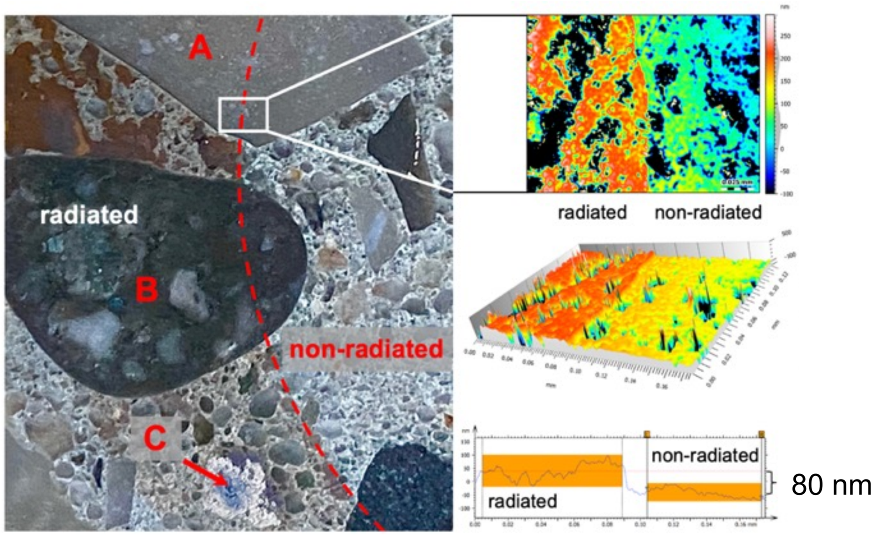
Examining out-of-plane expansion of aggregate minerals in ion-irradiated concrete



Q. I. Roode-Gutzmer, S. Schymura, A. Barkleit, T. Stumpf

Bundesministerium
für Bildung
und Forschung

FORKA – Forschung
für den Rückbau
kerntechnischer Anlagen

Förderkonzept: Rückbau und Entsorgung

9. RCA Workshop

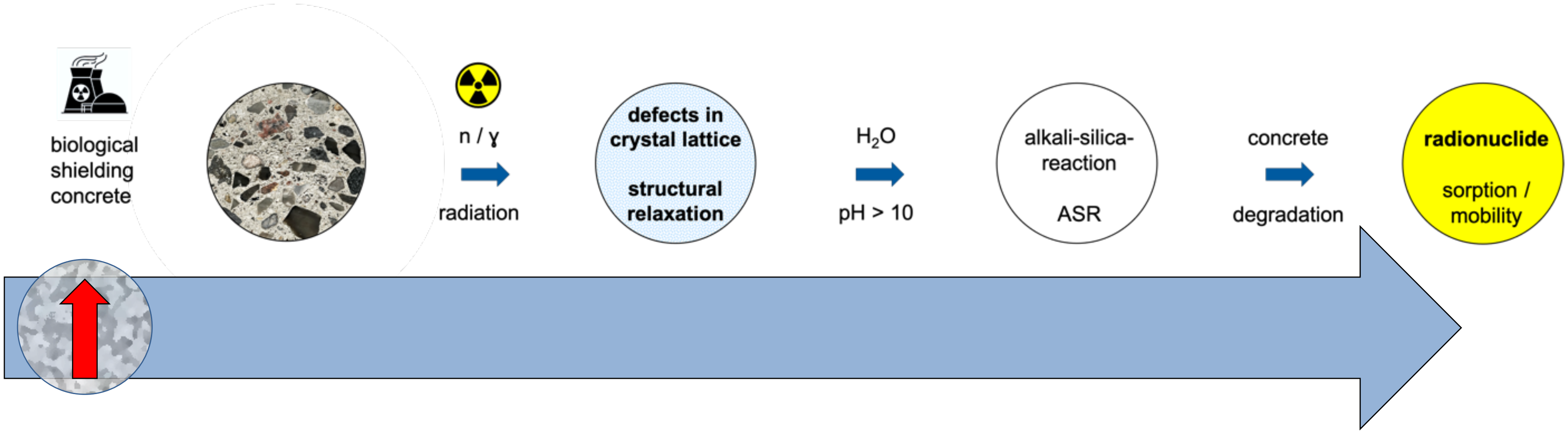
June 2021

Partner von
DRESDEN
concept
exzellenz-dresden.de

HZDR

HELMHOLTZ
ZENTRUM DRESDEN
ROSSENDORF

Project details and funding



PhD work conducted within the framework of the WERREBA project (Grant Number 15S9412)

