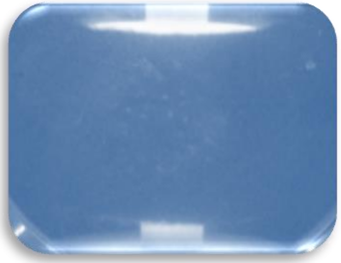
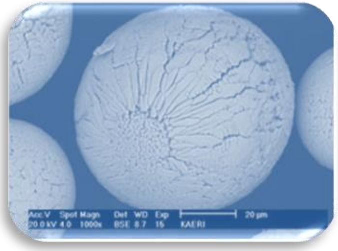
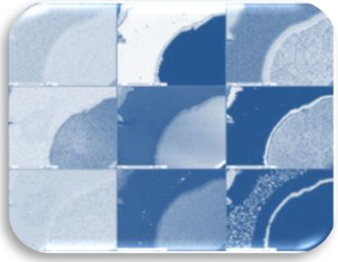
KNS Spring Meeting
OCT 20, 2022







Efficient cold immobilization of Bi^0–rGO iodine waste

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Outline

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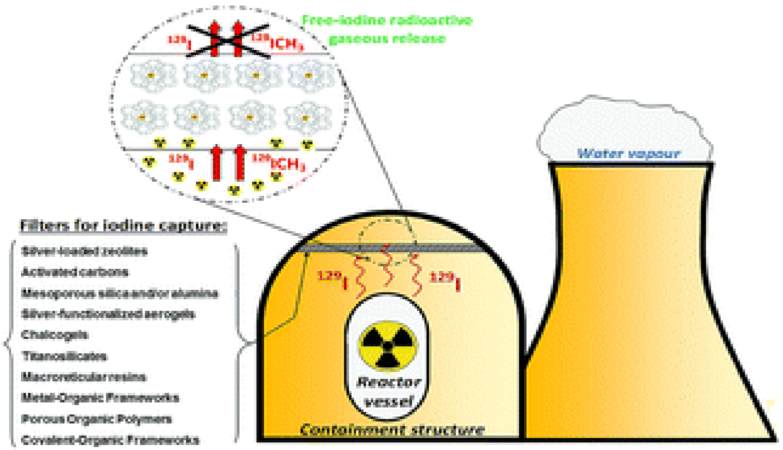
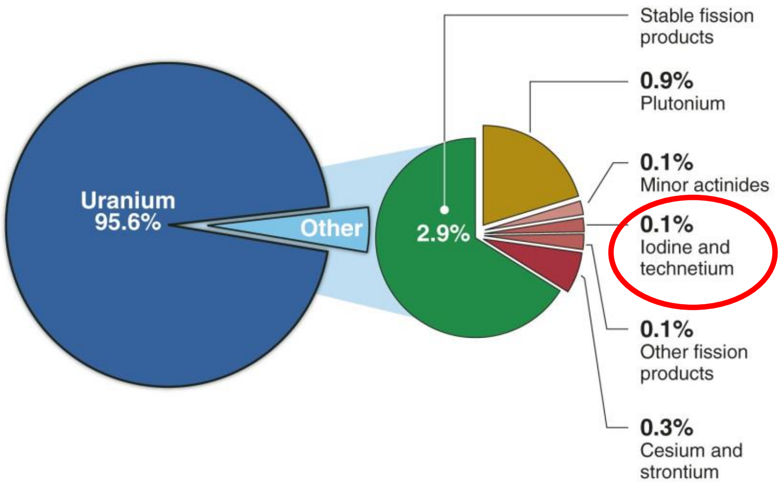
Background



- Composition of Spent Nuclear Fuel



Source: GAO analysis of DOE data.

