Sustainable Waste Form Development: Solidification of Cobalt Captured Zeolite Waste into Geopolymer

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Study Roadmap

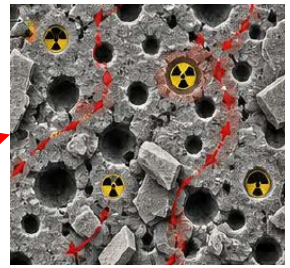
- Why Zeolites?
- How efficient can Zeolites be for decontamination?
- Solidification Co(II)-Zeolite waste in geopolymer
- Effect of Zeolite waste on geopolymer waste form
- Key Outcomes
- What's next?

Key challenges in radioactive waste management

Nuclear power generation produces a range of radioactive waste streams that require safe conditioning and disposal.

Complex wastes (Sludge, resins, salts, soils, organics, etc.)

Cement solidification



Radioactive waste-hosting materials has to be:

- **Versatile and Durable,**
- **High waste loading**
- **Reliable and cost effective**

- **Pore water radiolysis and hydration reaction interreference**
- **higher leaching, low thermal stability,**
- **High carbon emissions**

Need for advanced waste forms!!

*J. S Mcloy et.al, Materials Today, 2024

Geopolymers

What is Geopolymer?

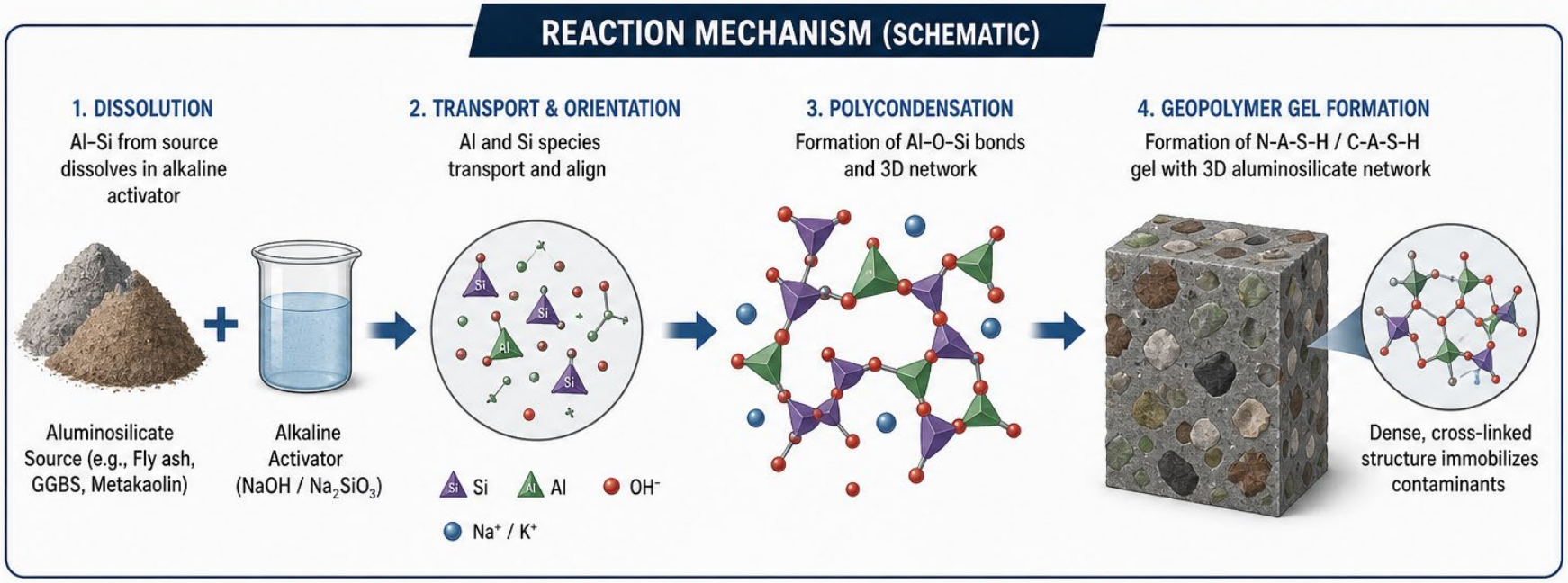
Inorganic polymers that form 3D long-range, covalently bonded, amorphous networks of Si-Al bridging with oxygen atoms.

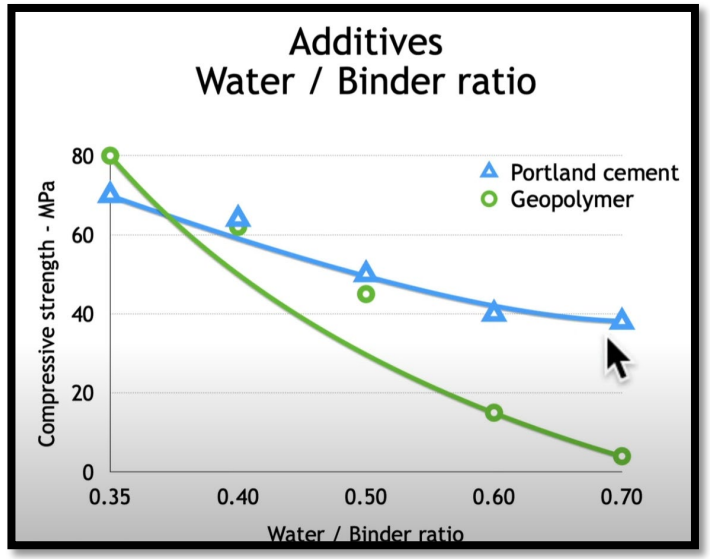
Raw Materials for GP:

Industrial wastes and natural aluminosilicate minerals (**Fly ash**, **Blast furnace slag, Kaolinite/Metakaolin, Red mud**)

Why Geopolymers?

