

31-01-2020

Mini-workshop on Geopolymers

The effect of gamma irradiation on CaO-FeOx-SiO₂ slag based inorganic polymers

*Radiation-induced mechanical degradation in alkali activated materials:
a combined experimental - computational approach*

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KUL



Inorganic Polymers

Belgian nuclear waste management

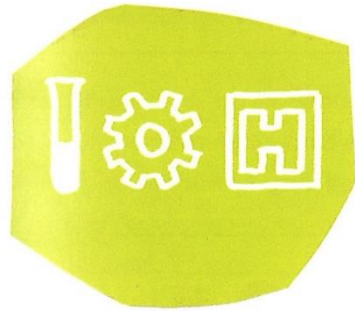
Possibilities of IPs in nuclear industry

Effect of gamma irradiation on
Fe-rich inorganic polymers

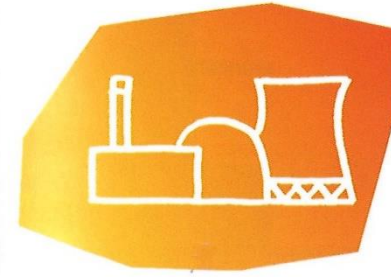
**as safety material
for nuclear industry**

Belgian nuclear waste management

Radioactive Wastes



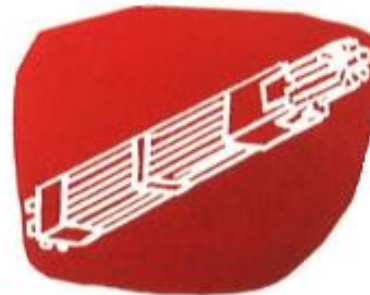
Nuclear medicine
Research
Industry



Energy production



Demolition



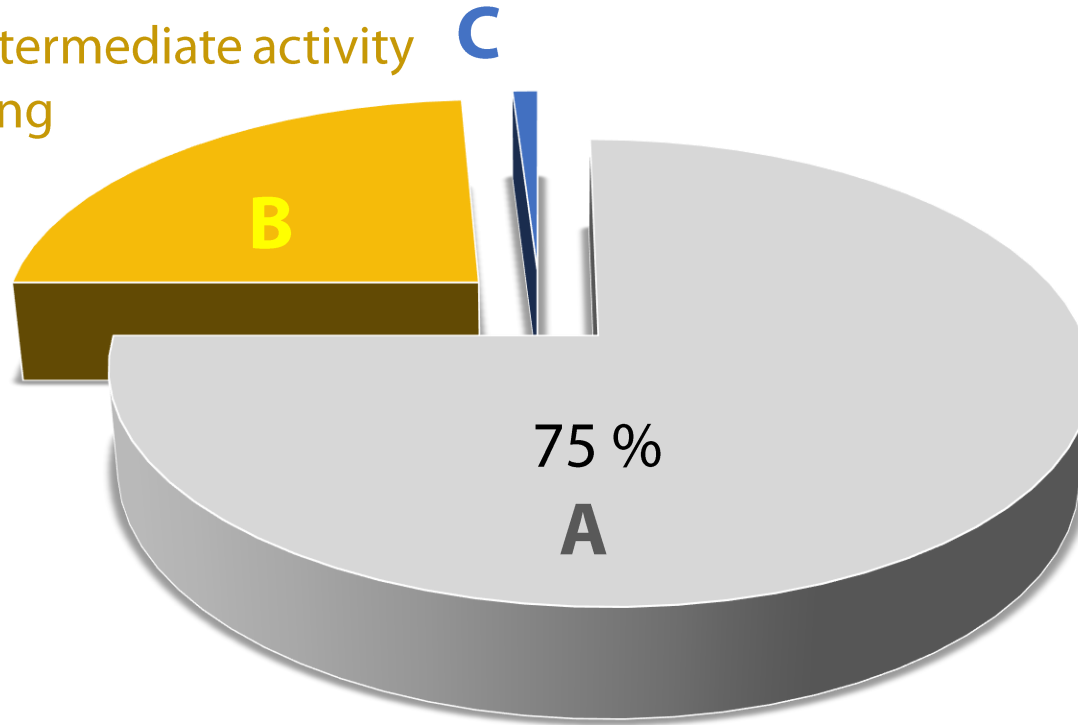
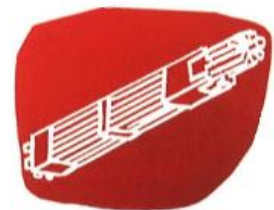
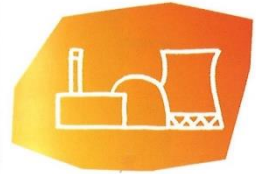
Nuclear fuel cycle

Belgian nuclear waste management

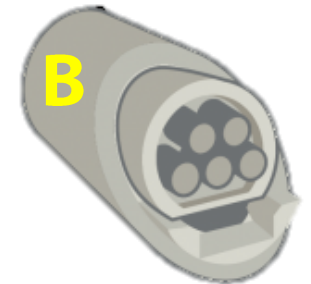


- High activity
- Long living

- Low & Intermediate activity
- Long living



- Low & Intermediate activity
- Short living



Challenges of Long-term storage of radioactive wastes

- Durability up to 100,000 – 1,000,000 years service life?
→ Dependent on type of nuclear waste & country specific
- Binding of radionuclides
- Low heat evolution
- Dimensional stability
- Controlled corrosion of reactive metals
- **Stability under irradiation**
- Properties of activated components
- ...

