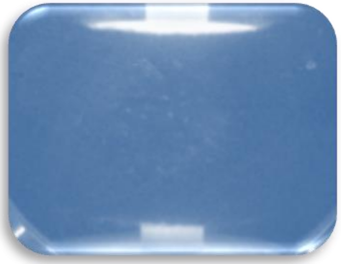
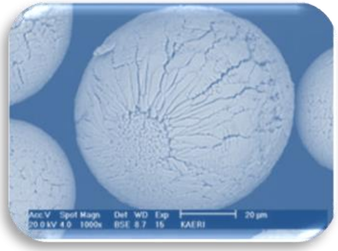
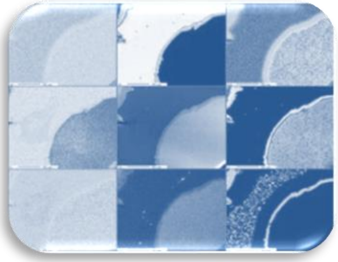
KNS Spring Meeting
May 18, 2023







Low Temperature Sintering for the Immobilization of Bi^0-rGO Iodine Wastes

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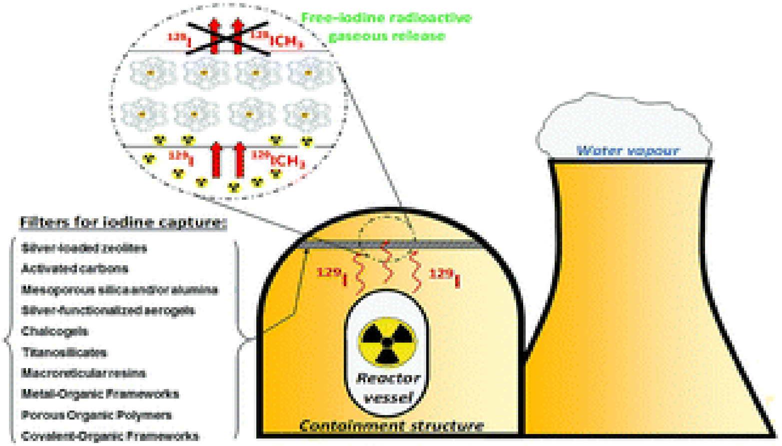
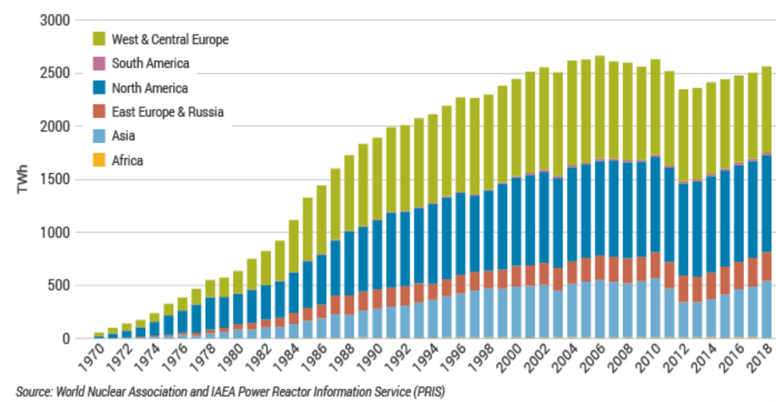
Outline

Growing need for safer energy

한국원자력학회 KOREAN NUCLEAR SOCIETY

Nuclear energy offers several advantages such as cleanliness, safety, & lower operating costs





Nuclear Electricity Production (1970-2018)

J.Huve. et al, RSC Adv. 8 (2018) 29248
https://world-nuclear.org/information-library/current-and-future-generation/nuclear-power-in-the-world-today.aspx

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Safe management of nuclear waste

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- Radioactive & acutely toxic
- Half-life: **~16 million years**
- Negative effects on the human health
- High environmental mobility when dissolved in groundwater

53
^{129}I
iodine

Capture & storage of off-gas iodine are essential !!!

