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The effect of gamma irradiation on Fe-rich slag-based inorganic polymers

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The effect of gamma irradiation on Fe-rich slag-based inorganic polymers

What is an IP?

Why using IPs in nuclear industry?

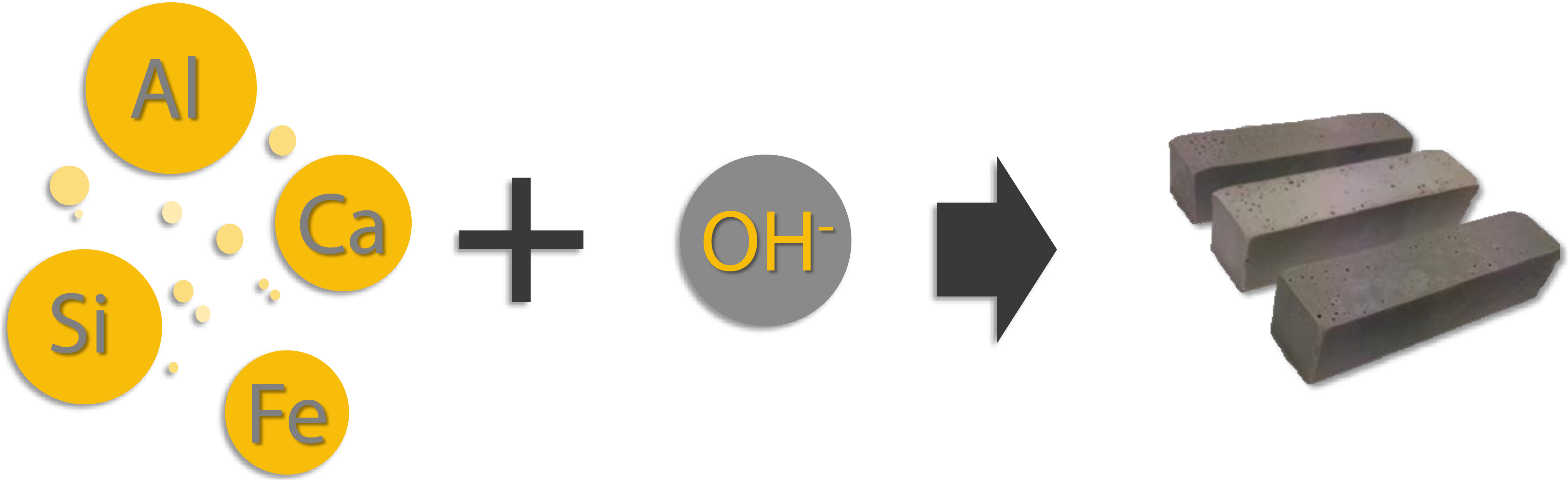
Which γ-irradiation effects occur in Fe-rich IPs ?

What is an IP?

What is an IP?

The production process

Alkali Activation



Amorphous precursor

copper slag, plasma slag,
metakaolin, fly ash, …

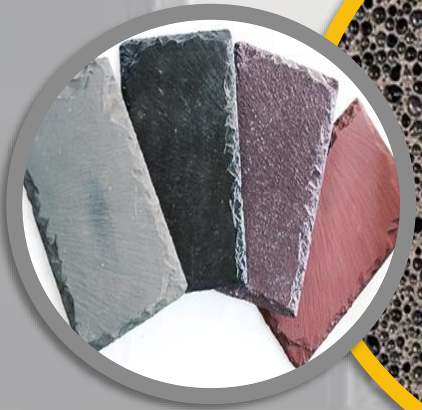
Alkaline activation solution

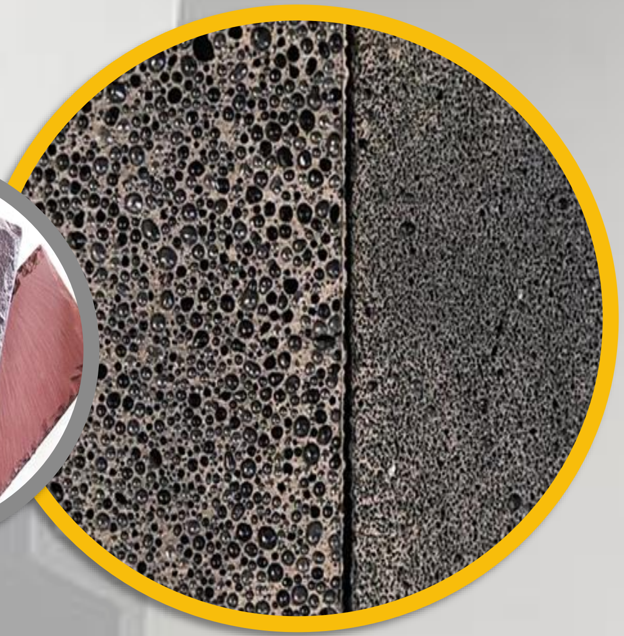
alkali hydroxides, silicates,
carbonates, sulphates, …

Inorganic Polymer

What is an IP?