- funded by the German Federal Ministry of Research and Education
- within the FORKA initiative

WERREBA Project

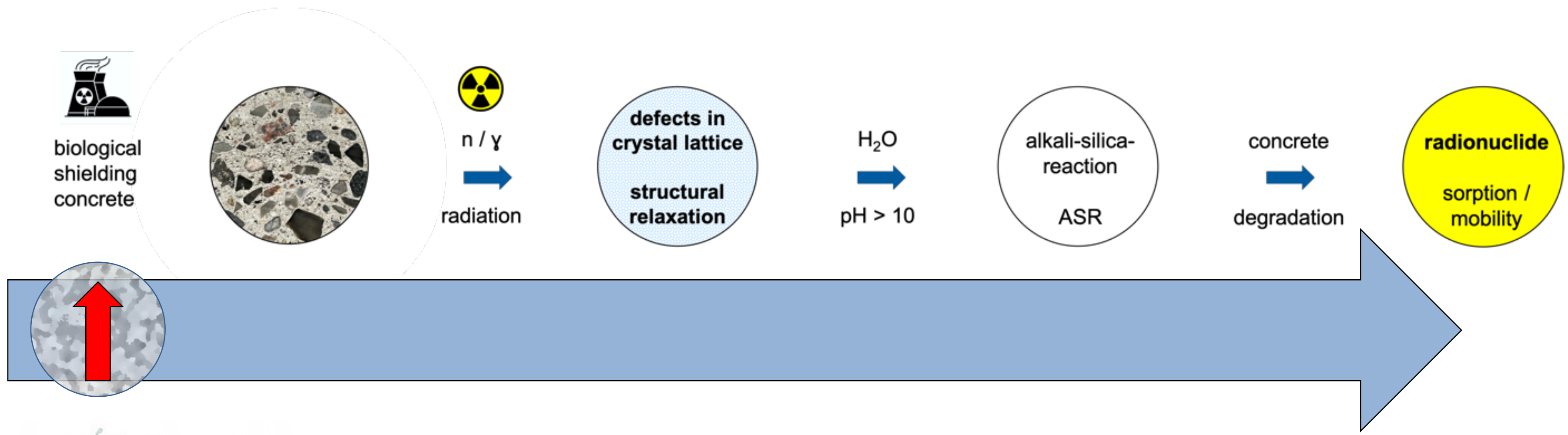
Primary objective:

- spatially reconcile calculated activity values with measured values of radionuclides contained in steel and concrete from the dismantled NPP Lubmin in Greifswald, Germany

Secondary objective:

- to determine where and how the radionuclides are bound in the concrete
- to determine the mobility of radionuclides under various leaching conditions