Problems related with I-129

- Most troublesome radioisotopes
- Radioactive and acutely toxic
- long-lived fission products: about 16 million years
- Negative health effects (thyroid diseases)
- High environmental mobility

  Iodide(I^-)predominates in aqueous solutions

	Half-life	Fission yield
I-127	stable	0.115
I-129	1.57×10^7y	0.706
I-131	8.02 d	2.878
I-133	20.8 h	6.59
I-135	6.57 h	6.39

J.Huve. et al, RSC Adv. 8 (2018) 29248

❖ Capture and storage of iodine is of great importance

Background

Technique	Advantages	Disadvantages	Material examples
Hot pressing	High densities Limited grain growth	Batch process	$Pb(Zr,Ti)O_3$ (Ba,Sr)TiO3 Diamond
Microwave sintering	Lower temperatures Faster process Multimode or single mode High densities	Limited success in mass production Inhomogeneity in coupling Limited yields for large parts	Al_2O_3 BN $BaTiO_3$ WC Metals
Spark plasma sintering	Wide range of materials Enhanced densification	Batch process Upscaling High cost	Metals $BaTiO_3$ ZrO_2 SiC AlN
Flash sintering	Densified, very fast rates Limited grain growth	Microcracking in thermal shocking	ZrO_2 $SrTiO_3$
Hydrothermal sintering	Low temperatures Environmentally friendly Meta stable materials can be sintered	Long processing times Lower densities in some cases	TiO_2 SiO_2 (amorphous and quartz) ZrO_2 $BaTiO_3$

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J. Guo et al., Annu. Rev. Mater. Res. 49 (2019) 275-295

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Objective

In conventional sintering:

- Processing temperature > 900℃
- Sintering time > 60 min
- Volatilization of radionuclides

To explore ways to transform materials to waste forms by encapsulating them at low temperatures into highly durable waste forms

Low temperature sintering and easy processing techniques need to be developed for long term disposal of I-129

↓

to minimize iodine loss

Immobilization of I-129 (2)

Primary role of a waste form:

- To immobilize radioactive and hazardous constituents in a stable & solid matrix

Criteria:

- Low cost fabrication
- Ease of fabrication
- High waste loading capacity
- Durability of waste form

Vitrification: Standard immobilization method

- low solubility of iodine in silicate melts
- high volatility at standard vitrification process temperatures
- instability in the alkaline environment of deep geological layers

Studies on bonding materials: Cement and Glasses

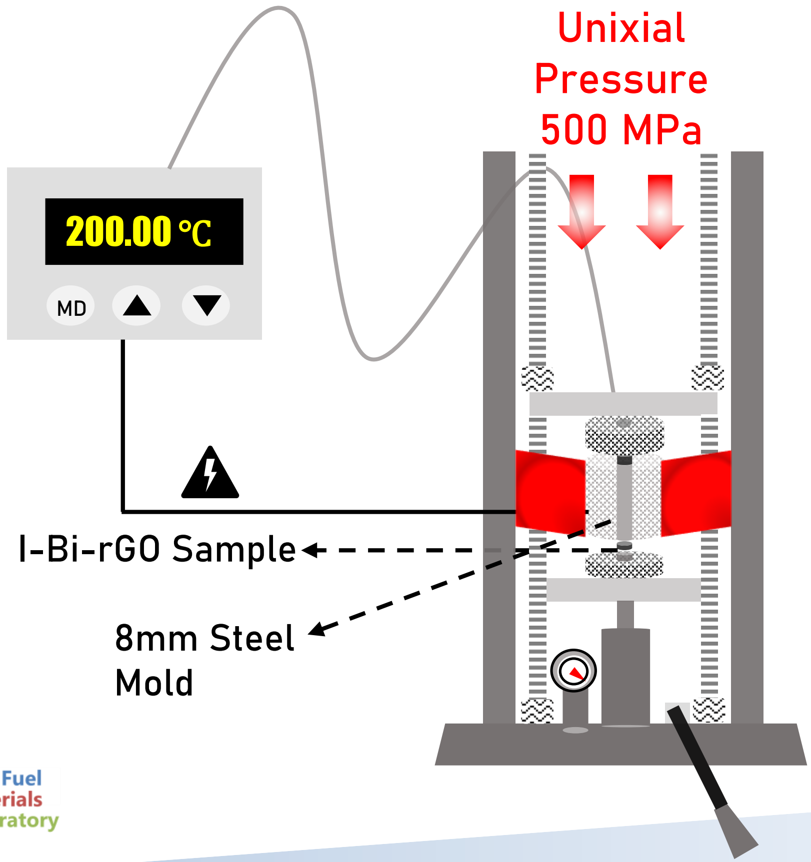
- Low waste loading capacities
- Long processing times
- High energy consumption
- High processing temperature