❑ Solid sorbents

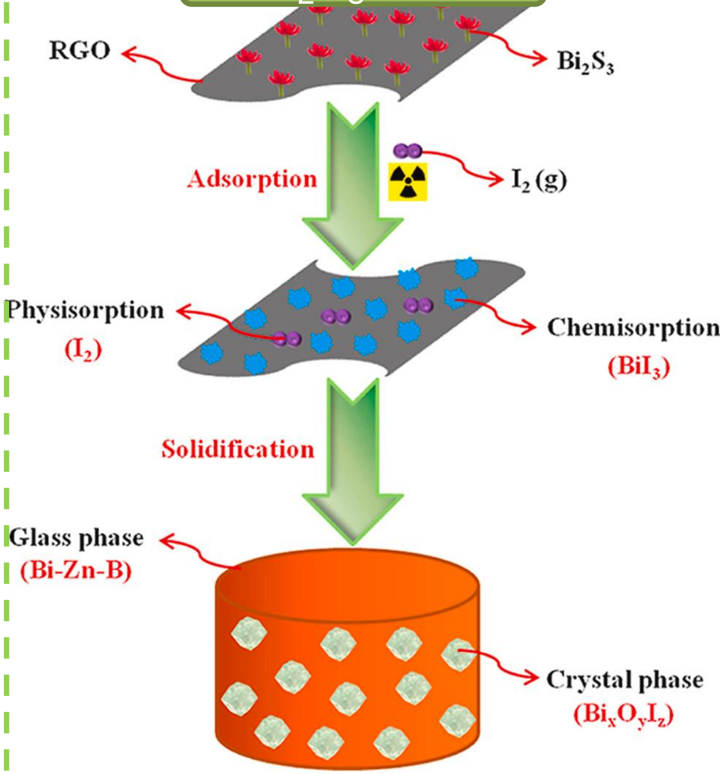
	Matrix	Advantages	Disadvantages
Inorganic	**Activated Carbon**	▪ Low cost	▪ Alteration of sorption properties under the influence of aging
Inorganic	**Silver-based materials**	▪ Good trapping performance	▪ High cost
Porous	**Metal organic frameworks (MOFs)**	▪ High I_2 sorption capacity	▪ Possibility of degradation at high temperatures

A. T. Reda et al., J. Environ. Chem. Eng, 9 (2021) 105279
K.W. Chapman. et al, J. Am. Chem. Soc. 132 (2010) 8897–8899
D.F. Sava. et al, J. Am. Chem. Soc. 133 (2011) 12398–1240

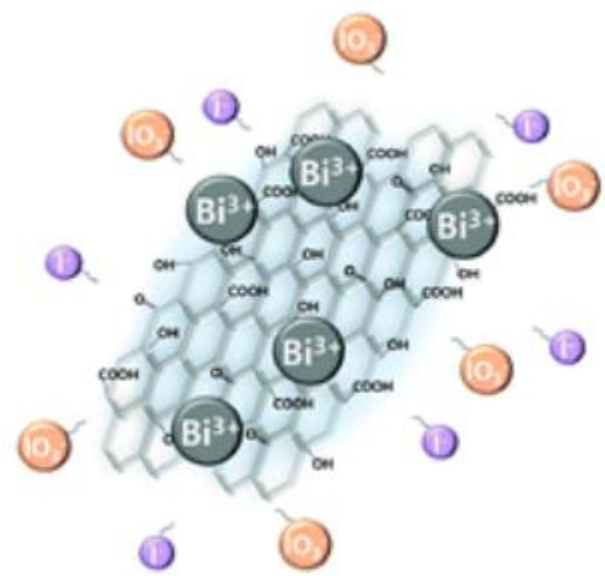
Bismuth-based materials

Bismuth-based absorbents have attracted considerable interest because of their unique properties & low costs