The end product





Application

Optimized

Mix Design

Why using IPs?

Why using IPs?

Closing the loop – from residue to construction material

Inorganic Industrial Residues

can be used as an input material, reducing disposal of residues as

Fly Ash

Red Mud

Blast Furnace Slag

Phosphogypsum

Plasmaslag

and can moreover offer a safe potential for (re)using materials containing

Naturally Occurring Radionuclides

Why using IPs?

Tailor-made materials with excellent performances

Design 4 Application

- ✓ High (early) strength
- ✓ Controllable porosity
- ✓ High chemical resistance
- ✓ High temperature resistance
- ✓ Low water content
- ✓ High alkalinity
- ✓ Good immobilization capacity
- ✓ No important ASR

Why using IPs in nuclear industry?

Tailor-made materials with excellent performances

Design 4 Application in Nuclear industry

ADVANTAGES of IPs:

- ✓ Less Ca
- ✓ Reduced water content
- ✓ High alkalinity
- ✓ Low porosity/high density

Radioactive waste encapsulation

- Less $Ca(OH)_2$
- No damage as a result of dehydration
- Formation of precipitates
- Protection from steel corrosion
- High immobilization capacity

Ionizing radiation Shielding

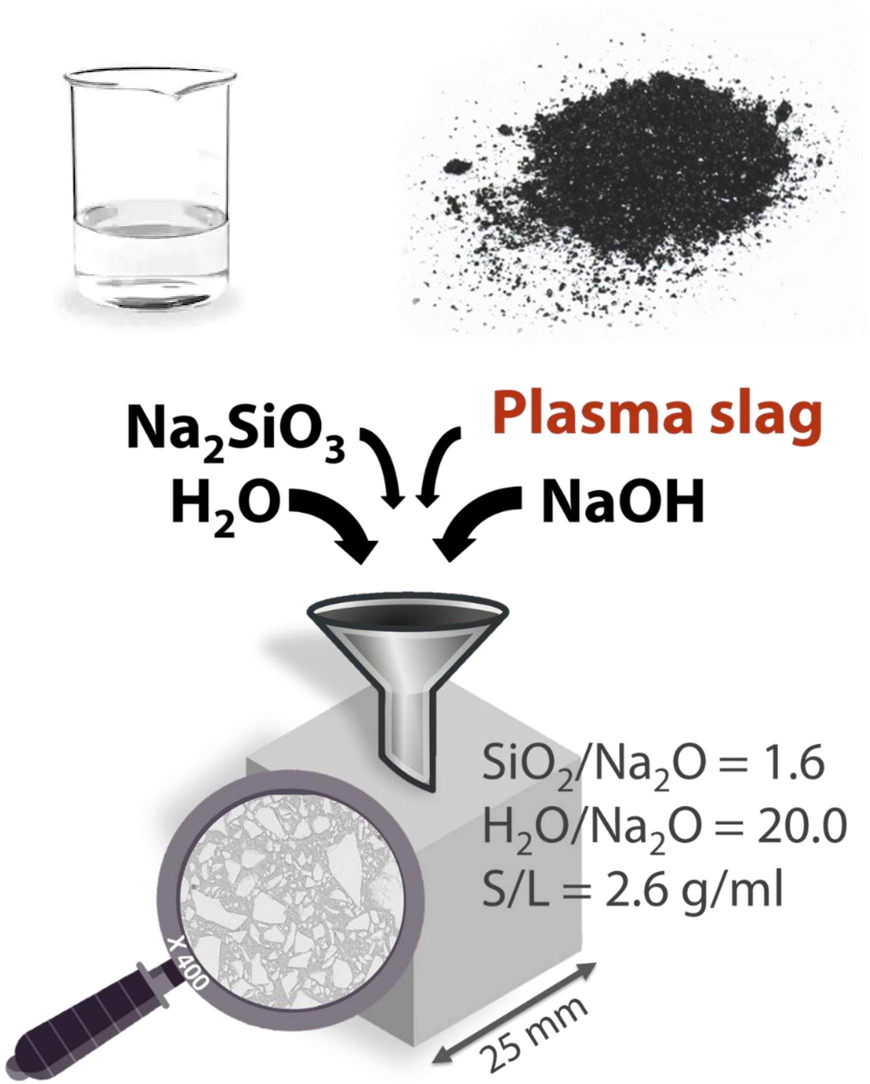
- Neutron activation of ^{40}Ca is reduced
- Low radiolytic H_2 yield
- Good shielding of γ-radiation

B. Mast, Y. Pontikes, W. Schroeyers, B. Vandoren, and S. Schreurs, "The use of alkali activated materials in nuclear industry," in Comprehensive Nuclear Materials, 2nd ed., R. Konings, Ed. Elsevier Inc., 2020.

Which γ-irradiation effects occur in Fe-rich IPs?