NOT EFFECTIVE

Immobilization of I-129 (3)

Cold Sintering

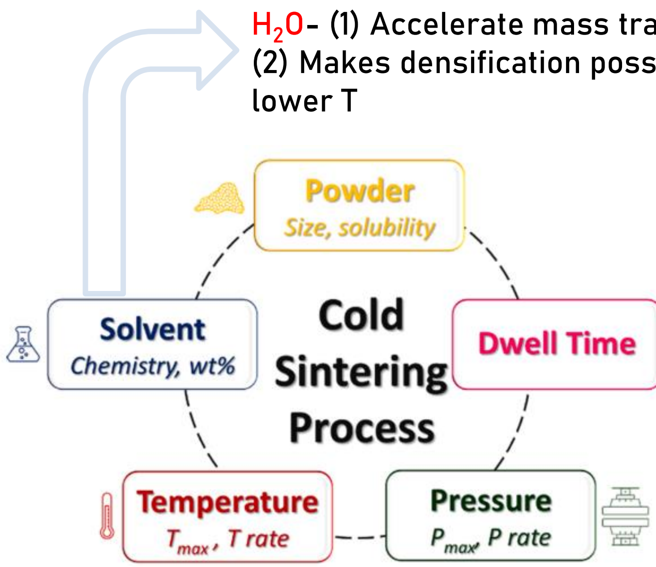
Extremely low-temperature <300℃ process that requires two phases:
(a) a parent powder from which to form a ceramic body
(b) a transport phase to facilitate mass transfer to and from the original particles



Parameters affecting the cold sintering process

Bi_2O_3- (1) added to create more oxygen vacancy
(2) applied to absorb iodide ➡ BIOI

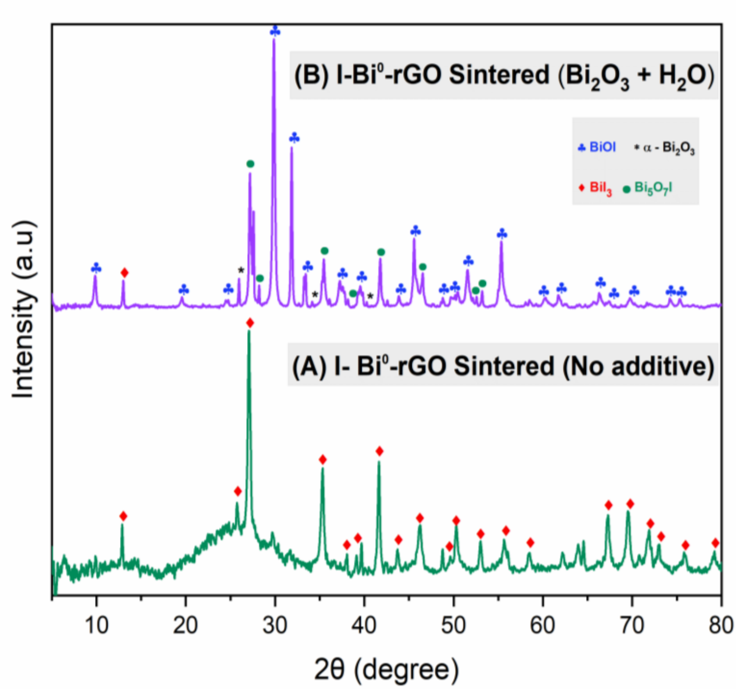
H_2O- (1) Accelerate mass transport
(2) Makes densification possible at lower T