Bi_2S_3-rGO



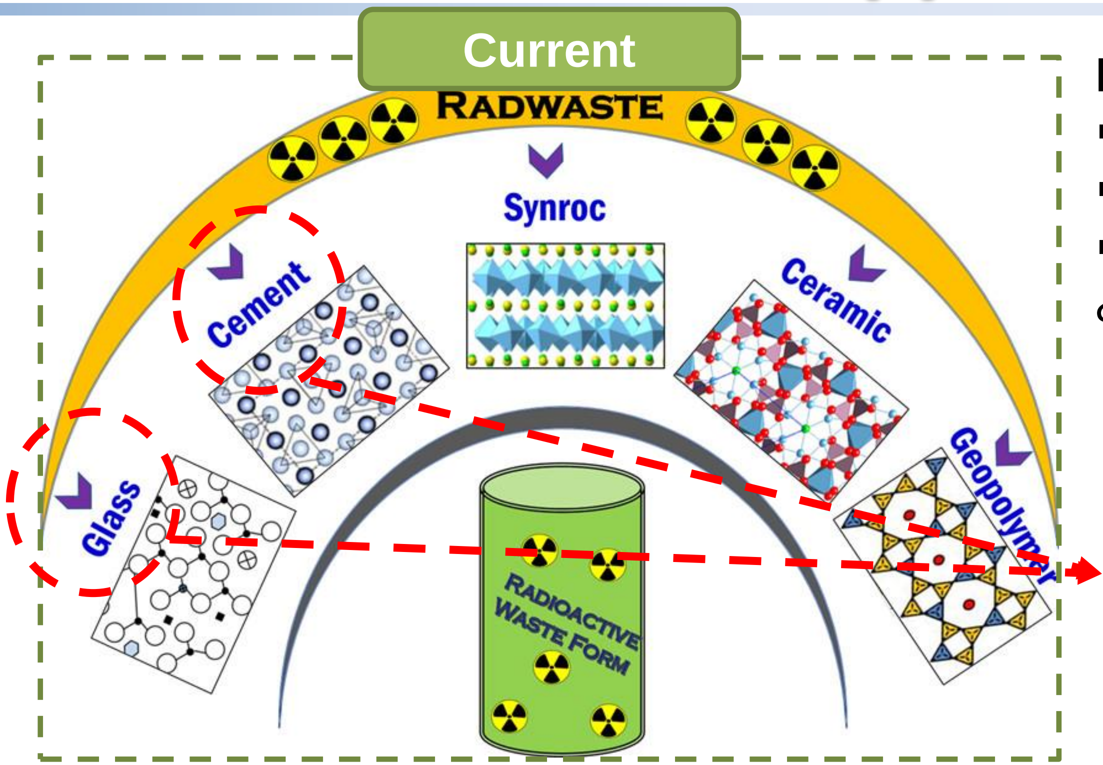
Bi–GO



No leaching resistance of the solidified waste forms was examined

Prog. Nucl. Energy. 135 (2021) 103705
Dalt. Trans. 48 (2019) 478–485

Immobilization of I-129 (1)



Designing suitable waste forms:

- Durability
- Stability
- Resistivity to various physical & chemical conditions

Issues

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

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

ACS EST Engg. 2021, 1, 8, 1149–1170

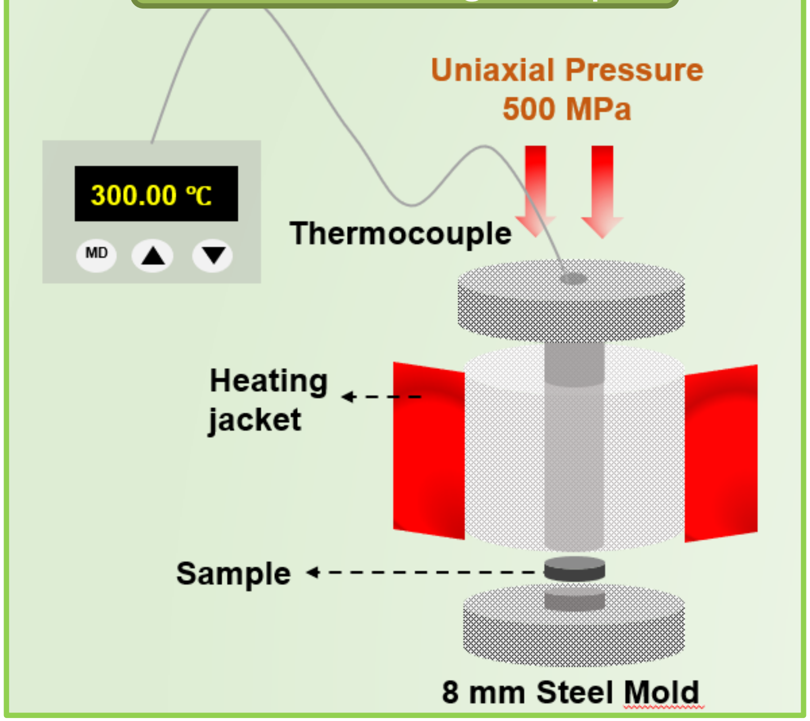
Immobilization of I-129 (2)

❑ Cold sintering Process (CSP):

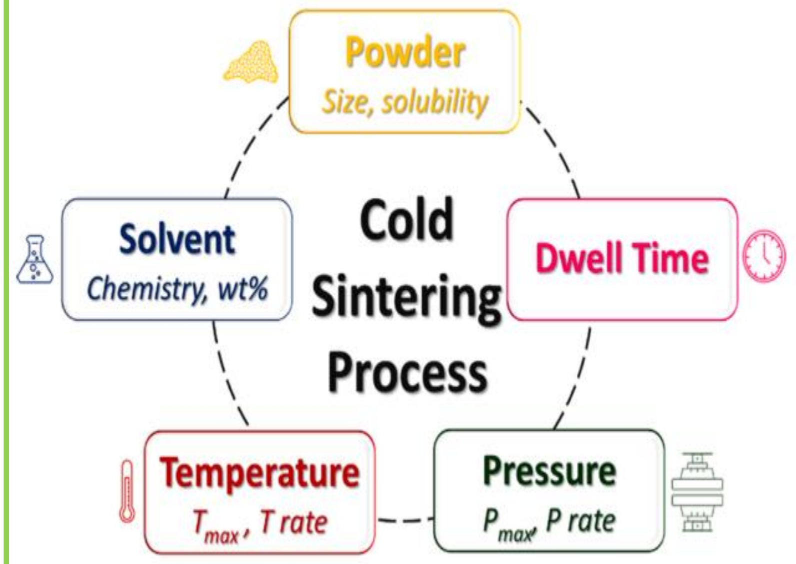
Extremely low-temperature <300 ℃ that requires two phases:

- a parent powder from which to form a ceramic body
- a transport phase to facilitate mass transfer to and from the original particles

Cold sintering setup



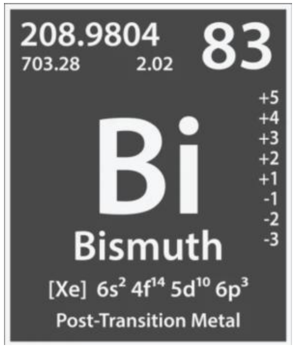
Parameters



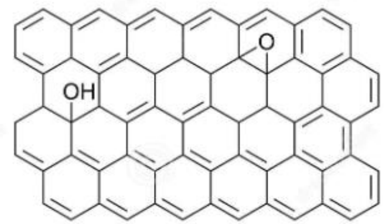
>>> CSP does not require heating waste to high temperatures

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

Objectives



- High sorption capacity
- low production cost
- High affinity to iodine species



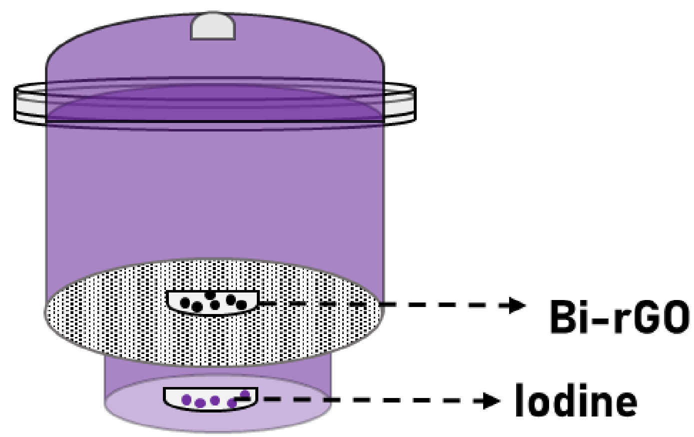
Reduced Graphene Oxide (RGO)

- Hydrophobic
- Low production cost
- Excellent mechanical properties

- Bi^0–rGO sorption tests
- Simple thermal annealing
- Optimize the CSP conditions