Effect of gamma irradiation on Fe-rich inorganic polymers

The experimental design

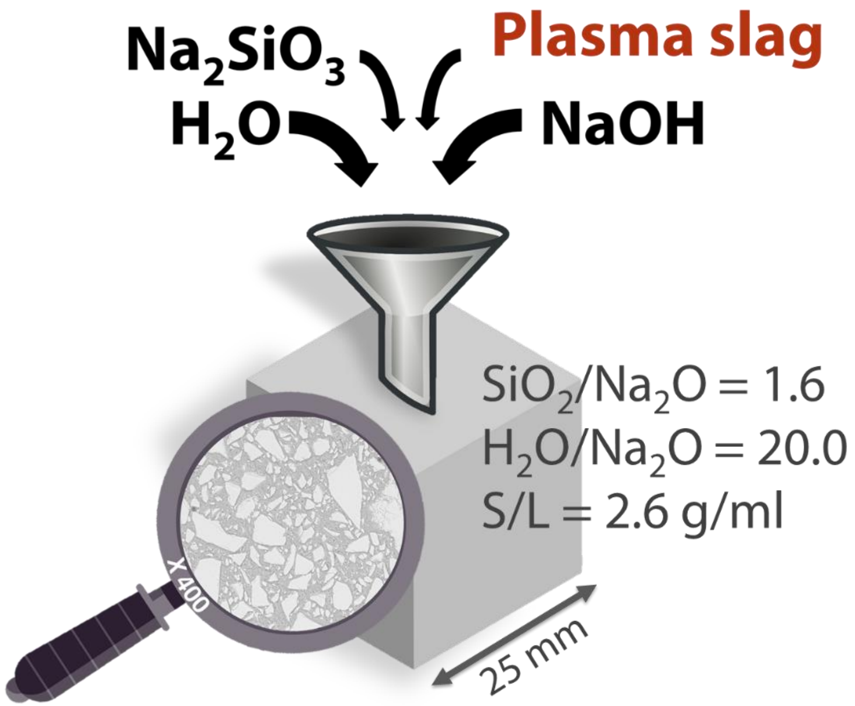


Plasma slag	wt.%
SiO_2	29.2
Fe_xO_y	28.2
CaO	26.7
Al_2O_3	13.4
Other	2.5

> 98 wt.% amorphous

Effect of gamma irradiation on Fe-rich inorganic polymers

The experimental design



Time of hardening prior to irradiation:

1 h: minimum time required
24 h: right after the main reaction peak
28 d: stable and fully cured

Effect of gamma irradiation on Fe-rich inorganic polymers

The experimental design

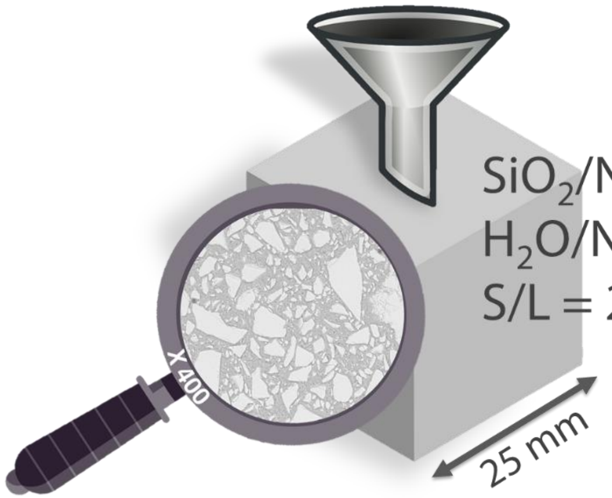
Set-up	Source	Dose rate
Brigitte sck cen	^{60}Co	8.85 kGy/h