A. Galotta et al., J. Eur. Ceram. Soc. 41 (2021) 1–17

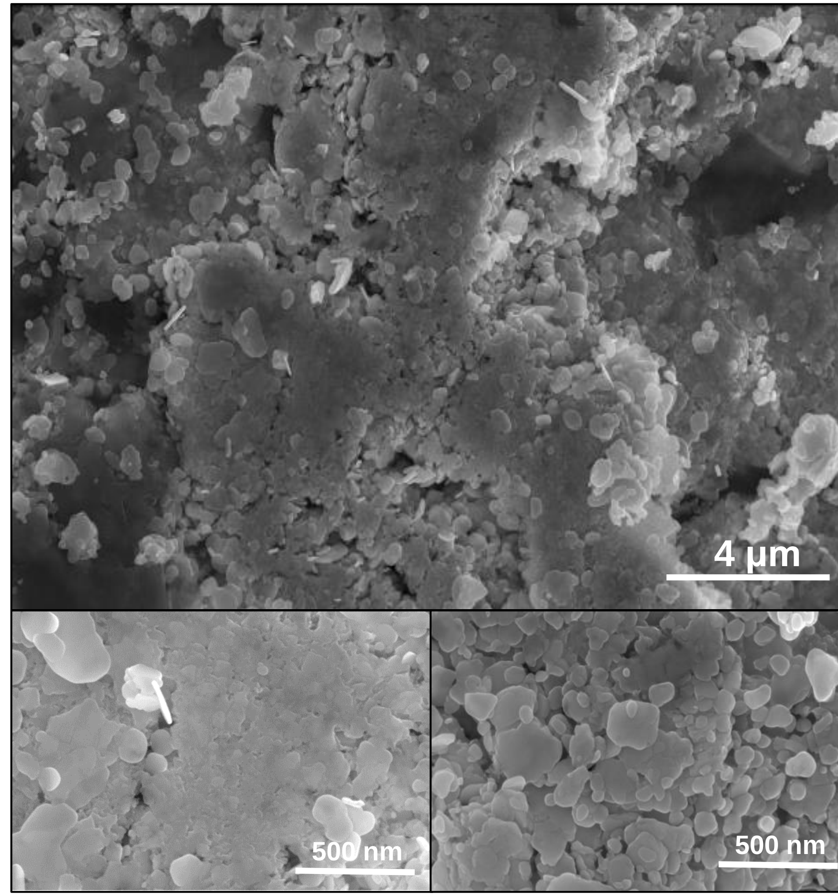
Results(1)

PXRD patterns



Sample	True density of unadsorbed Bi^0–rGO powder (g/cm^3)	Sintered relative Density (%)	Micro-hardness (GPa)
Cold sintered I-Bi-rGO (No additive)	4.25	93.7	0.55 ± 0.1
Cold sintered I-Bi-rGO (Bi_2O_3 + H_2O)	4.25	98.8	0.64 ± 0.1

SEM of fractured surface



removal of porosity by full densification

- a dry-sintered pellet is less dense than a pellet sintered with the addition of a liquid phase under the same conditions (pressure, temperature and dwell time)