Nuclear power plant Lubmin, Greifswald

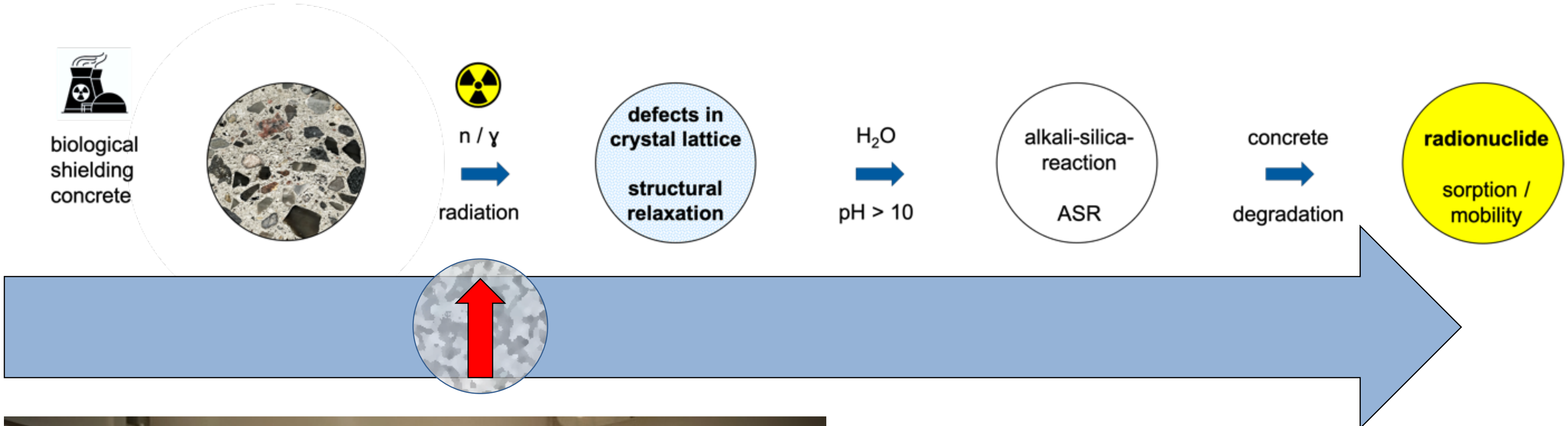


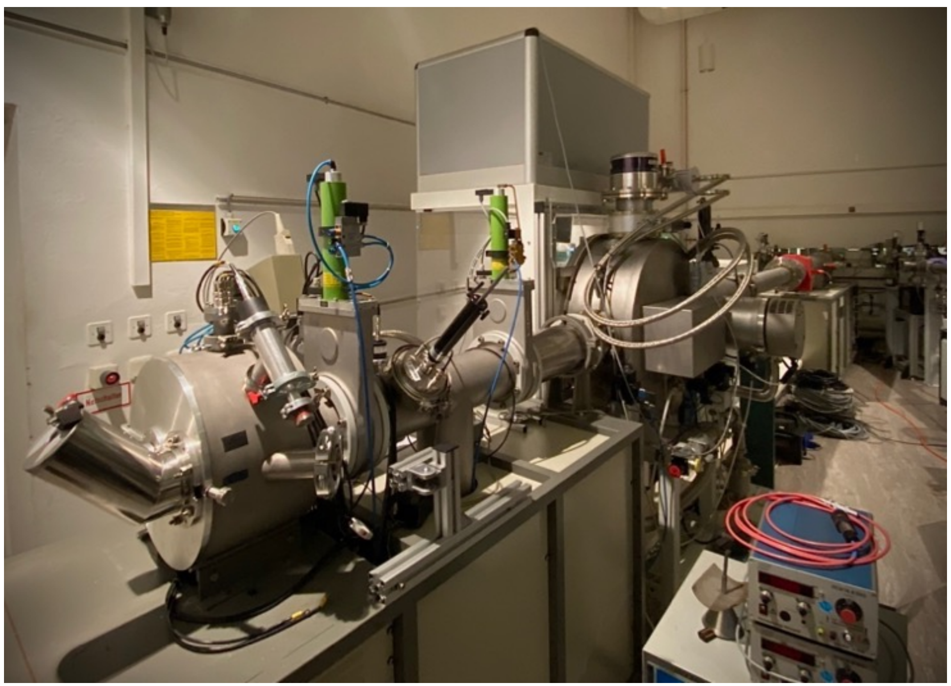
Empty reactor block 3, nuclear power plant, Lubmin, Greifswald. Decommissioned 1990.

Image credit: picture alliance/ZB/bwu pzi, 2015.

Data and image: BfE, 2015.

Simulating neutron radiation damage to concrete with an ion beam





Ion-Beam Centre (HZDR)

Advantages of ion-beam radiation

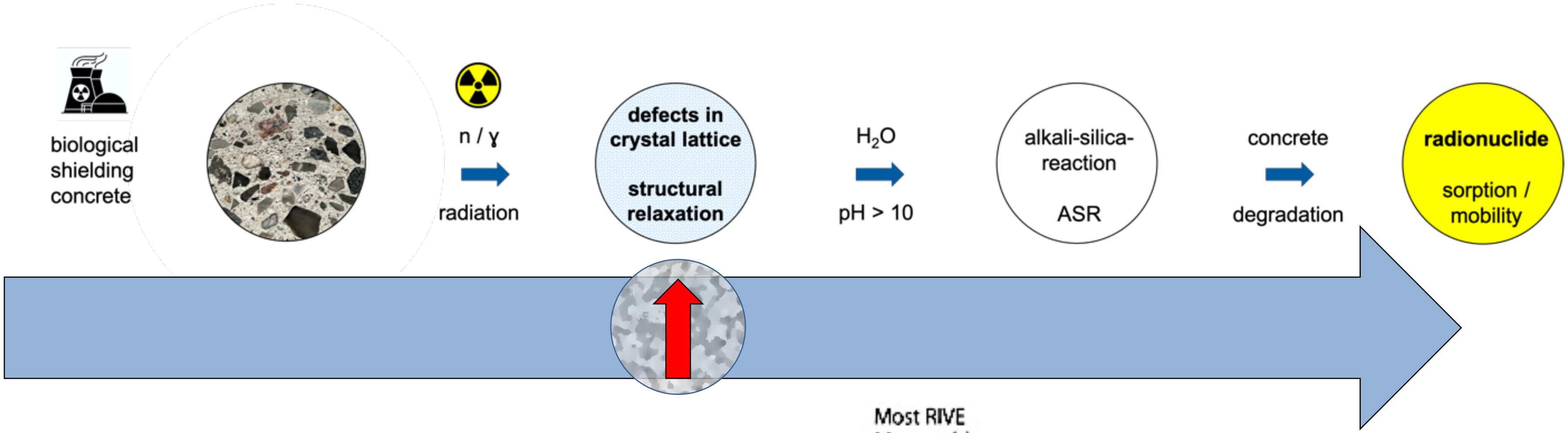
- locally accessible technology
- samples not radioactive
  no requirement for controlled area laboratory
- neutron radiation damage over decades
  can be simulated within hours