- ❖ Less carbon emitting
- ❖ Low water content, thermally stable
- ❖ Low calcium
- ❖ Can host diverse waste, organic, and oils, etc

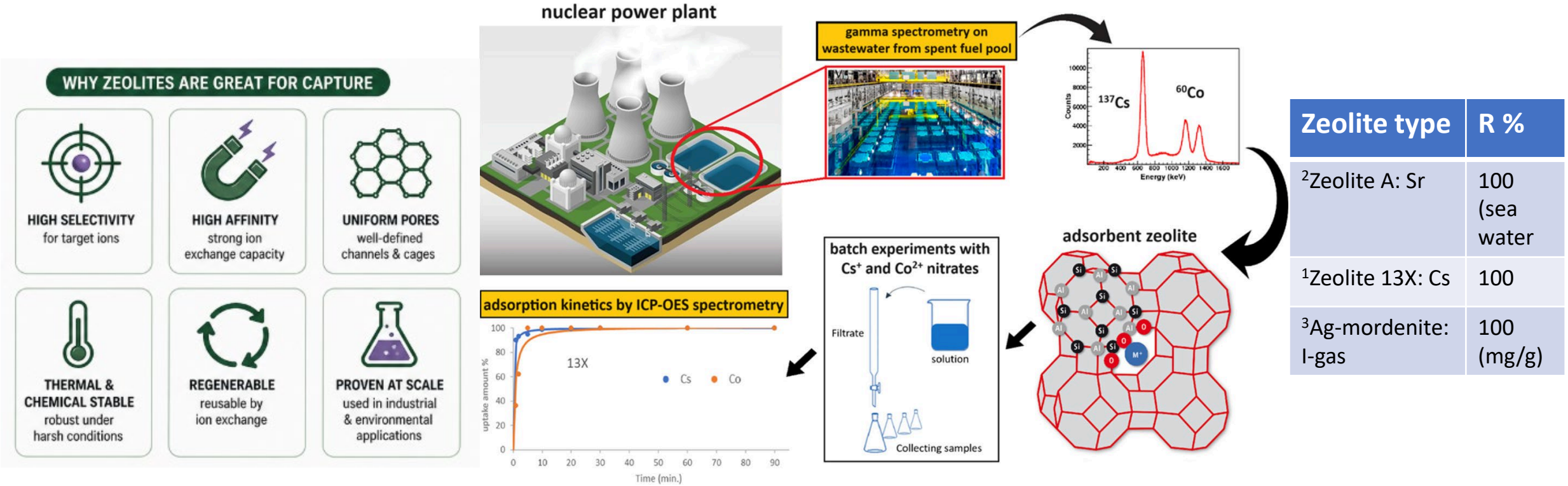




A. Gharzouni et al., Journal of Non-Cryst. Solids 410 (2015) 127–134

Zeolites are Excellent Materials for Decontamination

- Zeolites are **microporous aluminosilicate minerals** with a well-defined crystalline structure.
- They consist of **SiO_4** and **AlO_4** tetrahedra forming a 3D framework that can trap a wide range of anions and cations.



Zeolite type	R %
[2]Zeolite A: Sr	100 (sea water
[1]Zeolite 13X: Cs	100
[3]Ag-mordenite: I-gas	100 (mg/g)

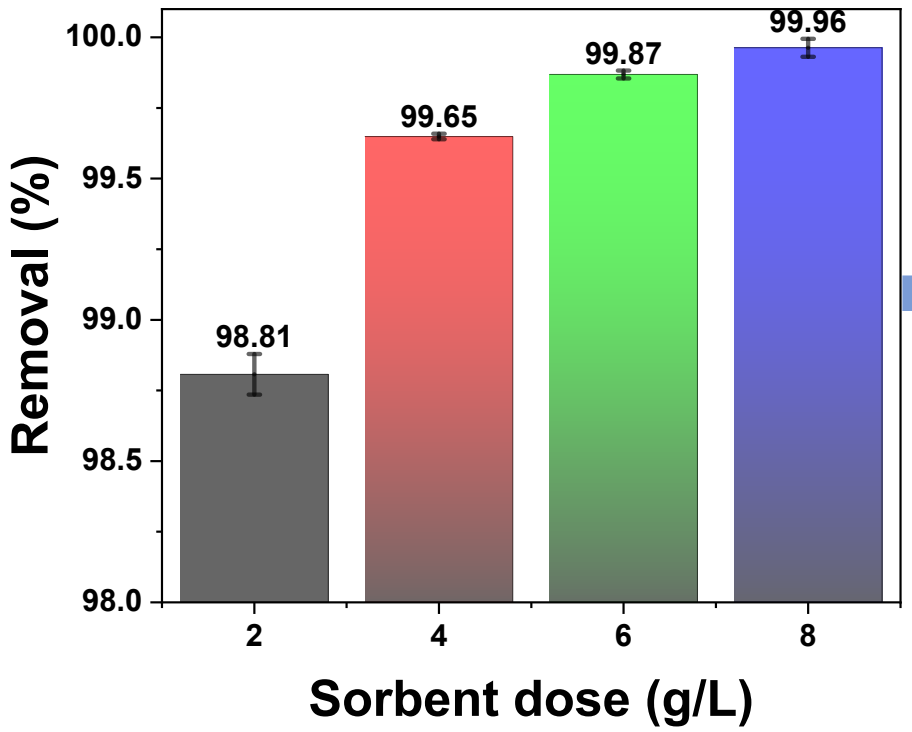
4 types of zeolites were studied: removal rate = ~100% in 20 minutes [1]