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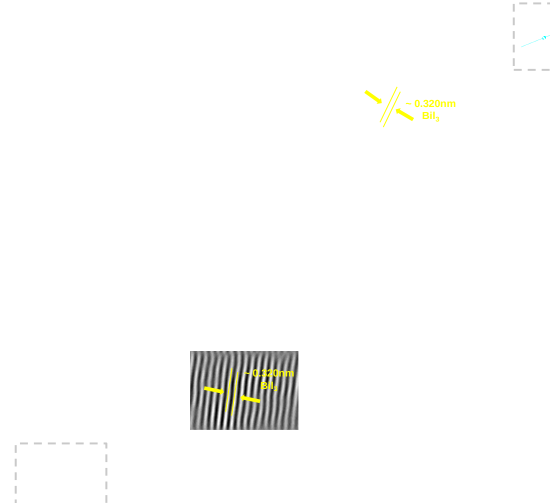
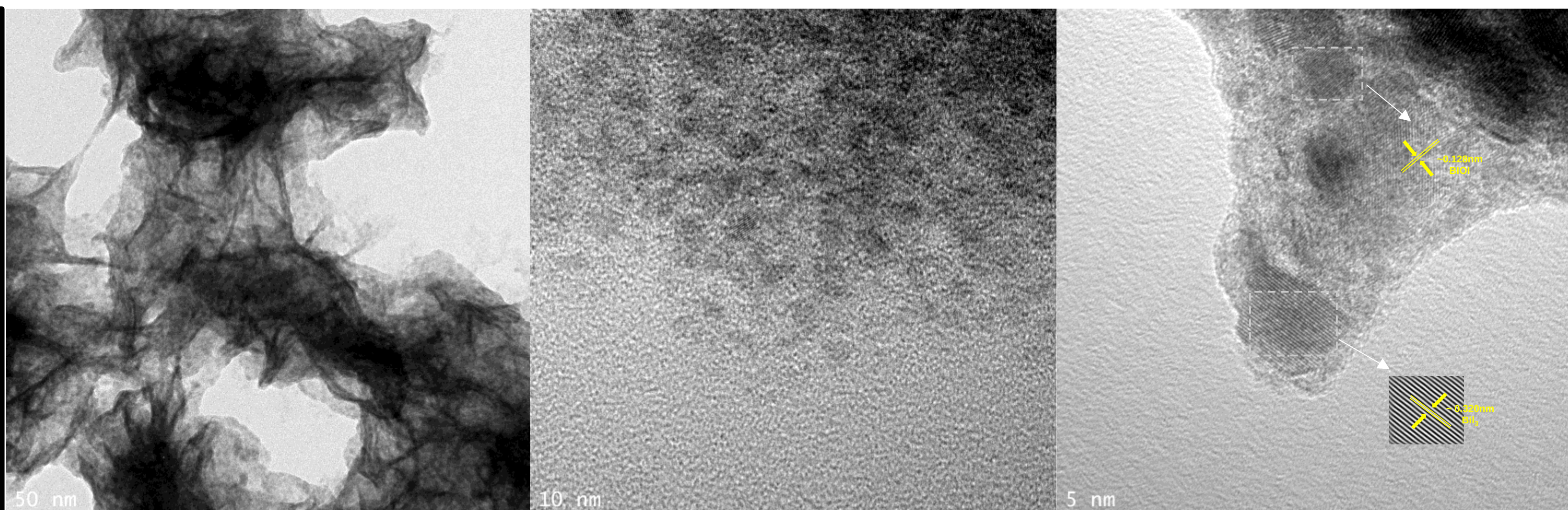
Results(2)

TEM of I-Bi-rGO sample



TEM of sintered sample



Results(3)

Chemical durability test → Stability of sintered samples

Protocol = ASTM-1285 standard
Sample = 1g crushed & sieved powder (150 µm)
Leachant = ASTM Type-I water
Temperature = 90 ± 1℃
Duration = 7 days

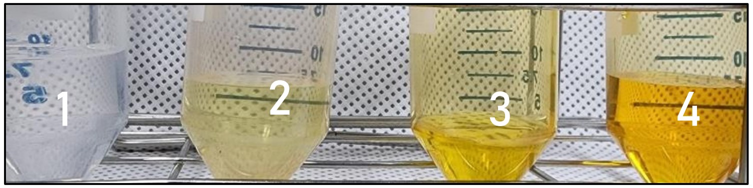
$$NLR_i = \frac{C_i}{t \times f_i \times S/V}$$

NLR_i = Normalized leaching rate i[th] element

C_i = Concentration of component I (g/L) leached into the water after a reaction time of t

f_i = weight fraction of the i[th] element in the starting sample

S/V (m^2 /L) = Specific Surface Area of the powdered waste form (1g)



1. Bi^0
2. BiI_3 before sintering
3. BiI_3 after sintering
4. BIOI after sintering

Summary

- Bi^0–rGO iodine wastes were immobilized with low temperature sintering
- 200 ℃ under a uniaxial pressure of 500 MPa for 20 min with the addition of Bi_2O_3 & 20 wt.% deionized water
- Effect of the water content and Bi_2O_3 : shifting of BiI_3 to mostly BIOI
- Based on these prior data, cold sintering could be an option for immobilization of I-129

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