Si-ion irradiation
fluence of $5 \cdot 10^{14}$ ions/cm^2
at 300 keV

≈ 40 years
NPP operation

Quirina Isabella Roode-Gutzmer | Institute of Resource Ecology | www.hzdr.de

Neutron radiation damage to rock-forming minerals

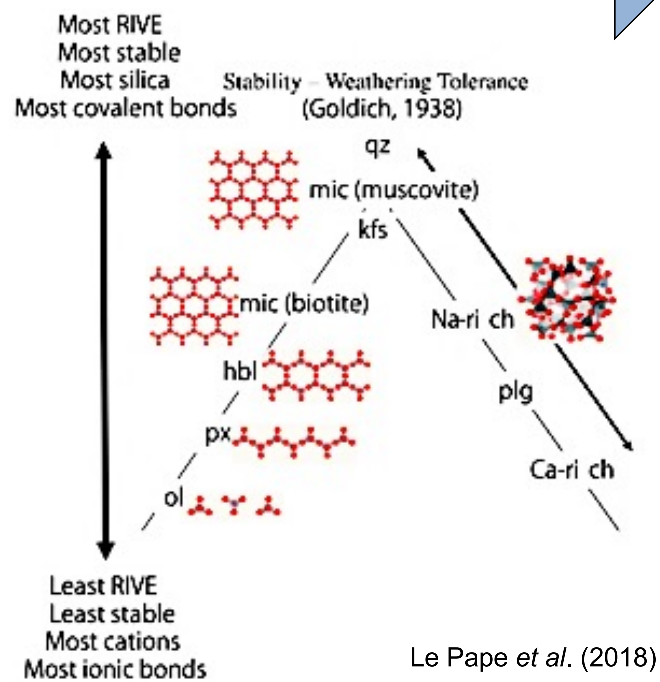


Radiation introduces defects in quartz structure

- when critical concentration of defects reached structure relaxes
  → amorphization
  → expansion
  → more reactive

Radiation Induced Volumetric Expansion (RIVE)
maximum RIVES for silicates (Le Pape *et al.*):

Quartz	17.8 %
Potassium feldspar	7.7 %



Le Pape *et al.* (2018)

Le Pape *et al. J. Adv. Conc. Technol.* **16 (5)** 191-209 (2018)