Zeolites solve **capture problem**, but not **disposal problem!!**

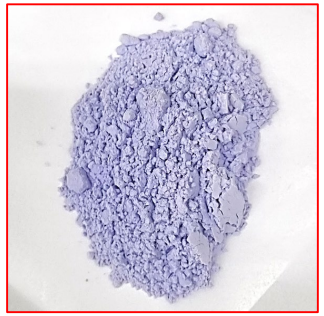
1] F. Ambrosino, et al., Journal of Hazardous Materials 16 (2024) 100458
2] I. Smiiciklas et al Scientific Reports, 11 (2021) 1738
3] A.T greany et al., Frontiers in Chemistry, 8 (2023) 1-7

Cobalt Capture by Zeolite

- ^{60}Co is the most important soluble ionic radionuclide in the PWR primary water system
- ^{60}Co emits high-energy gamma rays (1.17, 1.33 MeV), ($T_{1/2}$: 5.27 Y)
- Responsible for > 70% of the total radiation exposure
- Pose a threat to the maintenance & plant personnel



Co-sorbed Zeolite



~100% removal in 20 min

Solidification of Co(II) Captured-Zeolite by GP

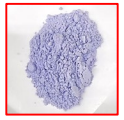
Co(II) spiked de-ionized water

Step 1

Zeolite-Based Ion Exchange Process

Captured Co^{2+} on zeolite

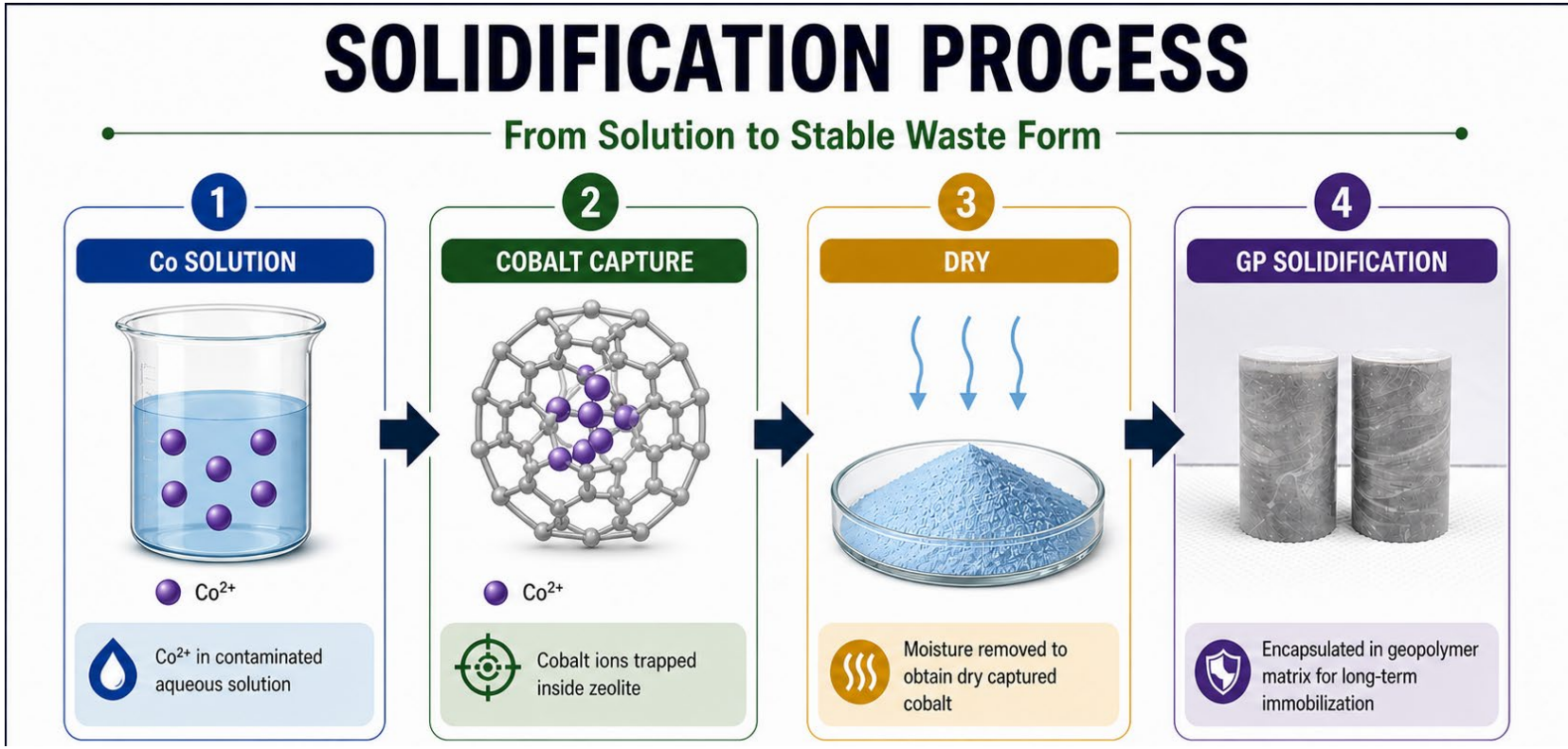
Drying of Radionuclide-Loaded Zeolite (110°C oven)



