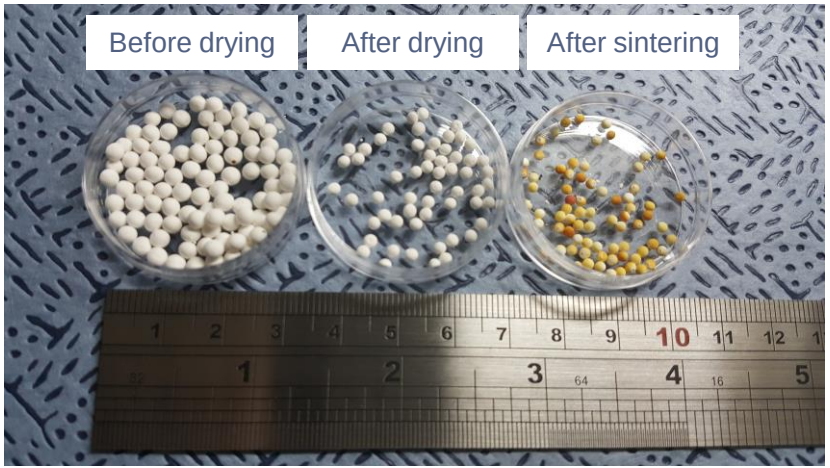
- **Conceptually innovative BA design introduced to eliminate the soluble boron.**
- **Gd_2O_3 ball fabrication: Drip casting → Drying → Sintering (~89%TD ball)**



Nguyen et al.