Akali silica reaction in concrete

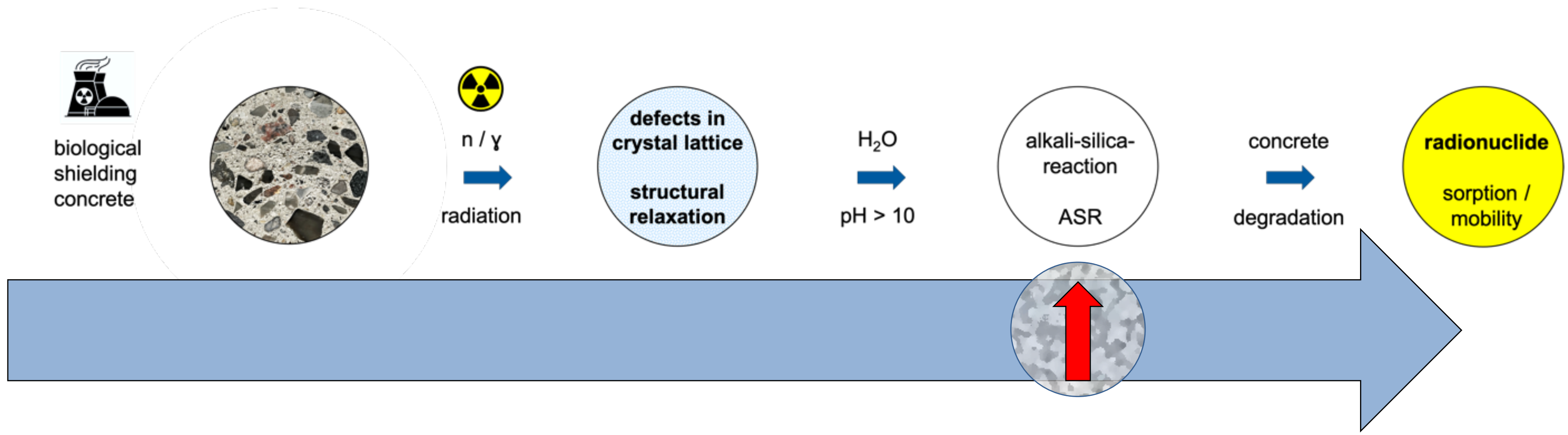


Image credit: Shinkolobwe, pillar at the National Gallery of Canada, 2017.

alkali silica reaction in concrete – also known as concrete cancer

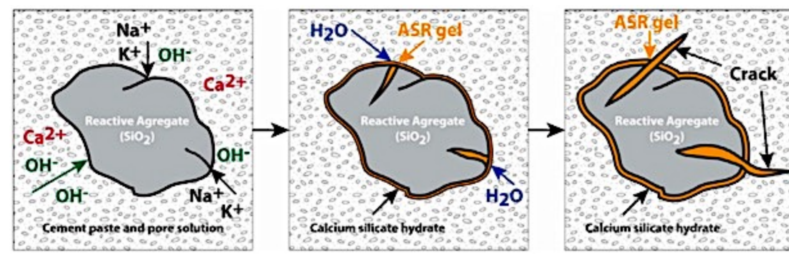
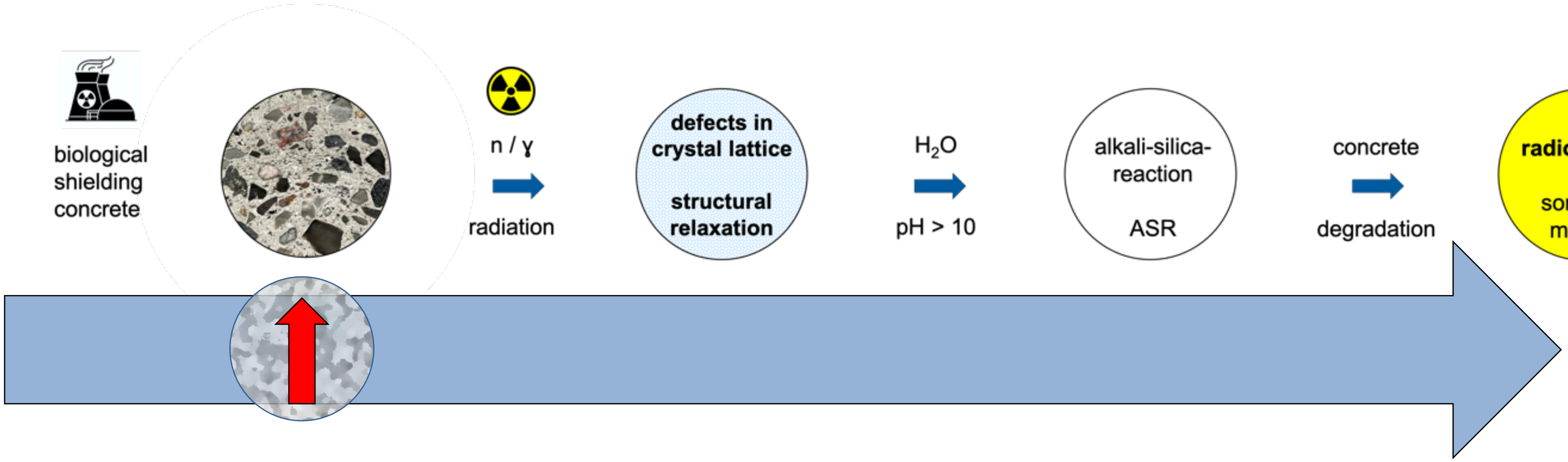