Step 2

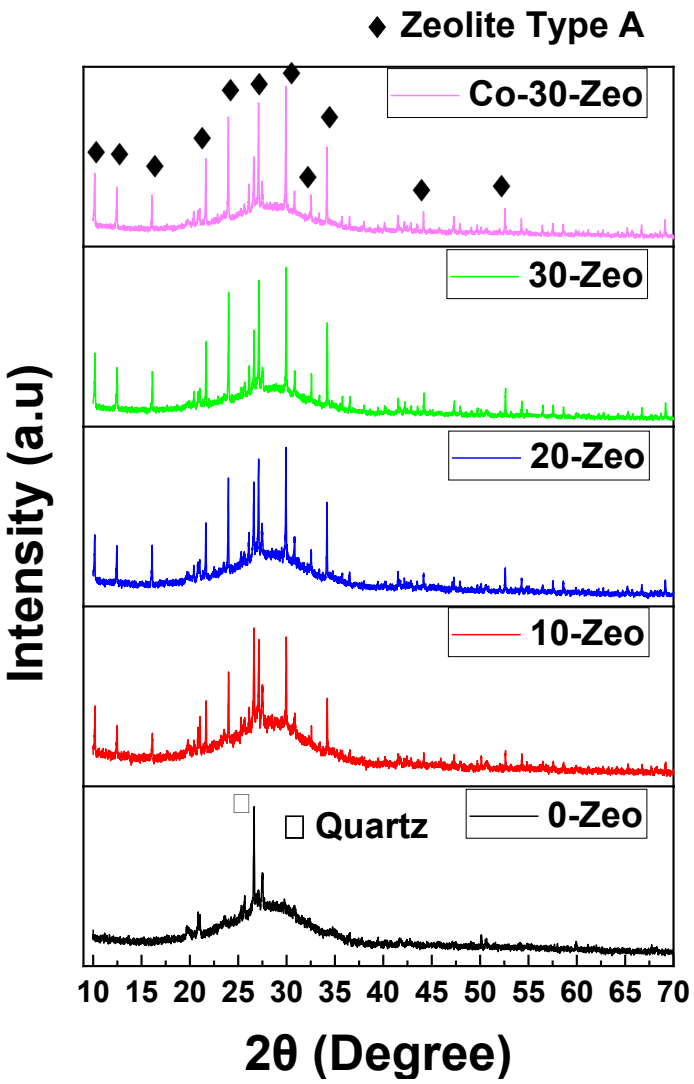
Mixing Dried Zeolite with GP Precursor/Paste

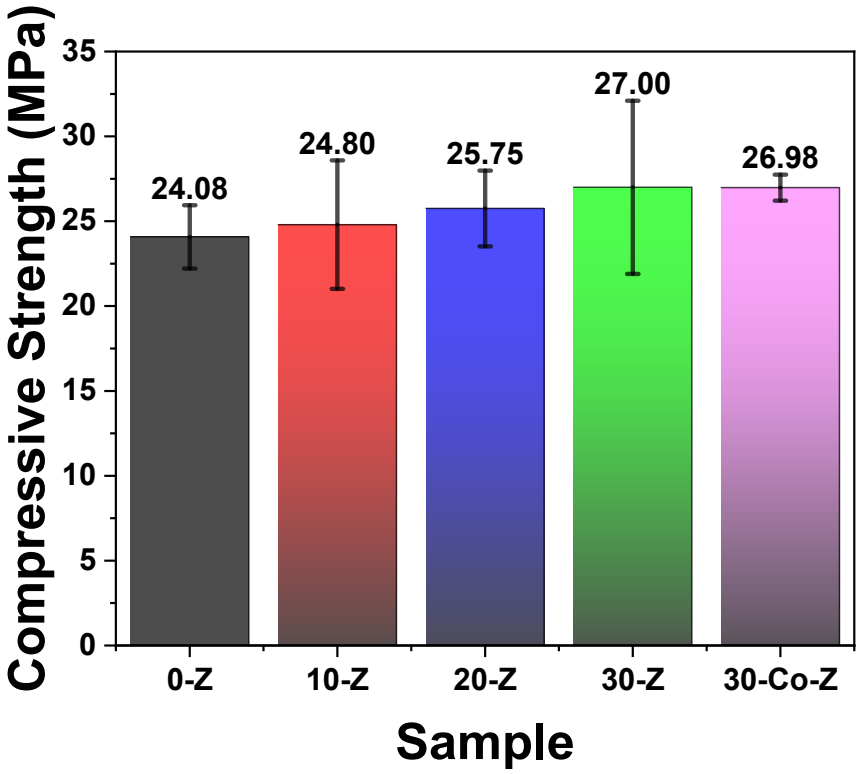
Casting and curing into cylindrical Molds (60 °C)



Chemical and Mechanical Durability Assessment

Zeolite A: (Card No. 96-153-2391)
Quartz: Card No.: 01-078-1252





Why geopolymer improve strength

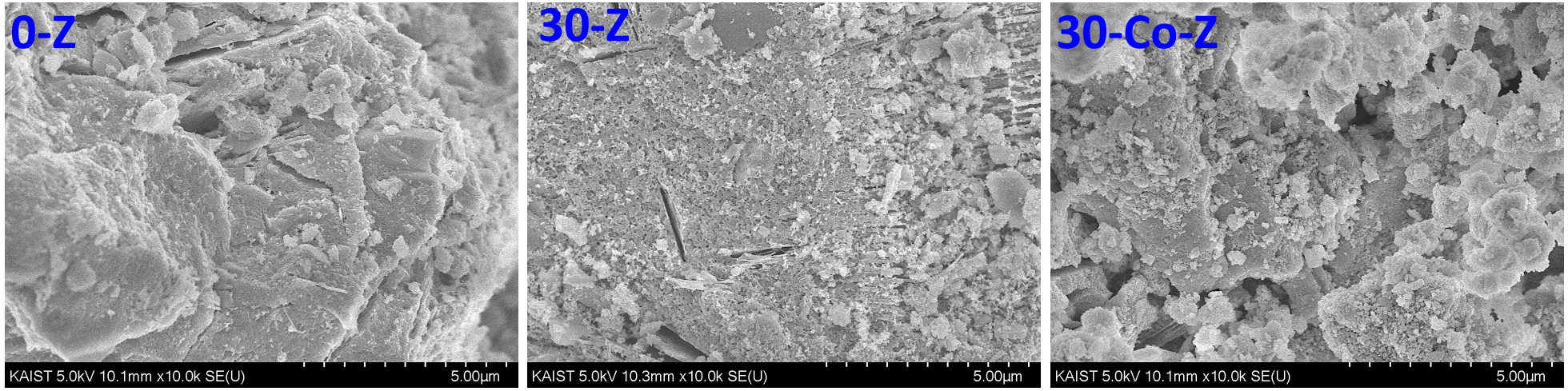
- **Micro-filler Effect**
- **Nucleation effect**