Effect of gamma irradiation on Fe-rich inorganic polymers

The experimental design

Na_2SiO_3 **Plasma slag**

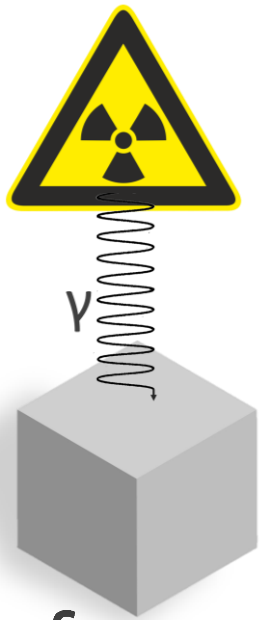
H_2O NaOH



$SiO_2/Na_2O = 1.6$
$H_2O/Na_2O = 20.0$
S/L = 2.6 g/ml

1 h, 24 h OR 28 d

22.5 h



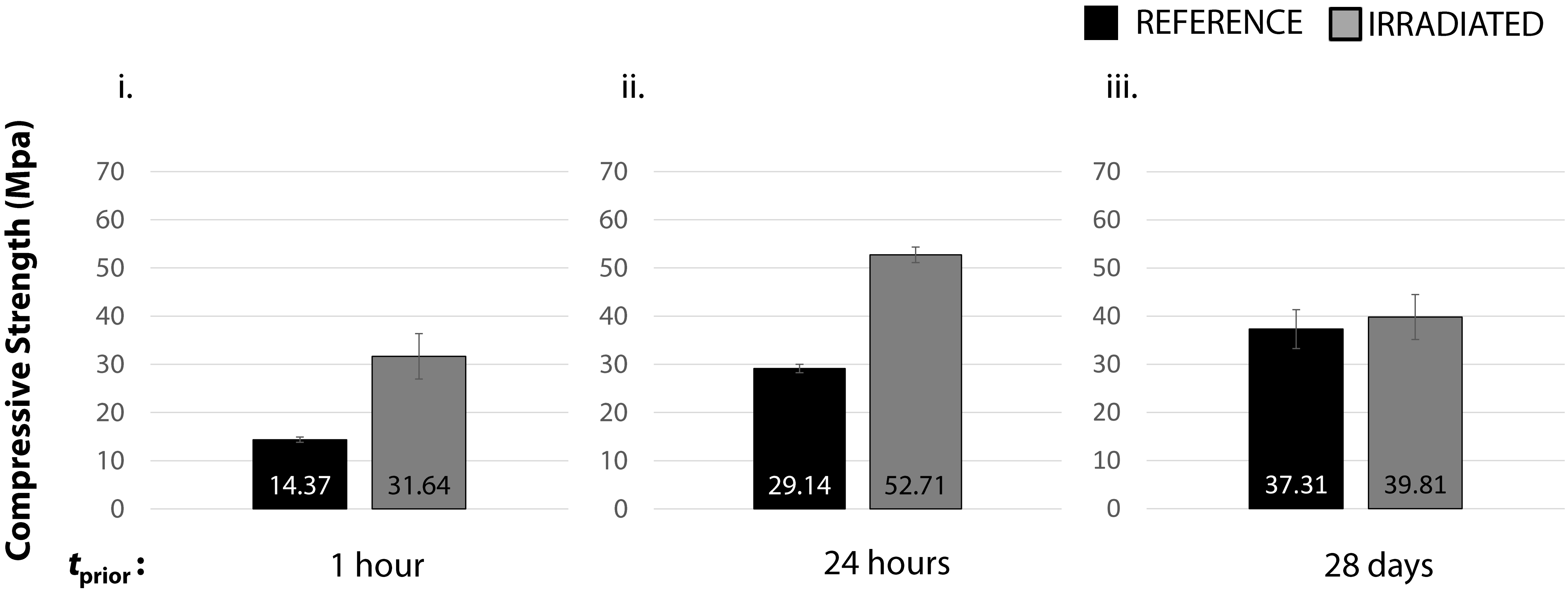
Set-up	Source	Dose rate
Brigitte sck cen	^{60}Co	8.85 kGy/h

Characterisation techniques:

- Compressive strength test
- Nanoindentation
- ^{57}Fe Mössbauer spectroscopy
- Mercury Intrusion Porosimetry
- Infrared Spectroscopy
- Thermogravimetry

Effect of gamma irradiation on Fe-rich inorganic polymers

Results of Uniaxial Compression Test – increase in compressive strength



Increase in compressive strength by factor x 2.2

Increase in compressive strength by factor x 1.8

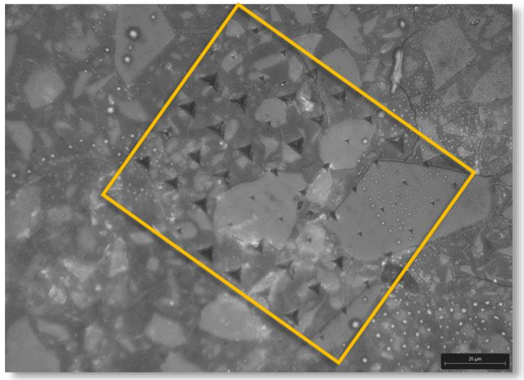
No change in compressive strength

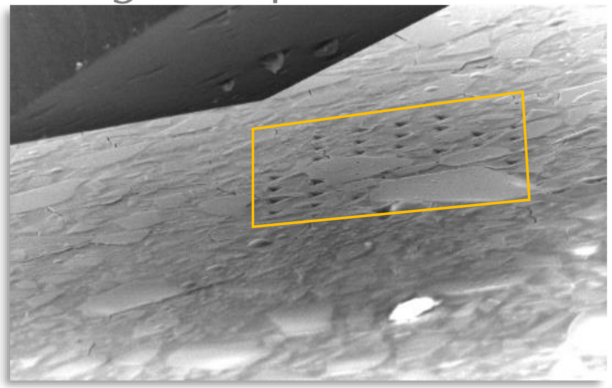
Effect of gamma irradiation on Fe-rich inorganic polymers