Image: Figueira *et al. Constr. Building Materials* **222** 903-931 (2019)

For ASR in concrete we require:

- ingress of water
- presence of alkalis (Na/K)
- reactive silicates (that fail the Oberholster test)

Concrete reference material



Concrete obtained during dismantling of a decommissioned NPP in Hamburg (kindly provided from DeBeKon project)

Mineral phase	Wt. %
α-Quartz (SiO_2)	51.0
Potassium feldspar microcline ($KAlSi_3O_8$)	17.8
Amorphous: C-S-H	25.6
Portlandite: $Ca(OH)_2$	2.8
Calcite: $CaCO_3$	2.8

Composition information provided from DeBeKon project

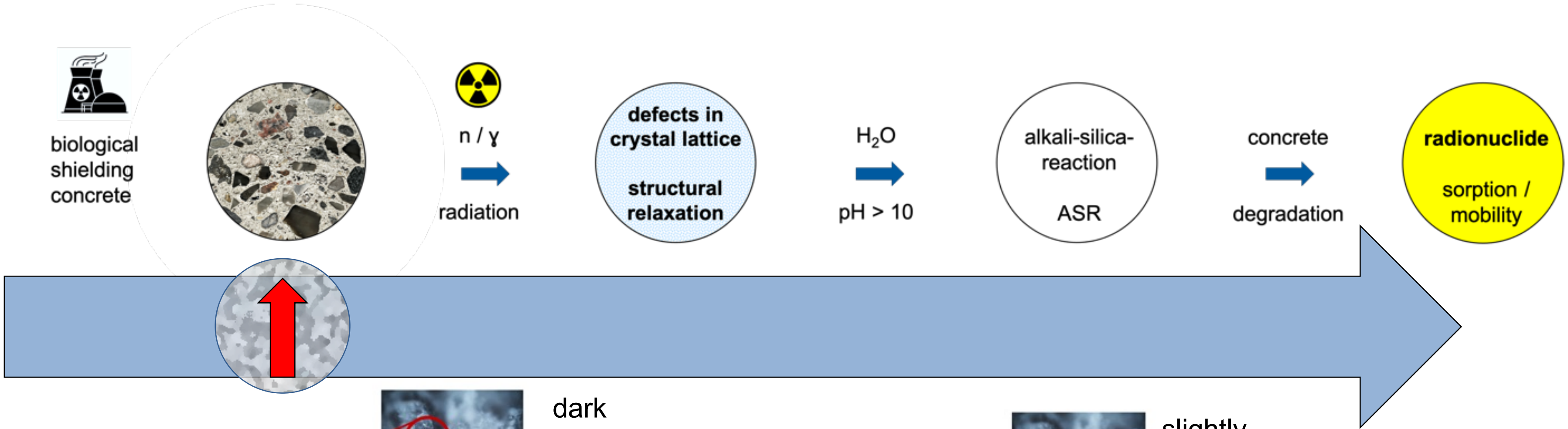
Phase identification by X-ray powder diffraction

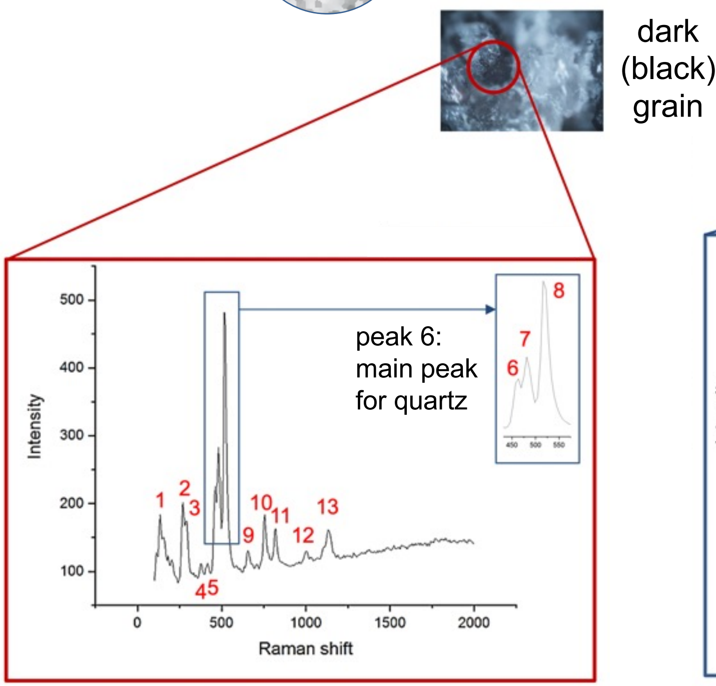
Quantitative phase composition determined by Rietveld refinement of XRD data