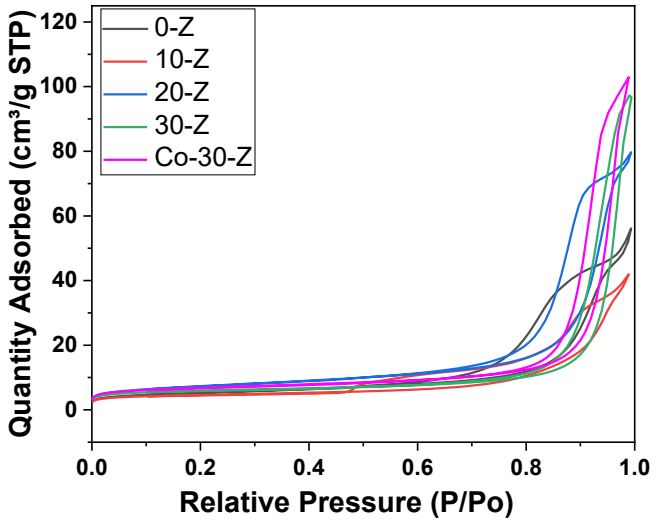
(partial dissolution of Zeolite-A (pH>11) >> extra $Si(OH)_4$, $Al(OH)_4^-$) → more geopolymer gel.

Zeolite not only acts as a cobalt sorbent but also slightly enhances the geopolymer matrix strength

V . GRÄMLICH et al, The crystal structure of hydrated NaA: A detailed refinement of a pseudosymmetric zeolite structure, Zeitschrift für Kristallographie-Crystalline Materials, 133, (1971) 134-149

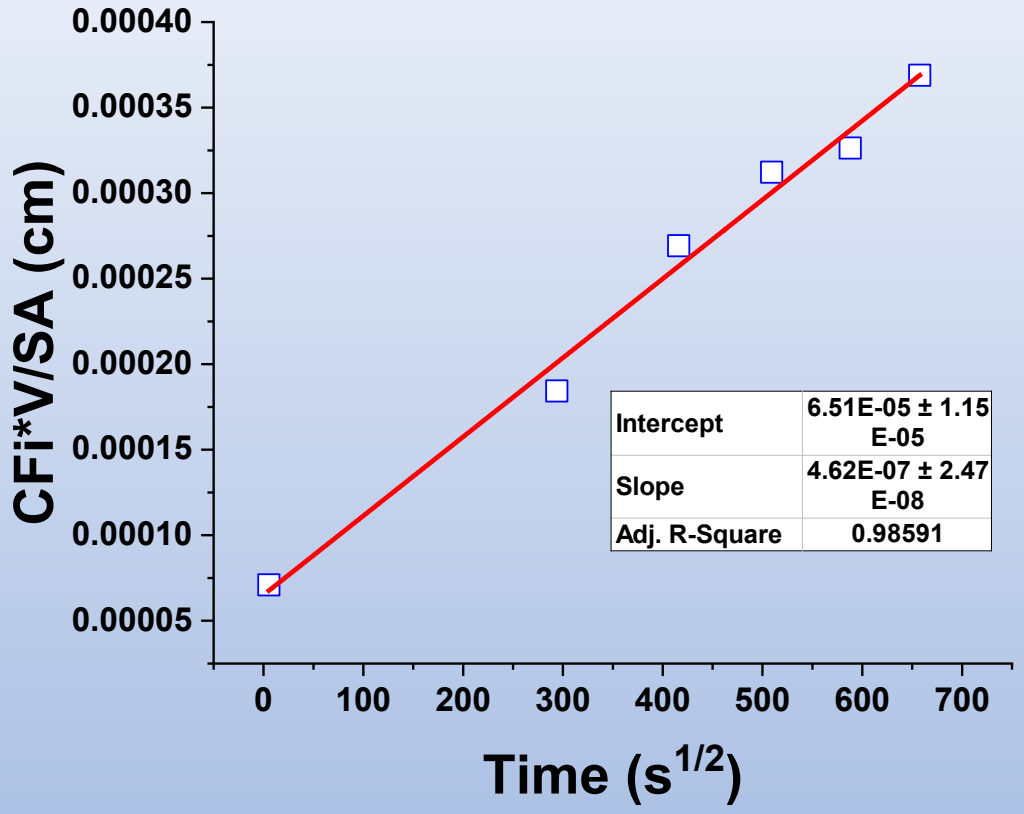
Morphology and Pore Structure Evolution with Zeolite Addition



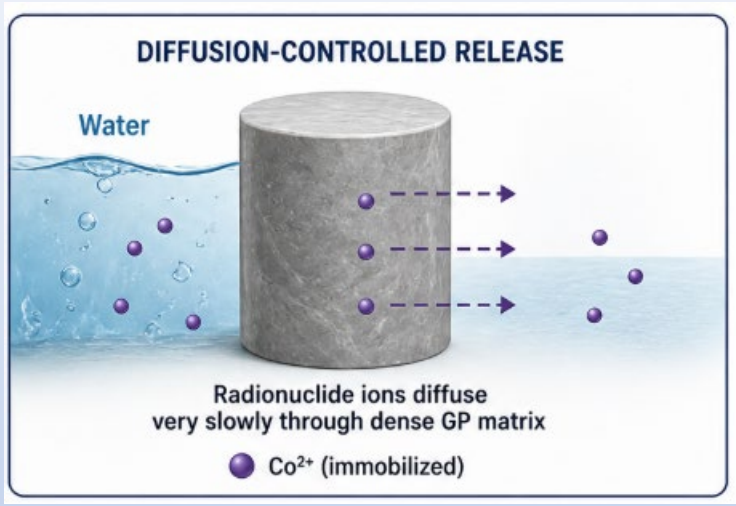


- SEM shows micropore increase with zeolite increase; however, no large cracks are showing, indicating good interfacial bonding.
- Although BET shows higher pore volume, SEM confirms these are mainly fine, well-distributed pores—not large connected defects