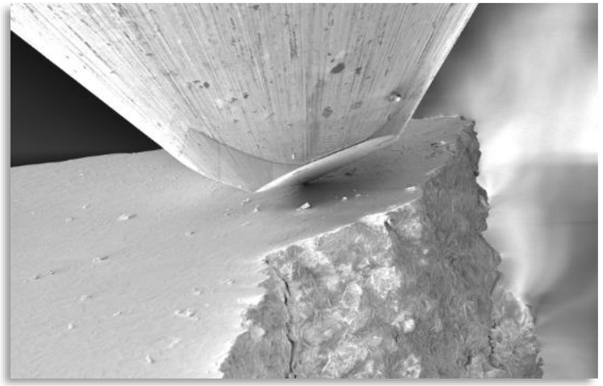
Results of Nanoindentation – decrease of hardness, elasticity and creep of the binder t_{prior} = 1 h

t_{prior}	1 h	24 h	28 d
Hardness (binder)	1.20 ± 0.07 GPa ↓ 0.87 ± 0.08 GPa	0.92 ± 0.03 GPa ↑ 1.19 ± 0.08 Gpa	≈ 1.05 ± 0.07 GPa
Elasticity (binder)	32 ± 1 GPa ↓ 21.5 ± 0.9 GPa	≈ 23 ± 1 Gpa	≈ 24 ± 1 GPa
Creep (binder)	8.2 ± 0.6% ↓ 5.9 ± 0.4%	≈ 5.9 ± 0.7%	≈ 7.2 ± 0.6%

Effects are highly dependant on the curing time prior to irradiation







Effect of gamma irradiation on Fe-rich inorganic polymers

Results of Mercury Intrusion Porosimetry– change in pore size distribution and porosity

—— REFERENCE - - - IRRADIATED

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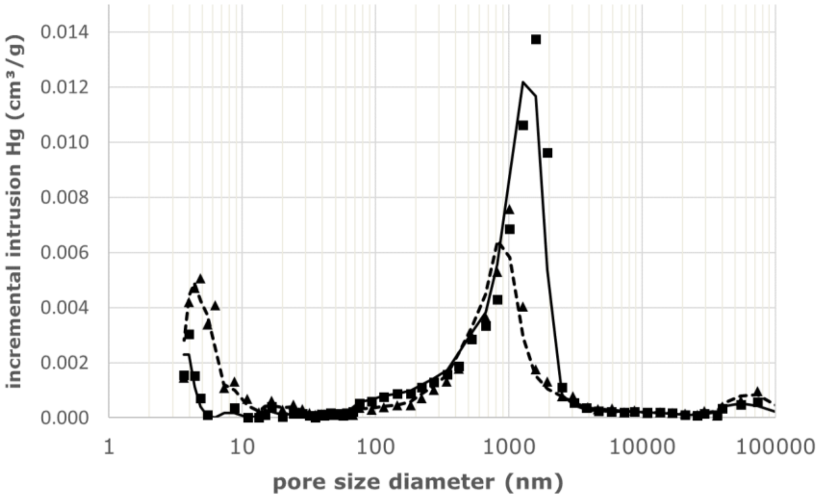

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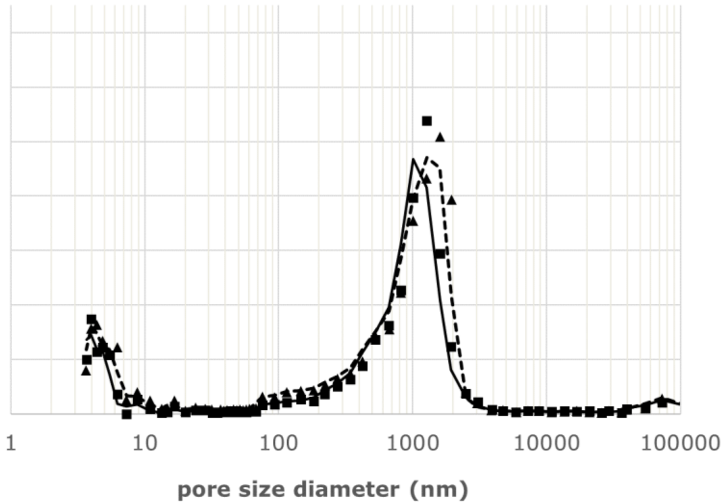

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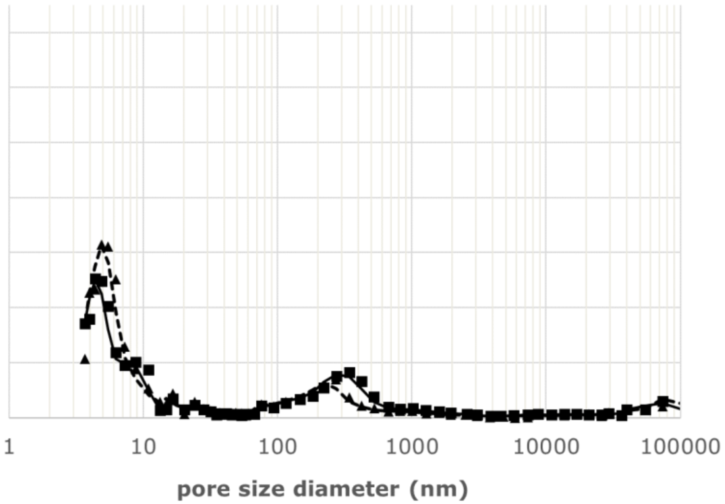


t_{prior}: 1 hour | 24 hours | 28 days

Decrease in porosity (-2.7%)