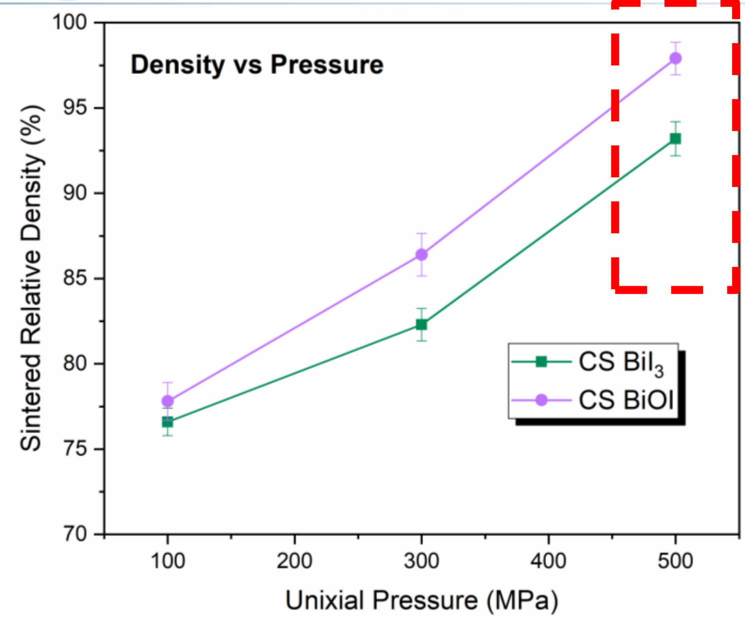
❑ **Characterization**

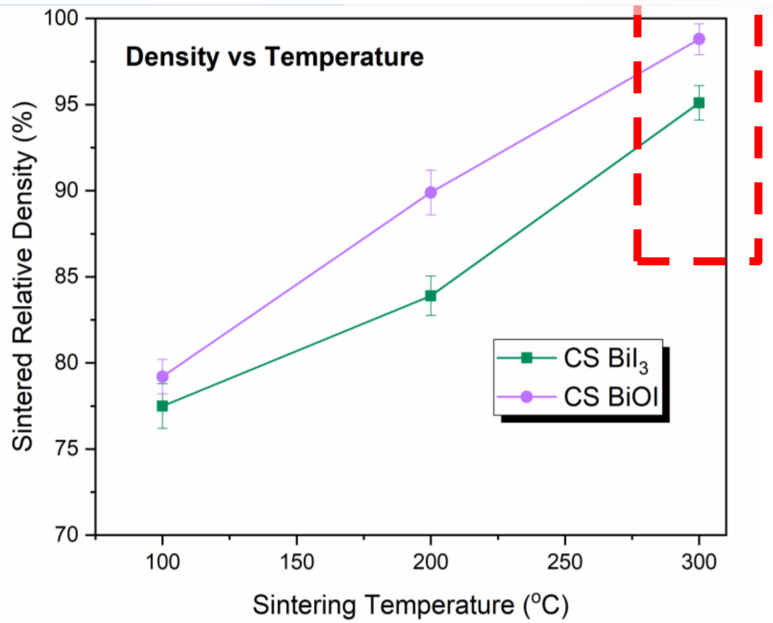
- PXRD for phase analysis
- SEM for microstructural changes
- TG-MS for thermal stability
- PCT Leaching tests for chemical durability