Leaching Test (ANSI/ANS-16.1-2019)



Radionuclide release is controlled by extra slow diffusion through a dense geopolymer



Performance matrices:

Effective diffusivity:

$D_e = 2.50 \times 10^{-13}\ cm^2\ s^{-1}$:

Leaching Index:

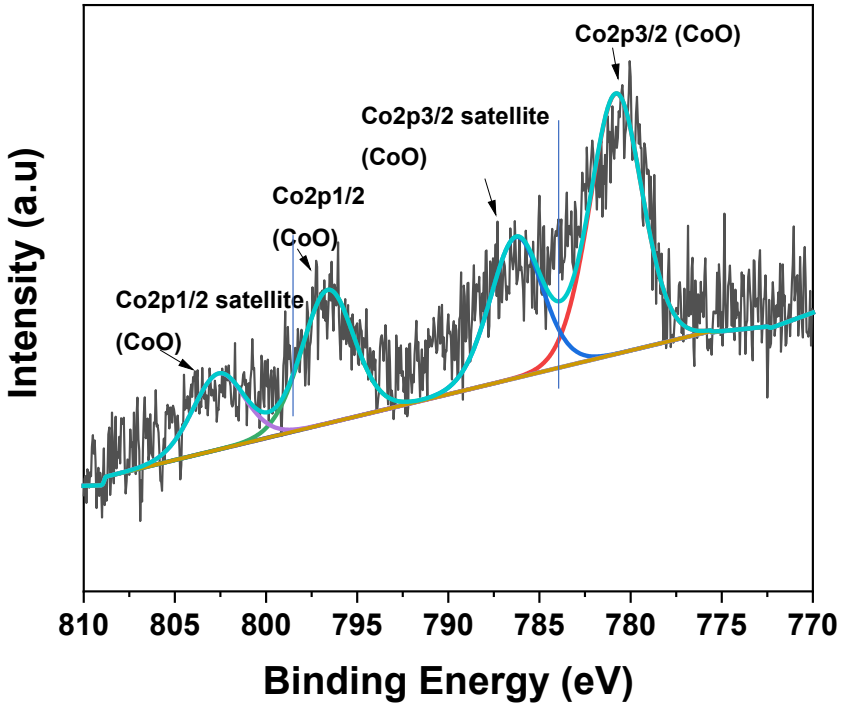
(L.I) = 12.60

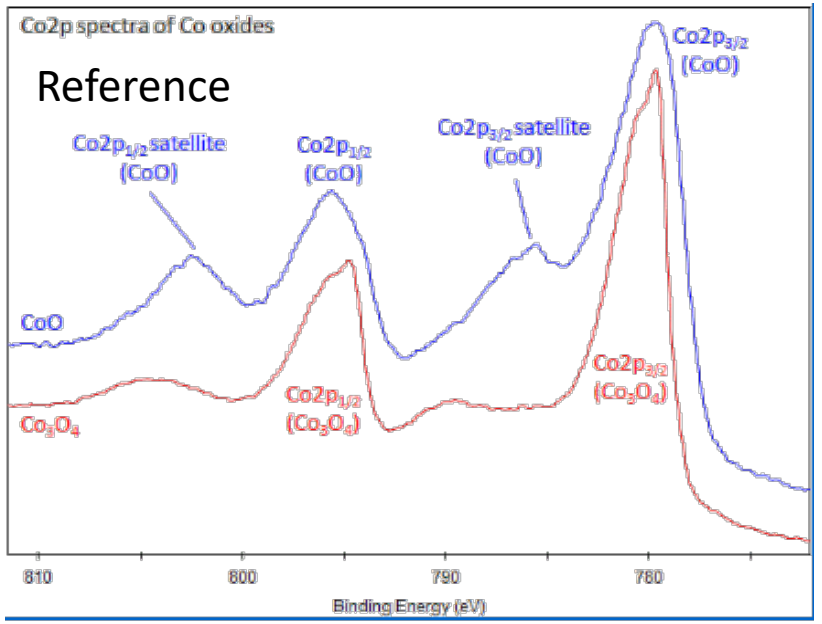
Requirement (WAC limit = 6)

*In ordinary Cement: ~6-8

Identification of the chemical environment of cobalt

The high-resolution Co 2p **XPS spectrum** exhibits two main peaks at approximately 780 eV and 795 eV corresponding to Co $2p_{3/2}$ and Co $2p_{1/2}$, respectively.