Challenges of Long-term storage of radioactive wastes

Possible effects of ionizing radiation on concrete:

- Dehydration

Radiolysis

→ decomposition of water

Heating

→ evaporation of water

Drying & Shrinkage

- Radiation-induced carbonation

Radiolysis

→ formation of H_2O_2

→ $\text{CaO}_2 \cdot \text{H}_2\text{O} \rightarrow \text{Ca(OH)}$

→ CaCO_3

Decomposition

Cracking & Loss of strength

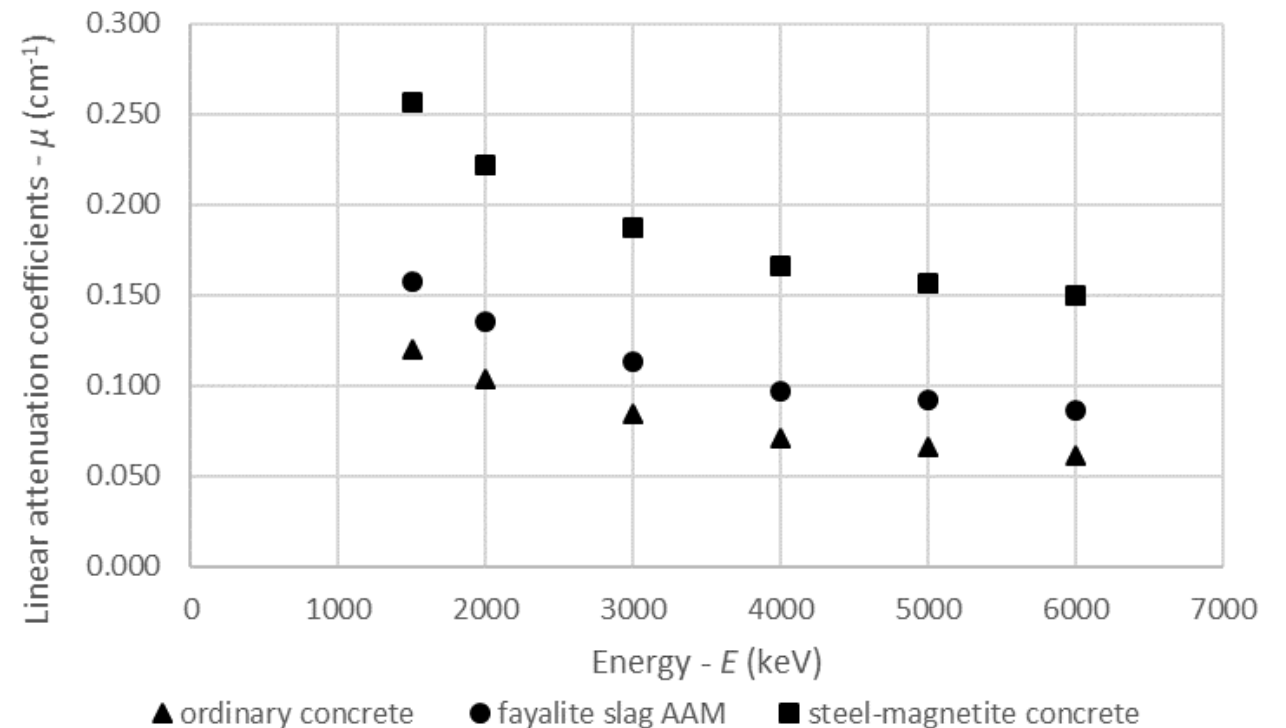
Possibilities of IPs in nuclear industry



B. Mast, Y. Pontikes, W. Schroeyers, B. Vandoren, and S. Schreurs,
"00135 Alkali Activated Materials - The use of alkali activated materials in nuclear industry,"
in *Comprehensive Nuclear Materials*, 2nd ed., 2020.

IPs as shielding material

- **Low radiolytic H₂ yield**
- **No damage as a result of dehydration**
- **Good Shielding of γ -radiation**