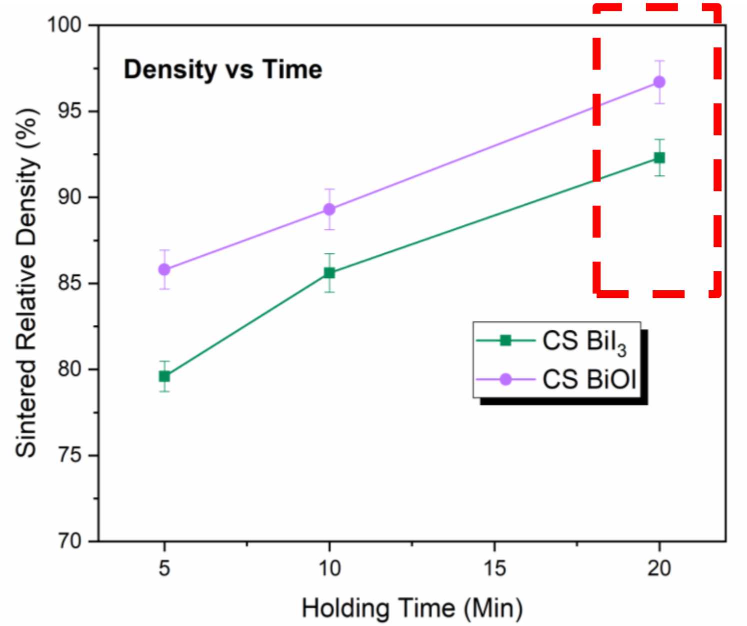
Desiccator

Optimized conditions for CSP

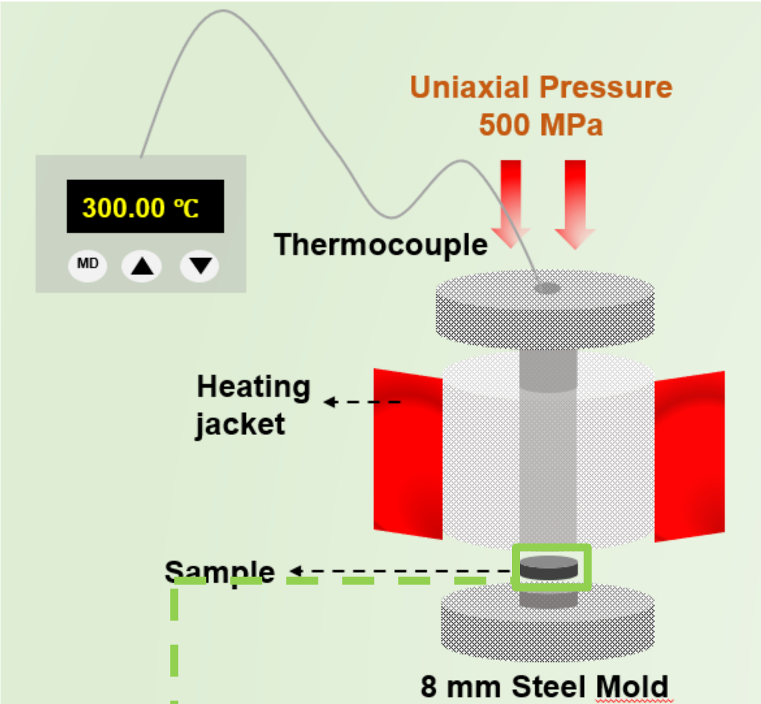


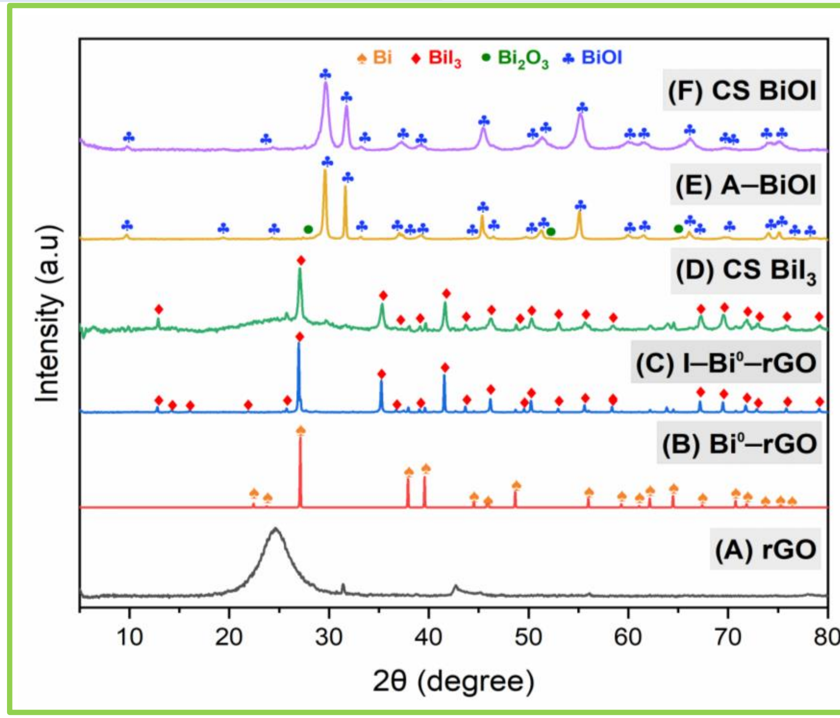


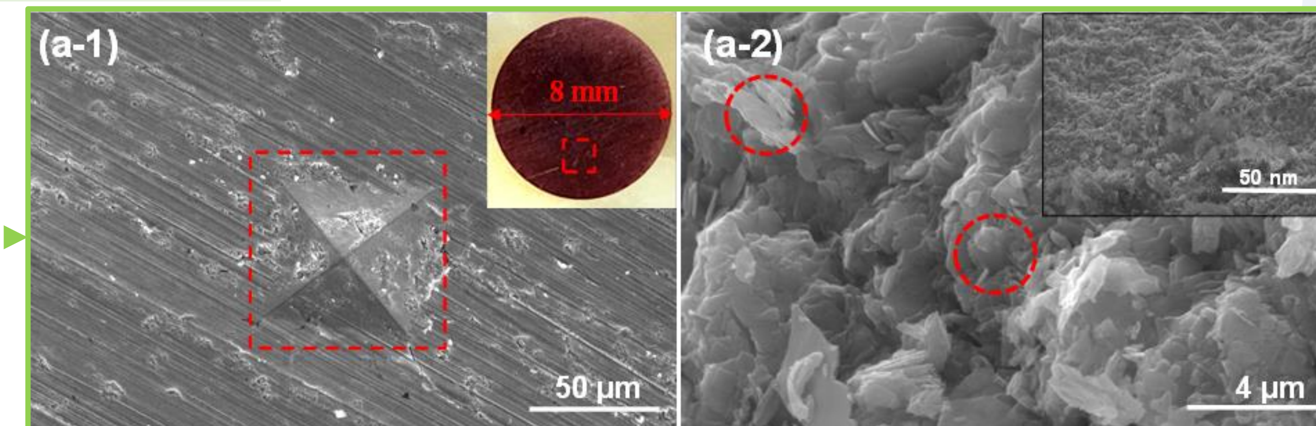
Sample	Sintered relative density (%)	Micro-hardness (GPa)
CS BiI_3	95.1	0.6 ± 0.1
CS BiOI	98.8	1.3 ± 0.1