- new technique for calculating the amorphous content (Qoku *et al.*)
- with mass balancing using quantitative chemical composition

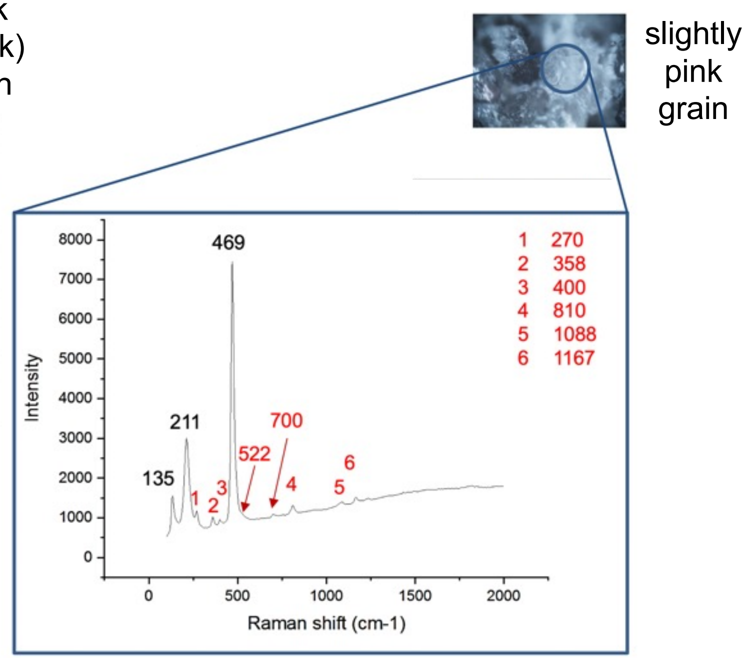
Qoku *et al. J. Build. Eng.* **12** 37-50 (2017)

Spatially resolved mineral characterisation using µ-Raman spectroscopy

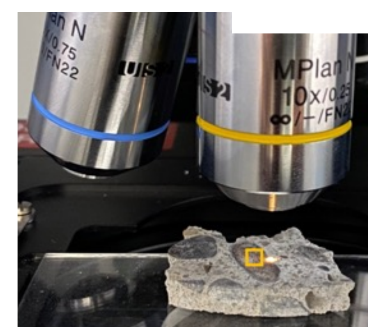




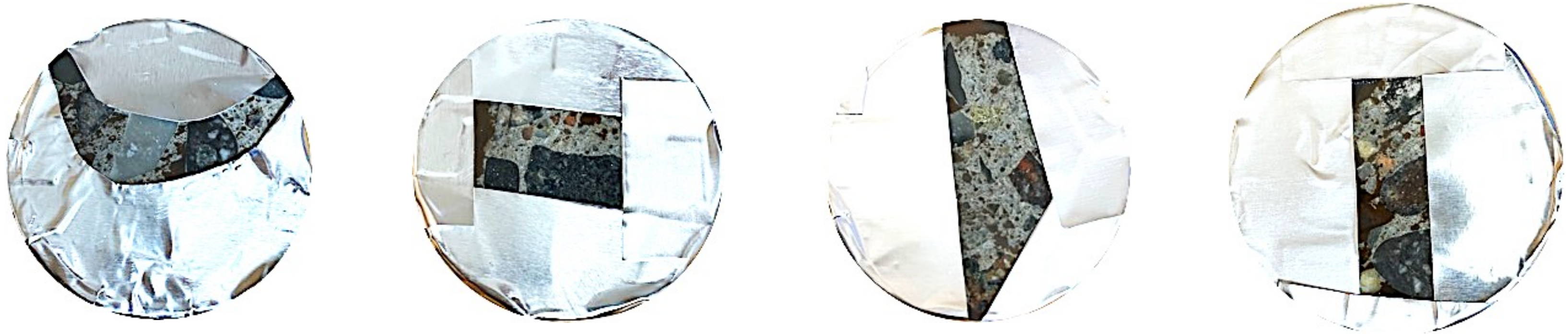
Microcline – $KAlSi_3O_8$



Quartz – SiO_2



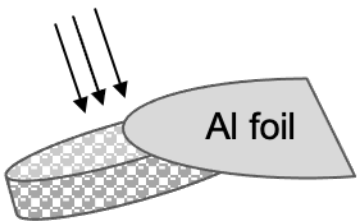
Ion irradiation of concrete – irradiation conditions – sample presentation