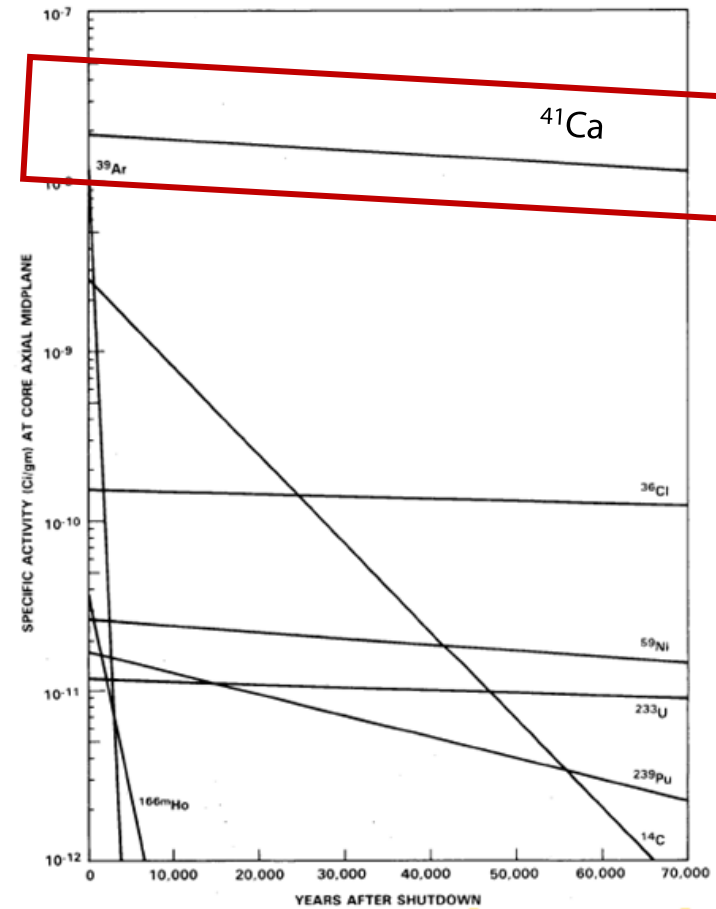
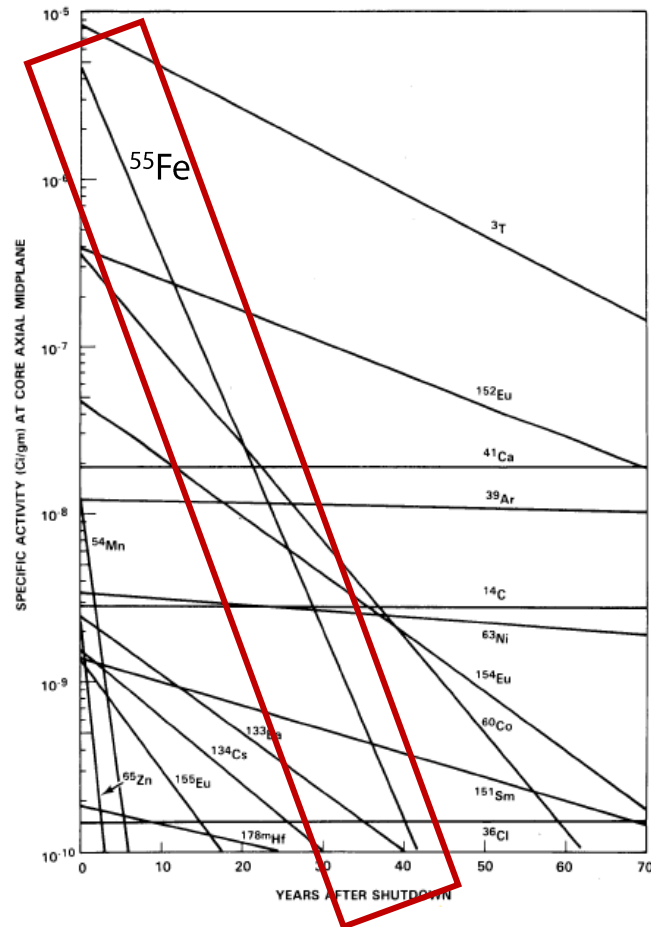


IPs as shielding material

- **Low radiolytic H₂ yield**
- **No damage as a result of dehydration**
- **Good Shielding of γ -radiation**
- **Activation with Ba(OH)₂ could improve the shielding performance due to high atomic number**
- **Boron could substitute Al for good neutron shielding**
- **Better neutron shielding than aged OPC concrete**
- **Neutron activation of ⁴⁰Ca can be reduced**

IPs as shielding material

- Neutron activation of ^{40}Ca can be reduced



IPs as conditioning material

Advantages

- **Absence of $\text{Ca}(\text{OH})_2$**
- **Reduced water content**
- **High alkalinity**
- **Excellent fire resistance**
- **Low porosity**

Mechanisms

- **Uptake as charge-balancing ion in the structure**
- **Incorporation in aluminosilicate structure**
- **Formation of precipitates**
- **Adsorption on self-generated or introduced zeolite structures**

IPs as conditioning material

Example: Mg-Zr alloys

At high pH:

High hydrogen gas generation as result of $\text{Mg}(\text{OH})_2$ formation

Solution:

Addition of sodium fluoride as Mg corrosion inhibitor \rightarrow formation of passivation layer

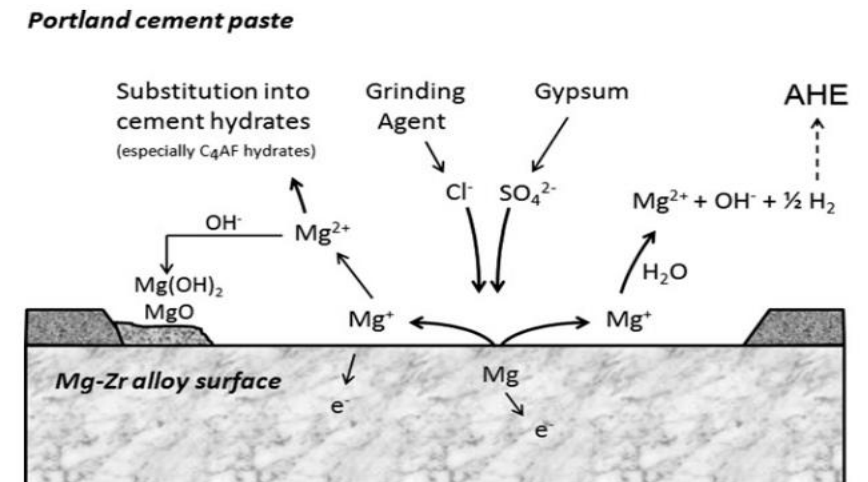
Problem in OPC:

Formation of CaF_2 precipitates \rightarrow deterioration

Destabilization of protection layer

Solution:

Ca-free low pH IPs



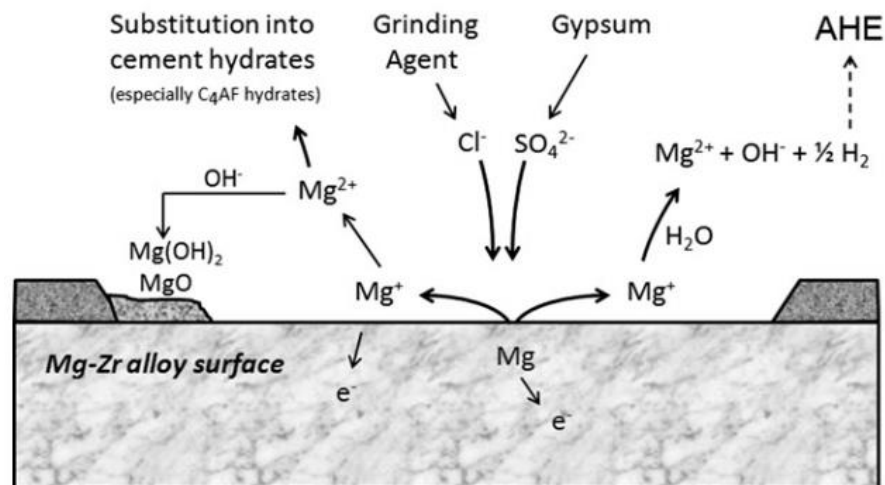
IPs as conditioning material

Example: Mg-Zr alloys

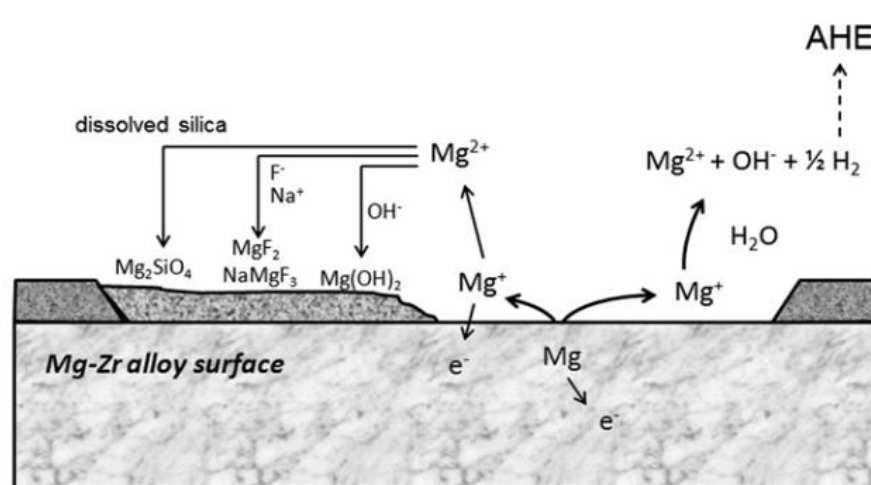
Solution:
Ca-free low pH IPs

- Formation of MgSiO_4 passivation layer

Portland cement paste

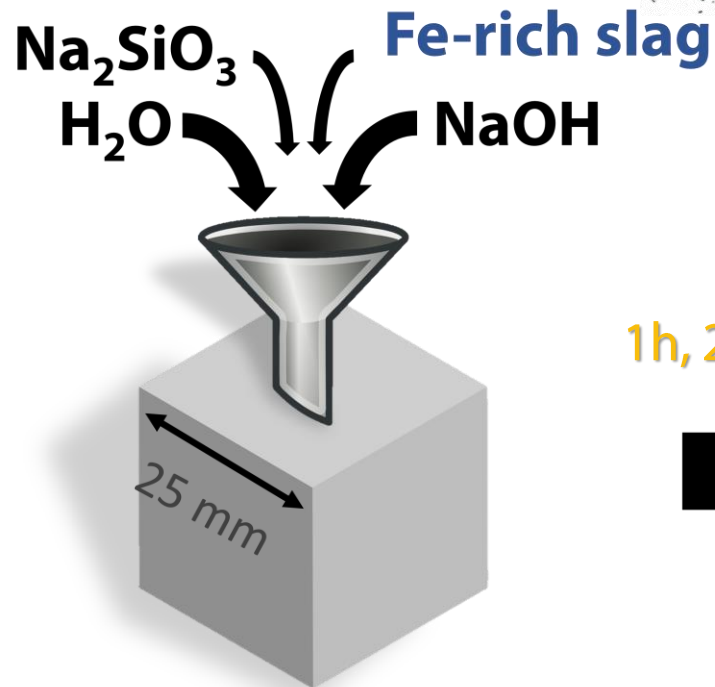


Geopolymer Paste (containing NaF)

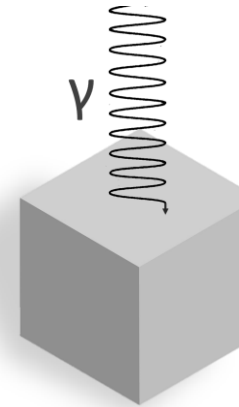
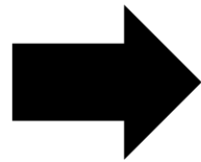


Effect of gamma irradiation on Fe-rich inorganic polymers

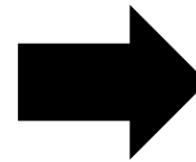
Effect of gamma irradiation on Fe-rich inorganic polymers