Shift to smaller pore sizes

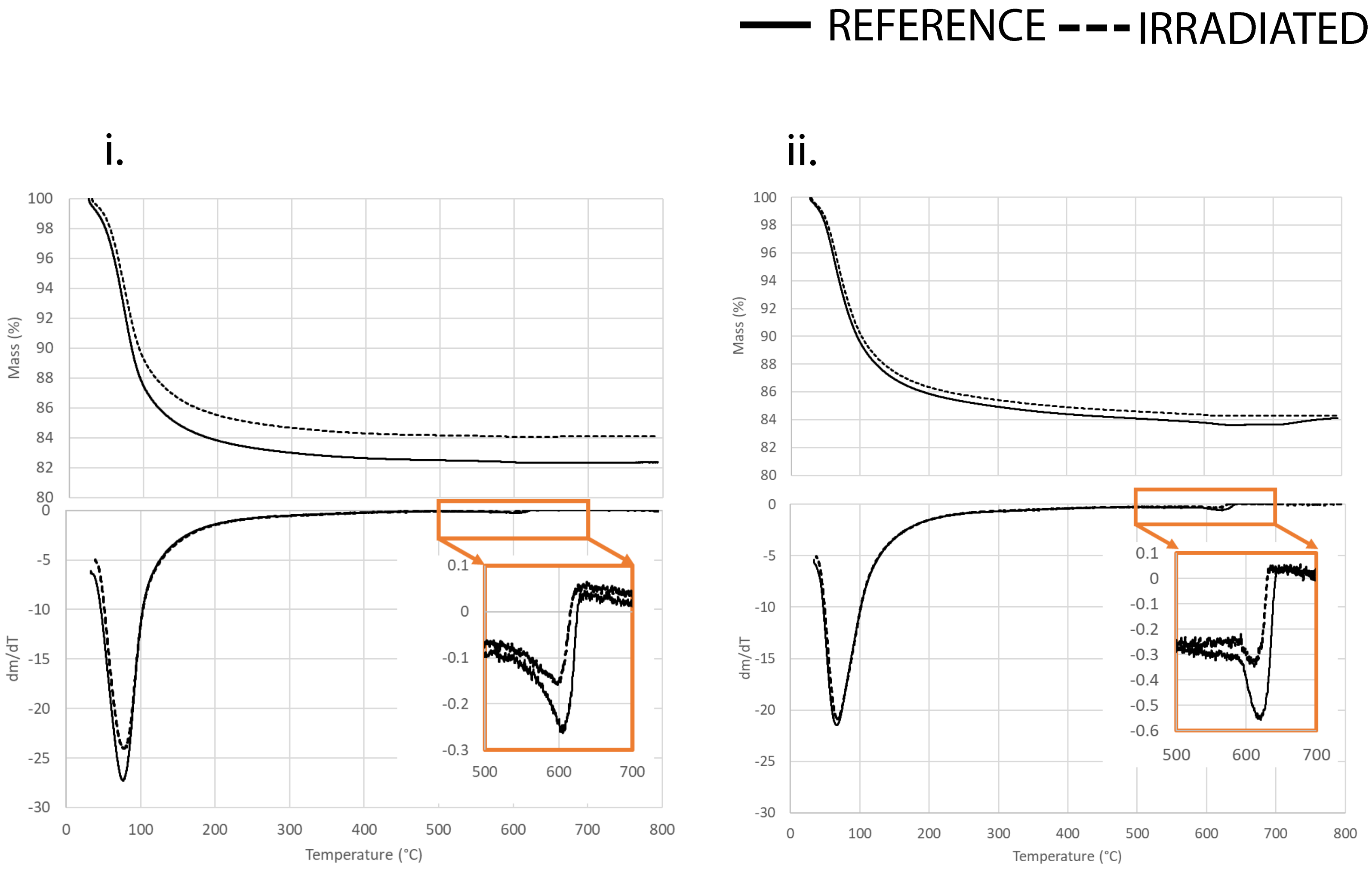
Increase in porosity (+3.0%)

Shift to larger pore sizes

No change in porosity

Effect of gamma irradiation on Fe-rich inorganic polymers

Results of TGA (and IR)– decrease in water content & radiation-altered carbonation