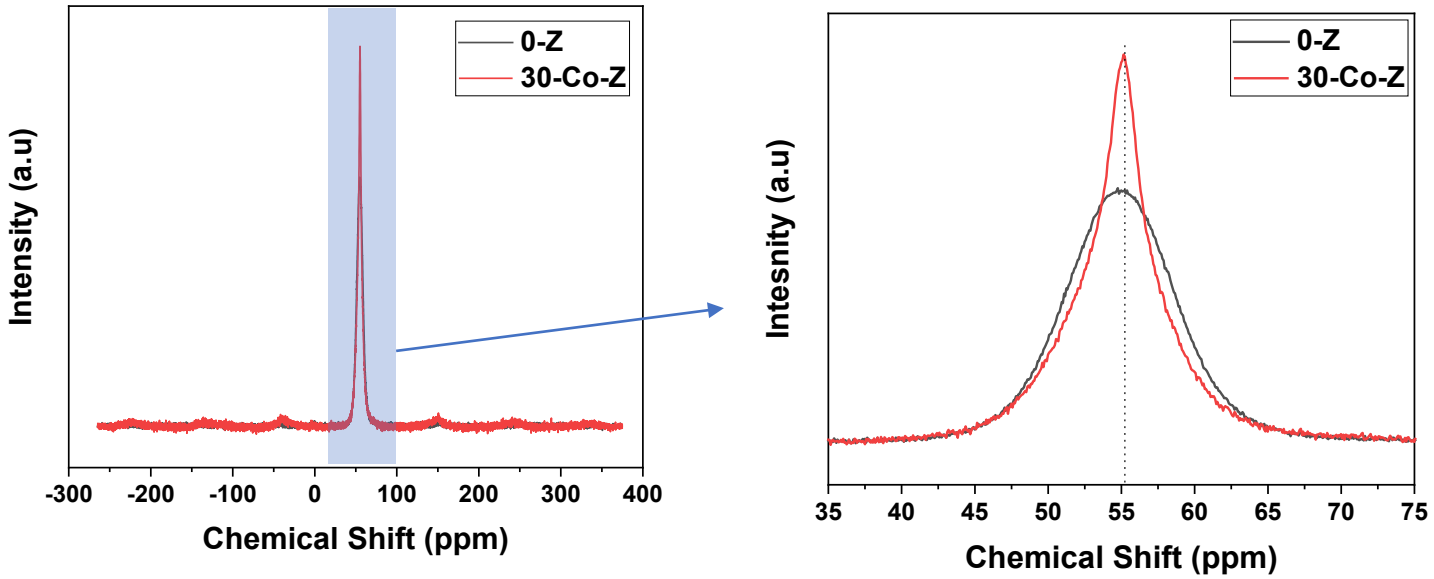




- **It's not metallic cobalt**
- **Co^{2+} is confirmed likely related to CoO**
- **Possible Exchange mechanism: $Si/Al{-}O^{-} + Co^{2+} \rightarrow Si/Al{-}O{-}Co^{+}$**

Identification of chemical environment of cobalt

^{27}Al MAS NMR technique was applied to explore the chemical bonding of Co with Al

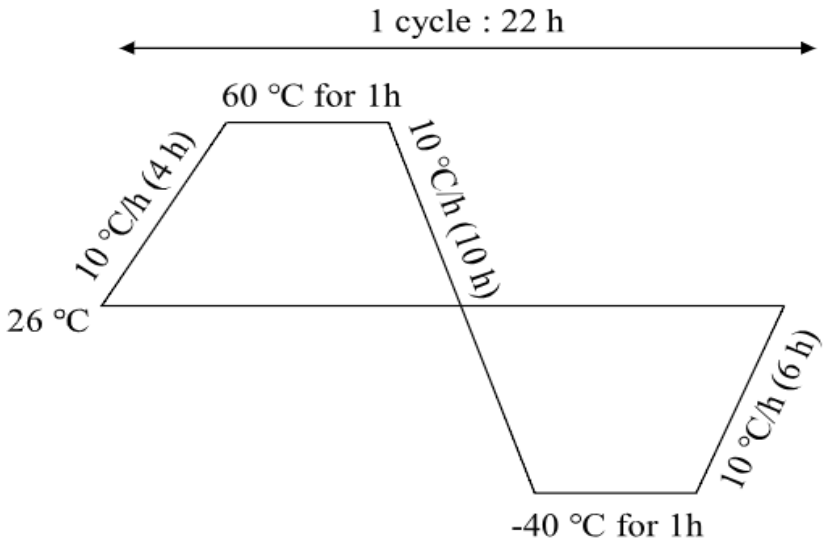


- Framework Al remains tetrahedral (~55 ppm): structure preserved
- Sharpness of the 30-Co-Z peak is due to the crystallinity of the zeolite phase
- The absence of additional NMR peaks suggests that cobalt is too dilute relative to Al to produce detectable signals and does not significantly alter the aluminum coordination in the geopolymer framework.

Waste Form Stability under Extreme Environments: Thermal Cycling Test

- **Seasonal temperature variations** (e.g., -20°C to +50°C) can cause thermal expansion and contraction, leading to degradation of WF.
- To ensure the long-term durability and stability of WF under extreme environmental conditions, a thermal cycling test was conducted using the **ASTM B553 method*.**