1h, 24h, 28d



2 Gy/h – 10 000 Gy/h

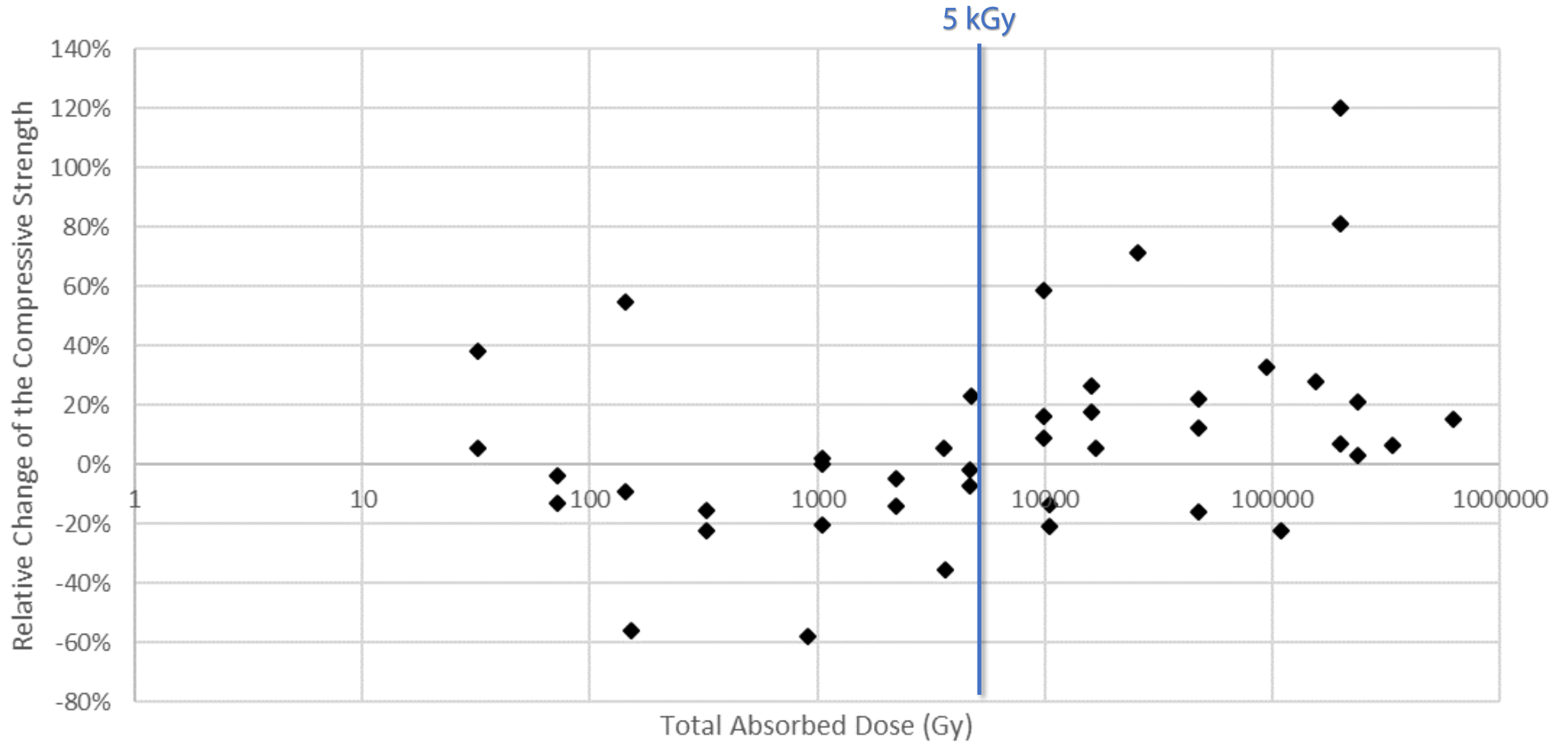


Evaluation:

- Nanoindentation
- Compression strength test
- ATR-FTIR
- NMR
- MIP
- SEM
- TGA
- ^{57}Fe Mössbauer spectroscopy