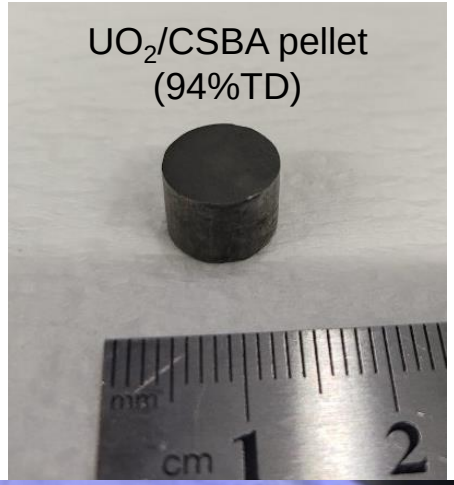
Design of CSBA fuel

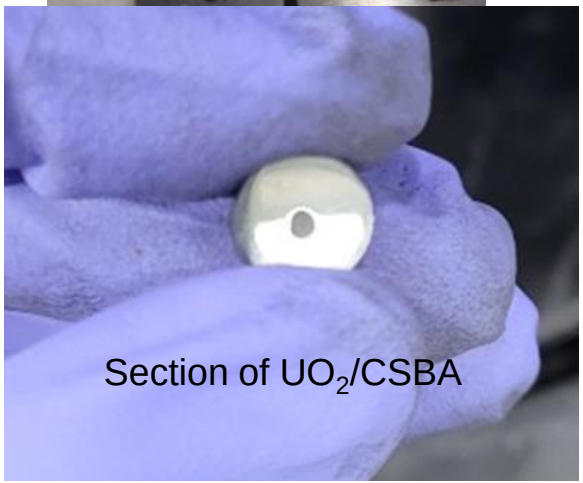
Gd_2O_3 ball drip casting

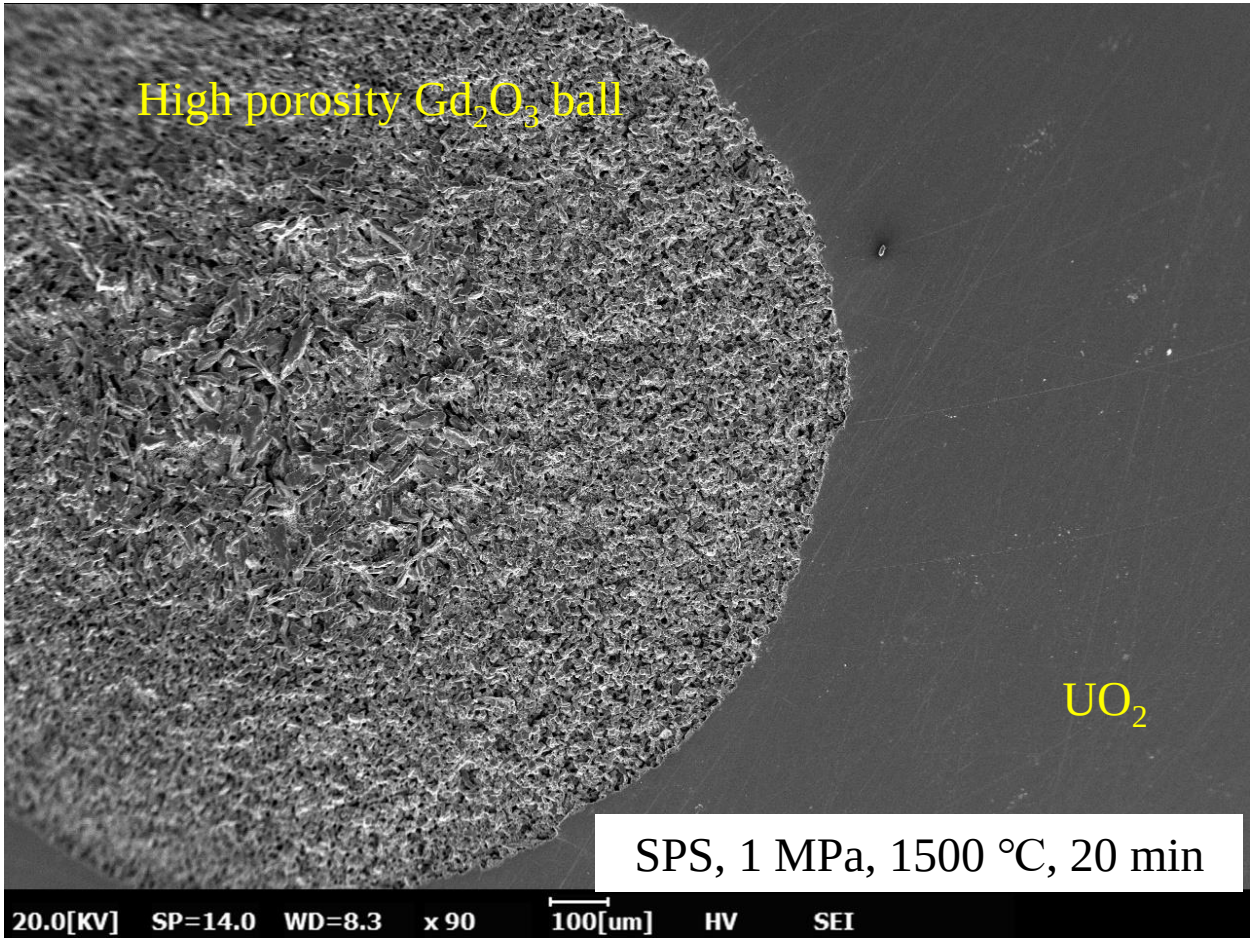


Gd_2O_3 ball sintering

UO_2/CSBA pellet fabrication (using SPS)