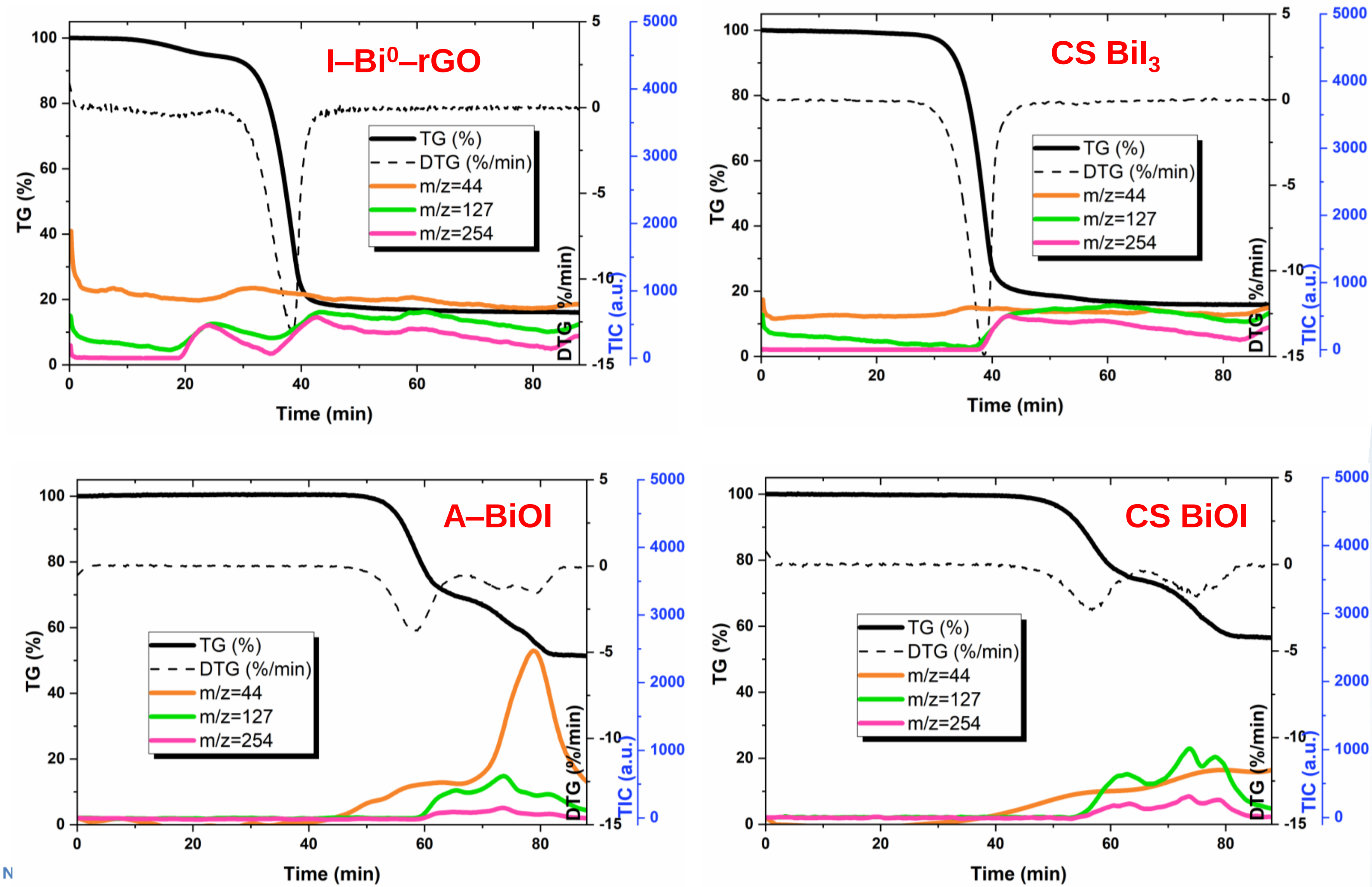
Results – Crystal structure & Microstructure