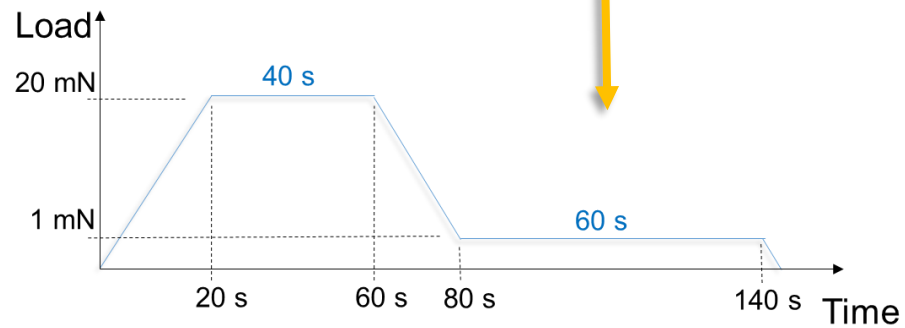
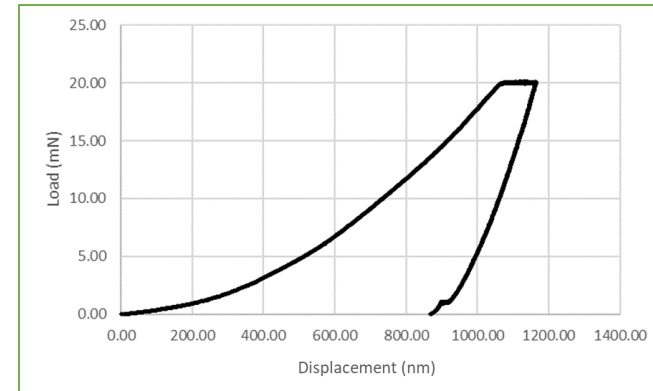
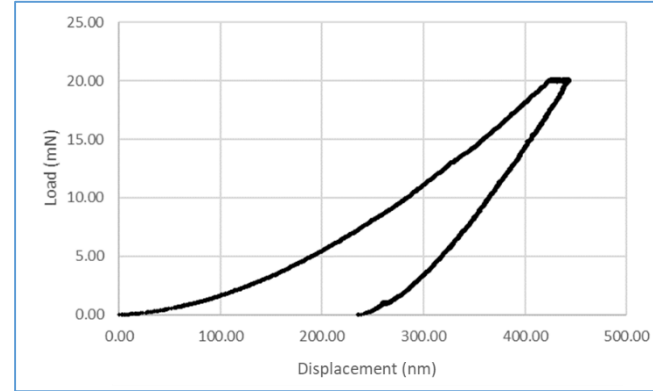
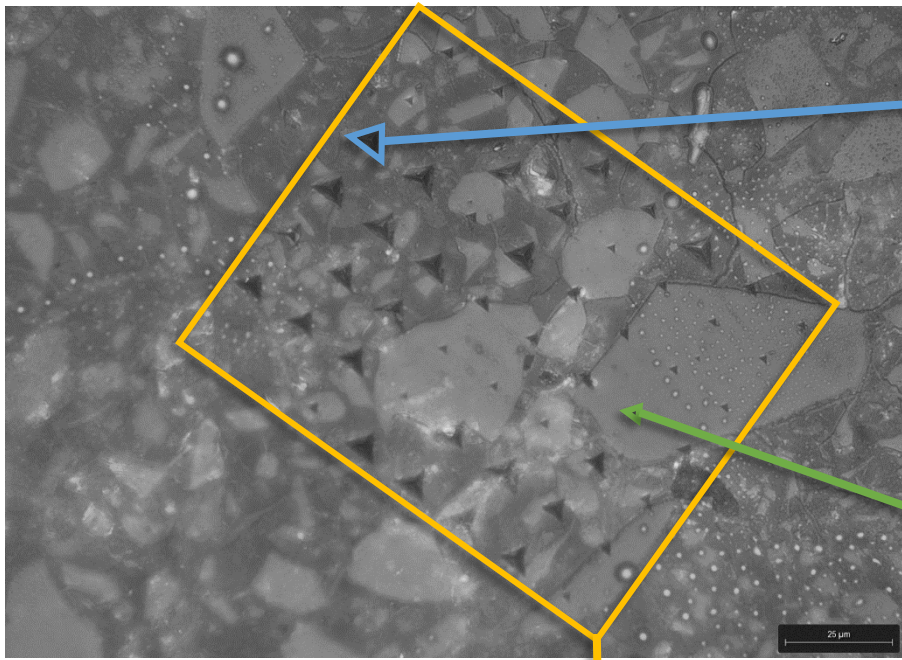
Effect of gamma irradiation on Fe-rich inorganic polymers