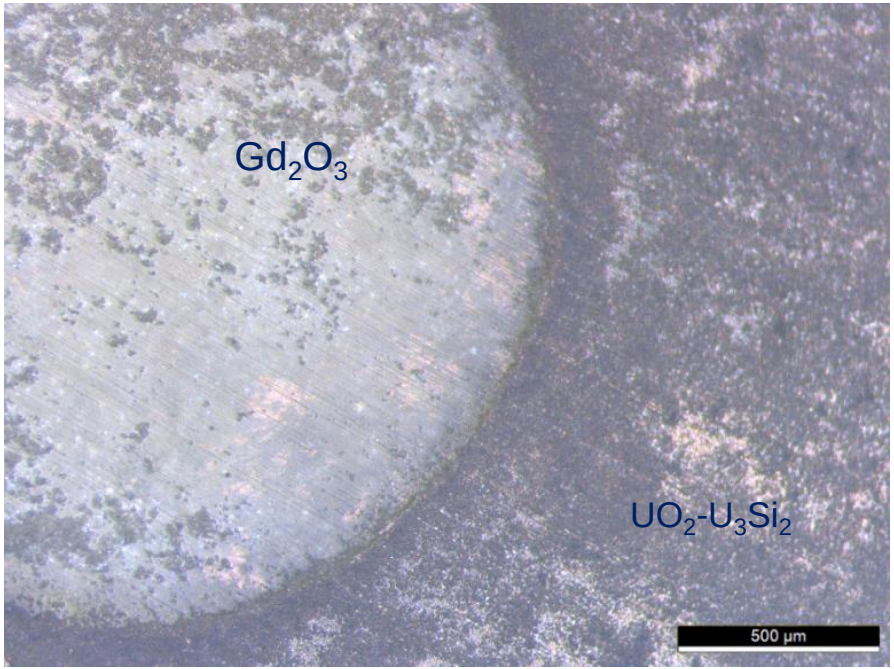


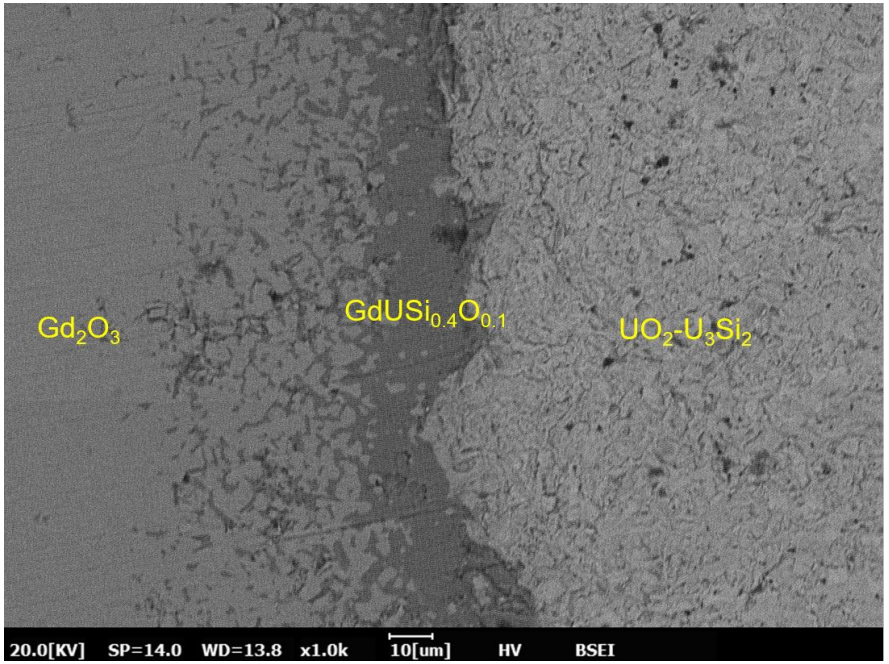




UO_2-U_3Si_2/CSBA pellet thermal stress resistance

- **Load following operation → thermal cycle on fuel → thermal stress**
- **Thermal cycle test on UO_2-U_3Si_2/CSBA pellet**
  - 600~800 ℃, 10 cycle, 200 min/cycle
  - Interaction layer observed but no crack formed for both Gd_2O_3 ball and UO_2-U_3Si_2.





Summary

Summary

Background

- **Lower fuel temperature to soluble boron free operation & acceptable corrosion resistance**
  → High *k* UO_2-based composite fuel concept
- **Uranium silicide (U_3Si_2):** high thermal conductivity & fissile density, acceptable corrosion resistance

Composite pellet fabrication

- **Spark Plasma Sintering** → 91~93%TD UO_2-U_3Si_2 pellet fabrication
- Secondary phase (UO) formation by exothermic reaction at 1050~1200 ℃
- UO_2 barrier formation along with U_3Si_2 grain boundary

Thermal conductivity measurement

- **Increasing thermal conductivity with increasing U_3Si_2 composition**
  - 10wt%U_3Si_2 pellet showed similar thermal conductivity with UO_2.
  - 50wt%U_3Si_2 : 90% thermal conductivity increase at 1000 ℃ w.r.t. UO_2
- **Increasing thermal conductivity with increasing sintering temperature**
  - Sintering @1000 ℃ pellet: very low *k* due to low sintering density
  - Too high sintering temperature accelerate 2nd phase formation which has low-thermal conductivity.

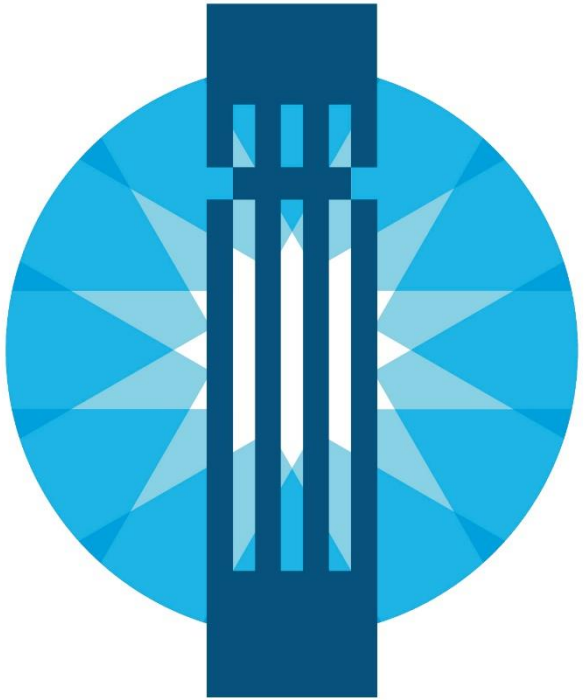
Oxidation resistance

- **Enhanced oxidation resistance of UO_2-U_3Si_2 composite pellet (air oxidation)**
  - Increased onset temperature of UO_2-50wt%U_3Si_2 pellet (450 ℃) compared to U_3Si_2 (300 ℃).
  - **Enhanced corrosion resistance of UO_2-U_3Si_2 composite pellet (pressurized water)**
  - UO_2-50wt%U_3Si_2 pellet showed no dissolution in pressurized water.

UO_2-U_3Si_2/ CSBA

- **No crack formation, but 2nd phase layer formation btw. UO_2-U_3Si_2 and Gd_2O_3 boundary.**

Q & A



Thank you for your attention