Results – Thermal stability



Results – Chemical durability

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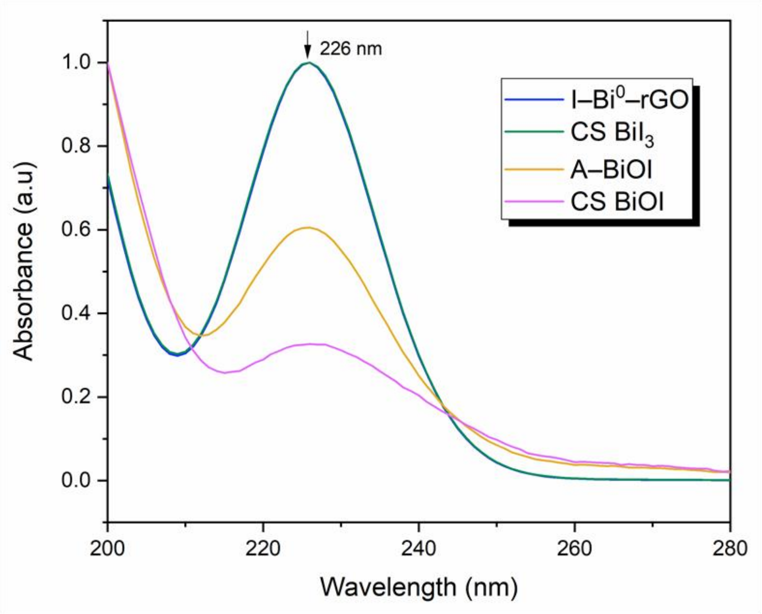
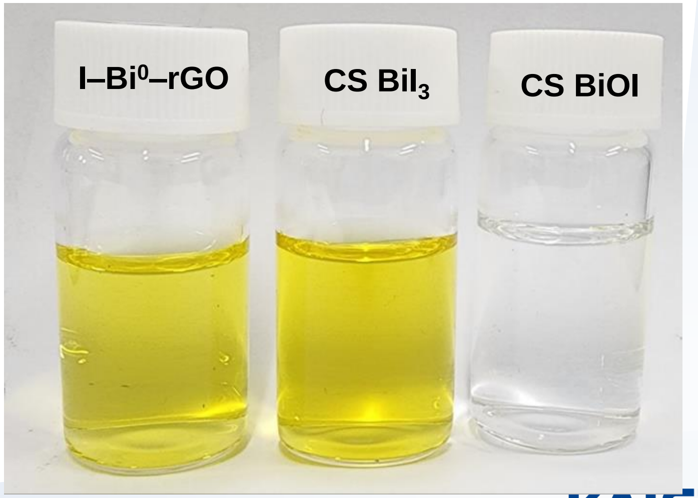
Material	Normalized leaching rate ($g \cdot m^{-2} \cdot d^{-1}$)
$I\text{–}Bi^0\text{–}rGO$	$1.8\ (\pm 0.003) \times 10^{-1}$
CS BiI_3	$2.1\ (\pm 0.005) \times 10^{-1}$
A–BiOI	$9.4\ (\pm 0.0002) \times 10^{-4}$
CS BiOI	$1.9\ (\pm 0.0001) \times 10^{-4}$





- **BiI_3-containing wastes:** cannot be safely disposed
- **BiOI-containing wastes:** formation of the stable iodine-containing BiOI phase via stronger chemical bonding

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Discussion

- High specific surface area is not always required for efficient iodine sorption
- The post-thermal annealing process is a promising approach for iodine stabilization
- The normalized iodine leaching rates of A–BiOI and CS BiOI (~ 10^{-4} g/m^2 day) may originate from the extremely low solubility of BiOI in the composite matrix

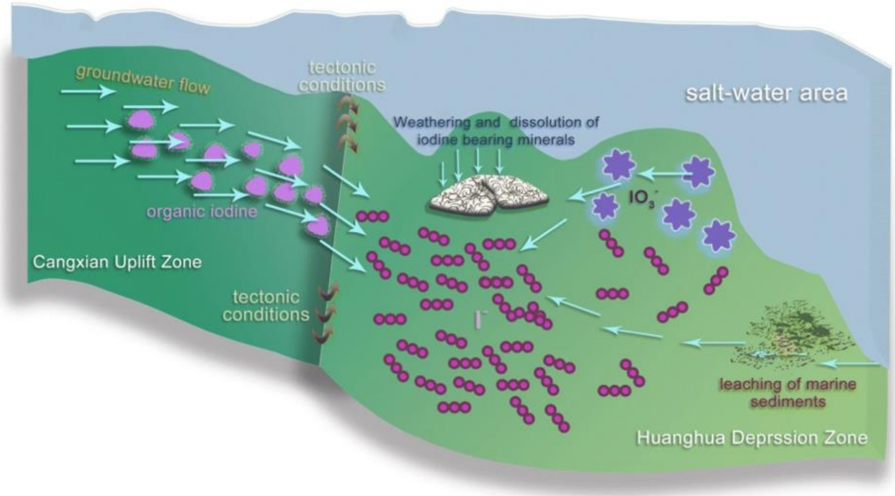
Limitations & future works

❑ Limitations

- Leaching tests simulate only the short-term exposure to leaching solutions with controlled pH values and anion concentrations

❑ Future works

- investigate the effects of complex groundwater chemistry, redox potential, and microbial activity on the iodine properties



Conclusions

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- A novel sorbent (Bi^0–rGO) was fabricated through solvothermal treatment
- High sorption capacity: **1116 ± 49 mg/g**
- A chemically durable iodine phase (BiOI) was obtained by simple thermal annealing
- The CS BiOI achieved a relative density of **~98%** and Vickers hardness of **1.3 ± 0.1 Gpa**
- The normalized iodine leaching rates were reduced by ~**3 orders of magnitude** due to the stable iodine phases (BiOI)

Q & A