Increase in compressive strength > 5 kGy



Effect of gamma irradiation on Fe-rich inorganic polymers

Nanoindentation : methodology



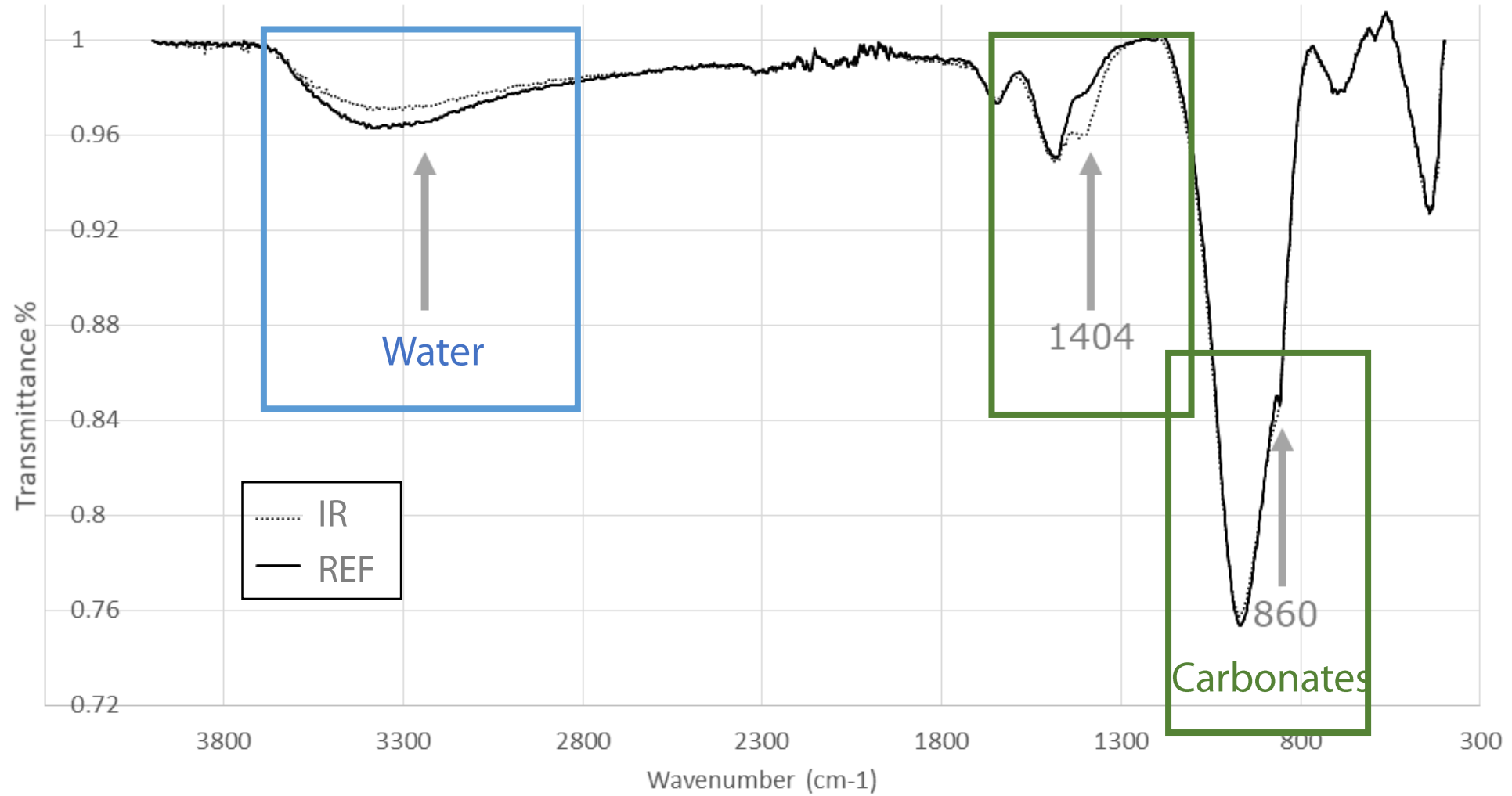
- Hardness
- Young's modulus
- Creep

Nanoindentation: effects on the binder

(1h) – 10 kGy/h

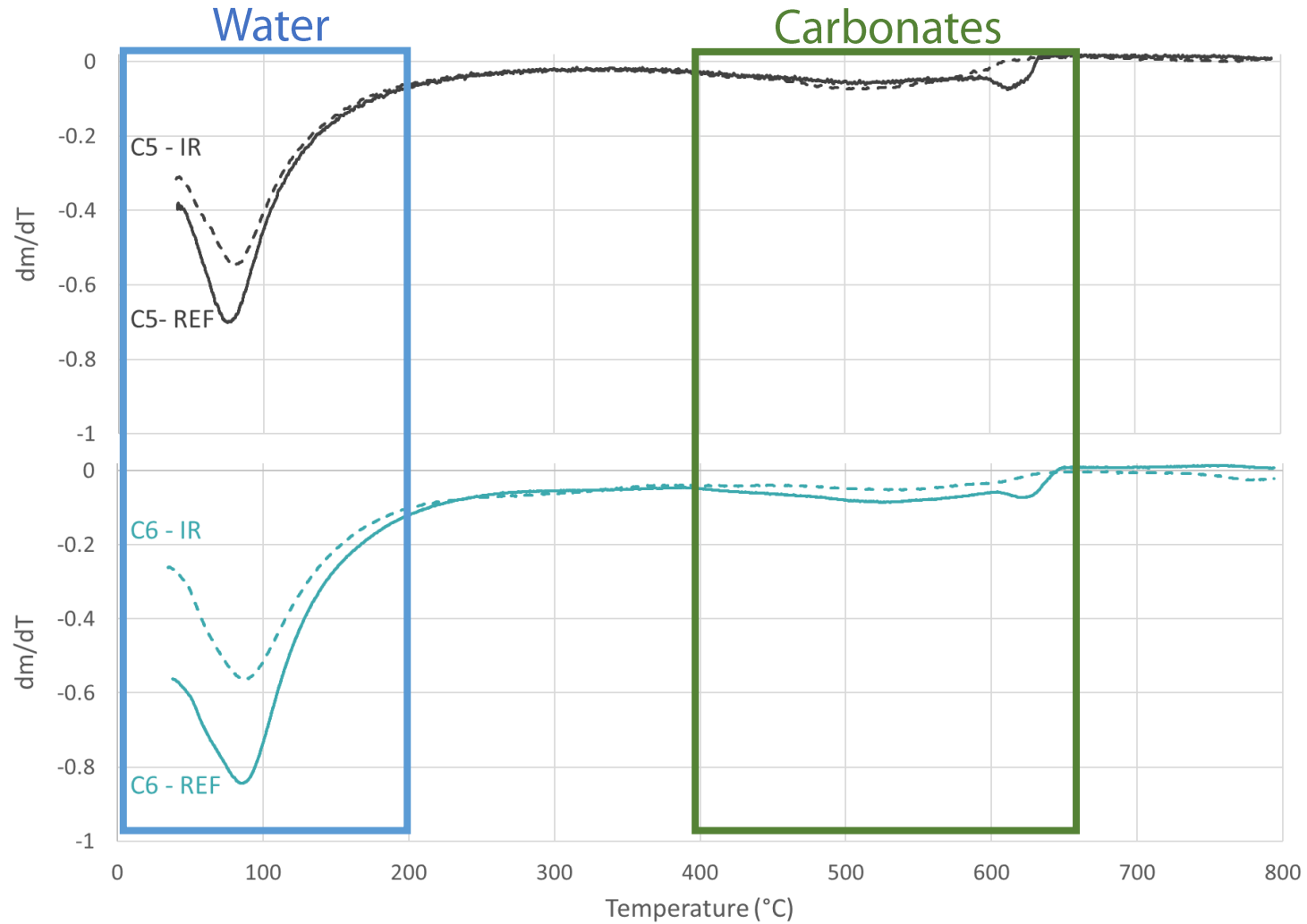
- Hardness ↓
- Young's modulus ↓
- Creep ↓