Total 30 cycles

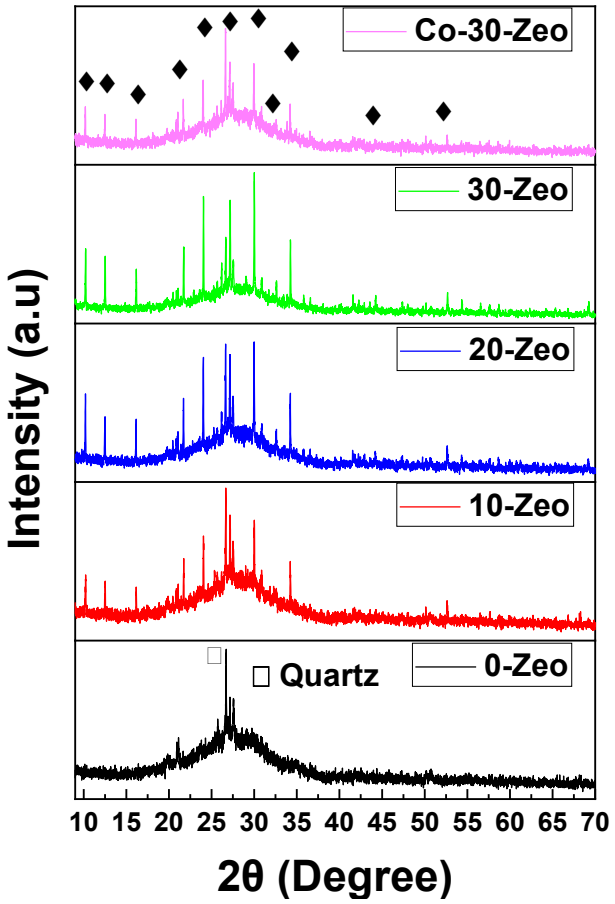


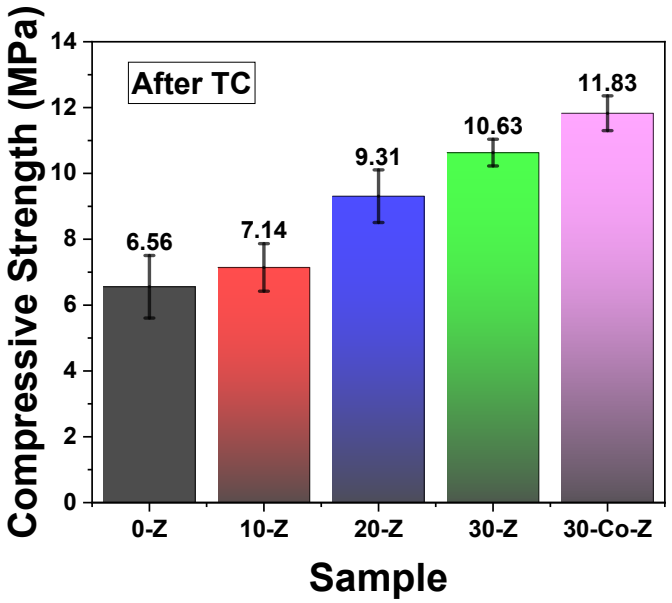
- **Evaluation Parameters**:
  - Mass loss (%)
  - Visual inspection for cracking or spalling
  - Change in **compressive strength**
  - Chemical or structural changes

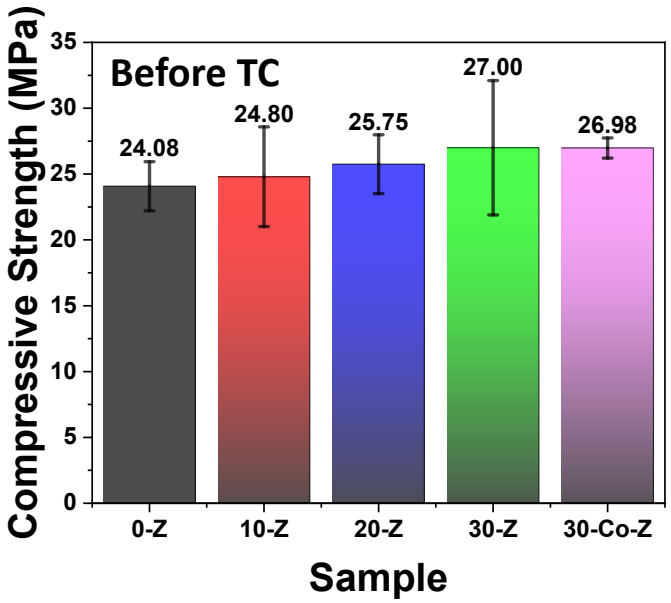
*American Society for Testing and Materials, B553, Standard Test Method for Thermocycling of Electroplated Plastics, ASTM, West Conshohocken, Pennsylvania, 1985

Thermal Cycling Test (Freeze-Thaw Test)

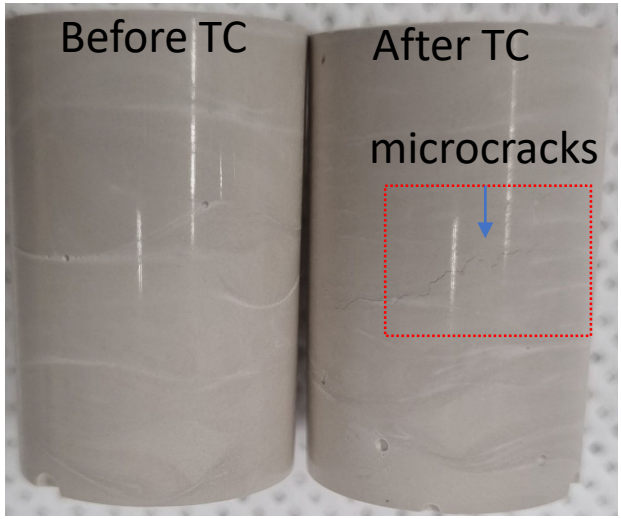
Zeolite A: (Card No. 96-153-2391)
Quartz: Card No.: 01-078-1252







- No phase change (XRD stable)
- **50 % drop in compressive strength** caused by **microcrack formation and internal damage.**
- **Zeolite improves Freeze-Thaw durability** by possibly acting as a stress buffer (due to high internal porosity) and internal curing agent in the geopolymer matrix



Key Outcomes

- Nearly **100% cobalt** removal was achieved using zeolite
- Up to **30 wt% Co-** zeolite waste was successfully immobilized into the geopolymer matrix.
- Zeolite incorporation **enhanced the mechanical strength** of the geopolymer.
- **Excellent durability (L.I. = 12.6): Strong retention, hindered transport.**
- Cobalt release is predominantly governed by **diffusion-controlled release** → long-term stability

This work demonstrates a simple but effective pathway to convert secondary radioactive waste into a durable and disposal-ready waste form

What’s Next?

- **Scale-up validation**
  - Pilot-scale geopolymer blocks (kg-scale validation)
- **Real waste testing**
  - Kori sludge / complex mixed waste streams
- **Long-term durability**
  - Extended leaching, thermal & radiation stability
- **Process integration**
  - Coupling decontamination → direct solidification
- **Regulatory alignment**
  - Meet WAC and disposal criteria?

Thank you for listening

Question?