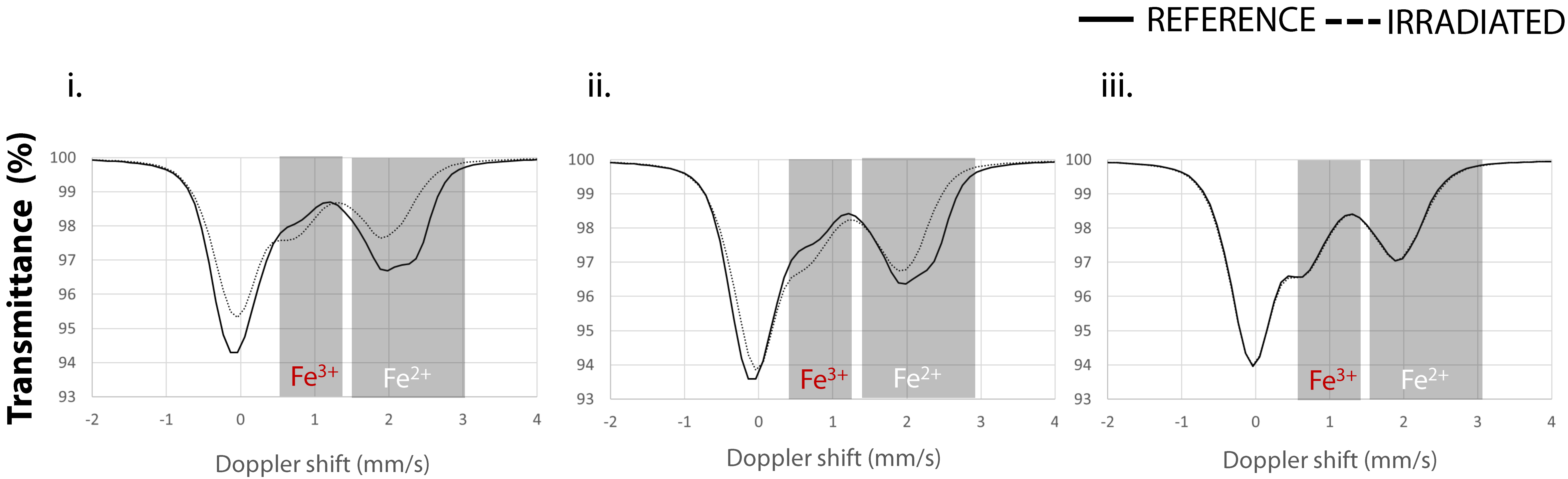
24 hours

28 days

t_{prior}:

Effect of gamma irradiation on Fe-rich inorganic polymers

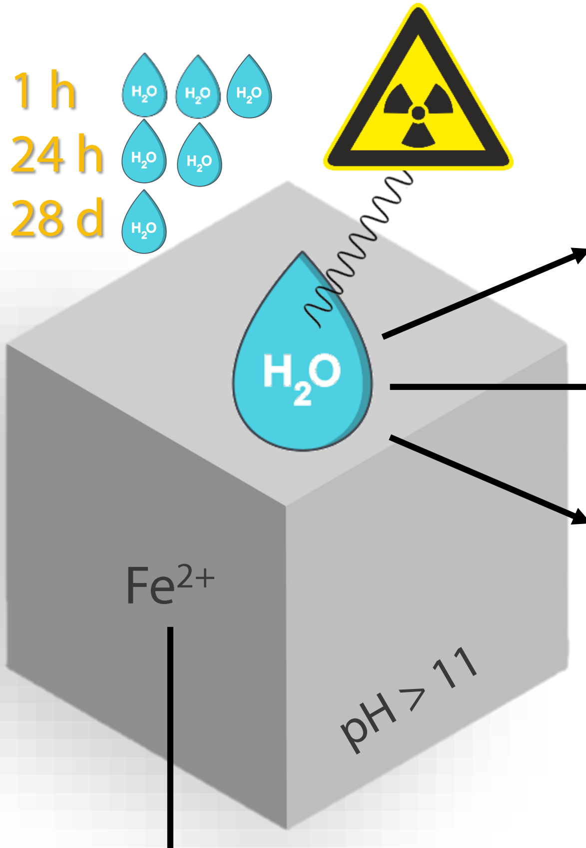
Results of ^{57}Fe Mössbauer spectroscopy – radiation-induced iron oxidation



t_{prior}:	1 hour	24 hours	28 days
Fe^{3+}_{REF}	20%	24%	43%
Fe^{3+}_{IR}	39%	38%	43%

Effect of gamma irradiation on Fe-rich inorganic polymers

Radiation-induced iron oxidation - explanation



H_2

H_2O_2 : strong oxidant → Fe^{2+} oxidation

O_2 acts as catalyst for $^{\bullet}e_{aq}^{-}$ removal

Fenton Reactions:

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + {}^{\bullet}OH + OH^{-}$$

$$Fe^{3+} + H_2O_2 \rightarrow Fe^{2+} + {}^{\bullet}OOH + H^{+}$$

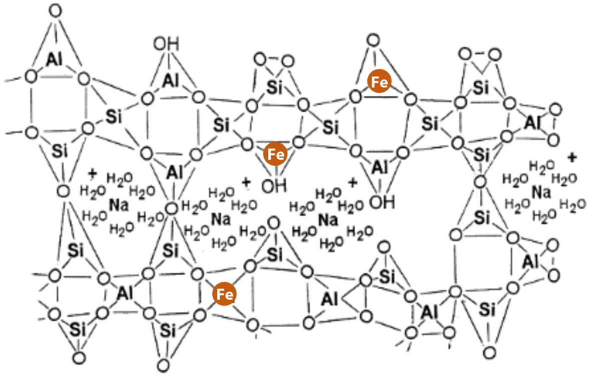
Very strong oxidants

Formation of FeO(OH) nanoparticles

Nucleation sites for polymerization

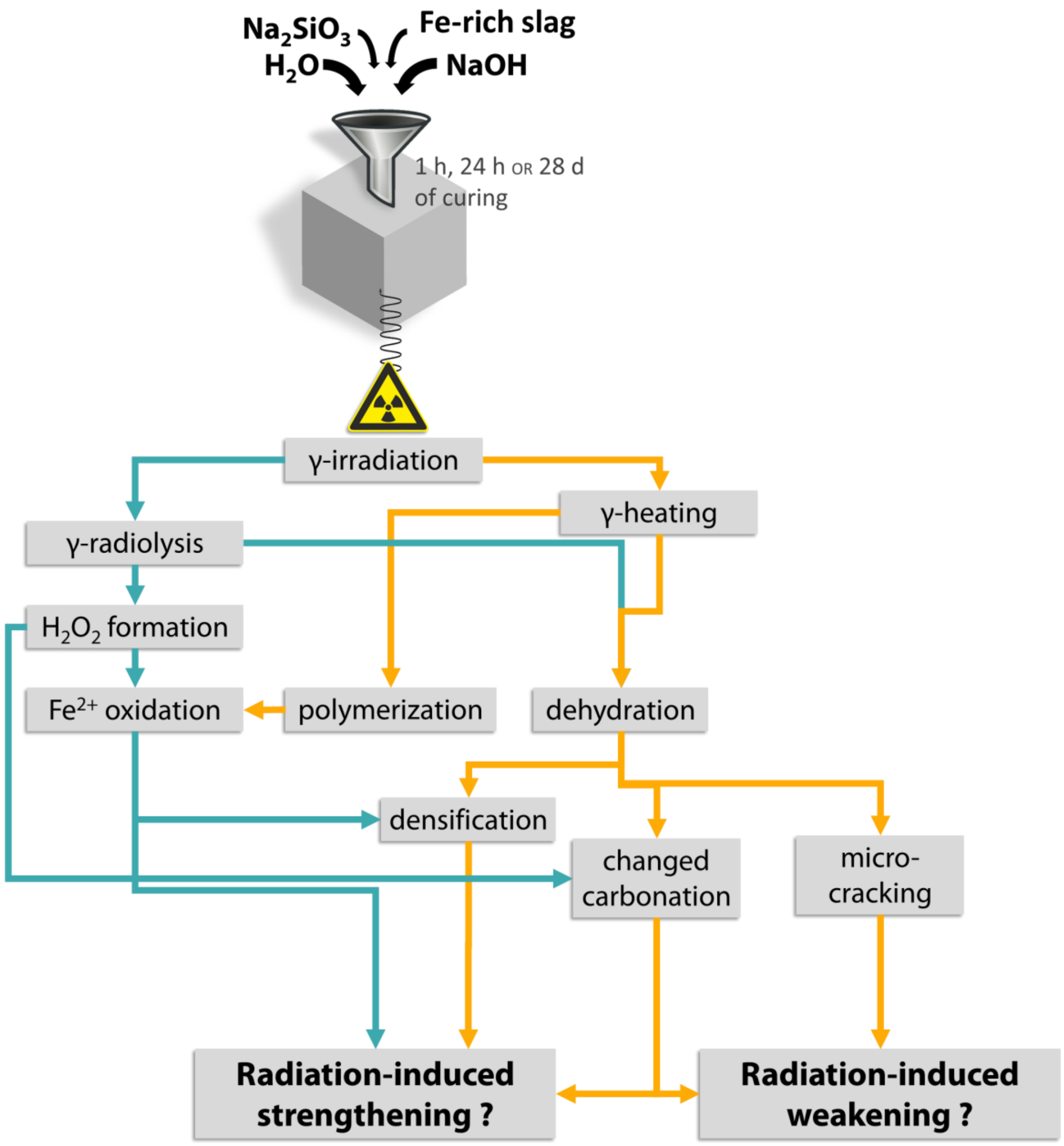
Fe^{3+} incorporation in tetrahedral network

Strength Increase



Effect of gamma irradiation on Fe-rich inorganic polymers

Summary



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Related work

[1] B. Mast, I. Gerardy, Y. Pontikes, W. Schroeyers, B. Reniers, P. Samyn, G. Gryglewicz, B. Vandoren, S. Schreurs, "The effect of gamma radiation on the mechanical and microstructural properties of Fe-rich inorganic polymers," J. Nucl. Mater., vol. 521, 2019.

[2] B. Mast, Y. Pontikes, W. Schroeyers, B. Vandoren, and S. Schreurs, "The use of alkali activated materials in nuclear industry," in Comprehensive Nuclear Materials, 2nd ed., R. Konings, Ed. Elsevier Inc., 2020.

[3] B. Mast , A. Cambriani, A. P. Douvalis, Y. Pontikes, W. Schroeyers, B. Vandoren, S. Schreurs, "The effect of high dose rate gamma irradiation on the curing of CaO-FeOx-SiO2 slag based inorganic polymers: Mechanical and microstructural analysis," J. Nucl. Mater., 2020.

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