FTIR: radiation-induced carbonation + dehydration (1h) – 152 Gy/h



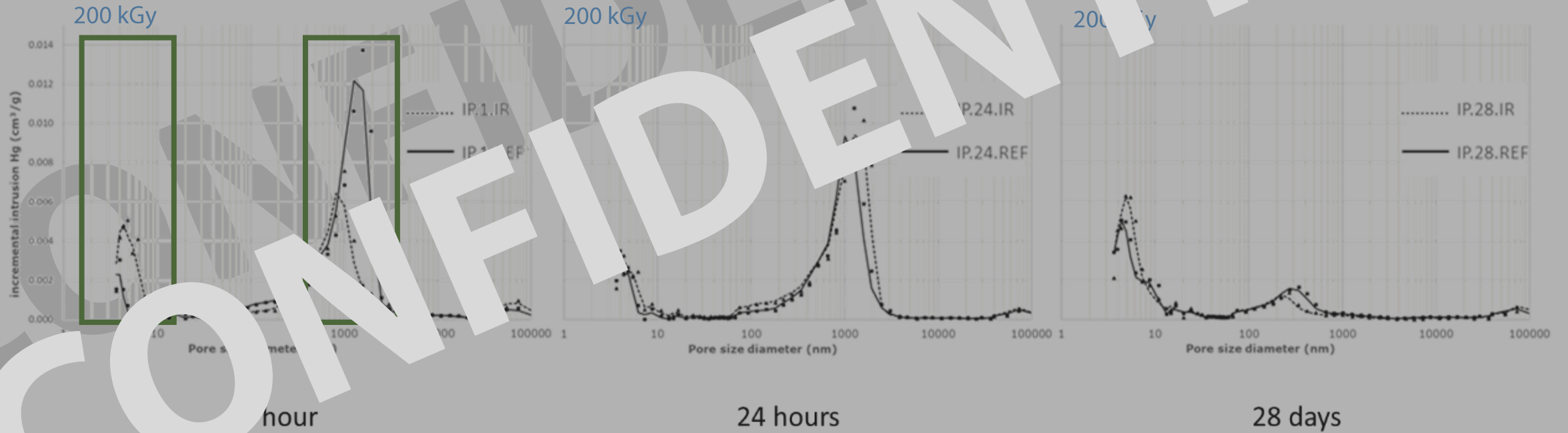
TGA: radiation-induced carbonation + dehydration

(1h) – 152 Gy/h

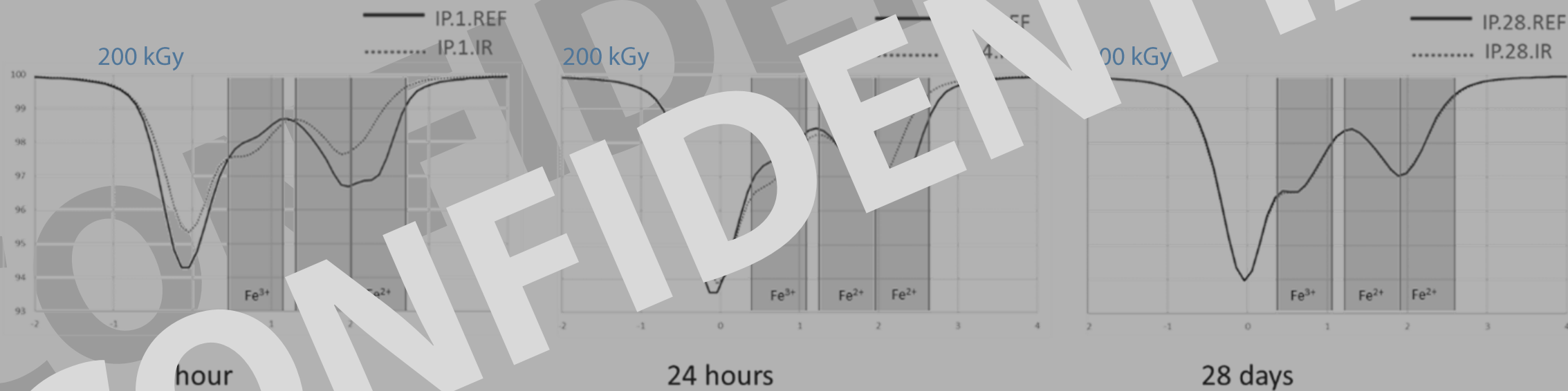


Effect of gamma irradiation on Fe-rich inorganic polymers

- Decrease in porosity
- Shift to the smaller pore sizes



Effect of gamma irradiation on Fe-rich inorganic polymers



Fe^{3+}_{REF} : 20 %
 Fe^{3+}_{IR} : 39 %

Fe^{3+}_{REF} : 24 %
 Fe^{3+}_{IR} : 38 %

Fe^{3+}_{REF} : 43 %
 Fe^{3+}_{IR} : 43 %



Summary

Radiation-induced:

- Carbonation
- Dehydration
- Iron oxidation

Leading to:

- Decrease in porosity
- Decrease in E-modulus
- Increase in Fe³⁺

Resulting in:

- Increased strength

Contact



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