Si ions 5 · 10^{14}

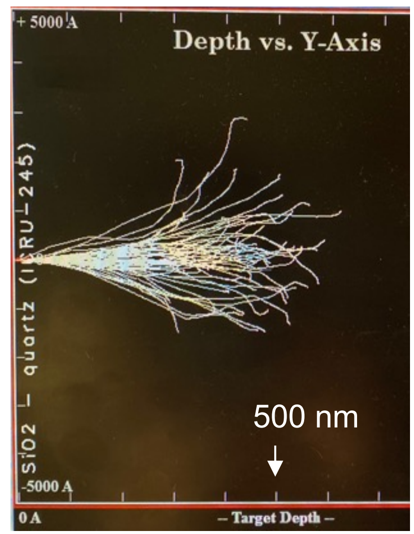
300 keV ions/cm^2

Simulating neutron radiation damage of 40 y of nuclear power plant operation



polished sections
vacuum impregnation
concrete (48 y)
non-radioactive

ion irradiation in vacuum
ambient temperature
without cooling



Stopping and Range of Ions in Matter (SRIM)

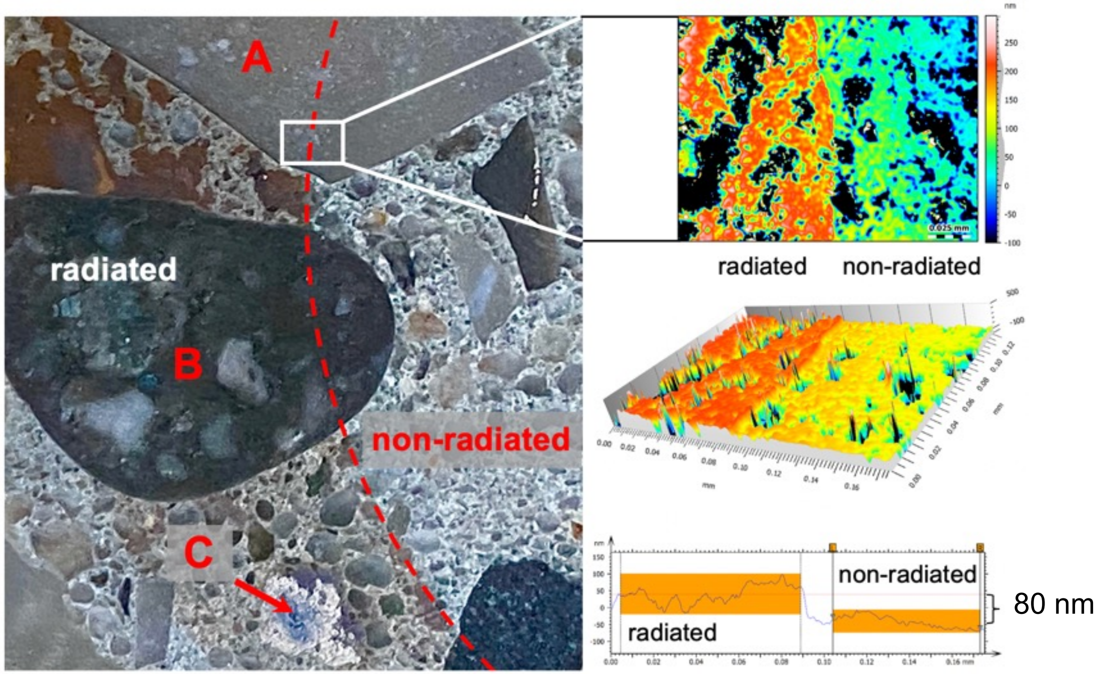
SRIM (Ziegler *et al.*)
Kinchin–Pease calculation

depth penetration expected for Si-ions of 300 keV

Quartz: 429 nm
Microcline: 604 nm

Ziegler *et al. J. Nucl. Instrum. Methods Phys. Res*. **B268** 1818-1823 (2010) http://www.SRIM.org

Ion irradiation in concrete – pilot study



Radiated area left of red dashed line.

A – quartz (SiO_2)

B – potassium feldspar

(microcline: $KAlSi_3O_8$)

intergrown with quartz

C – sputtering from Si-ion irradiation

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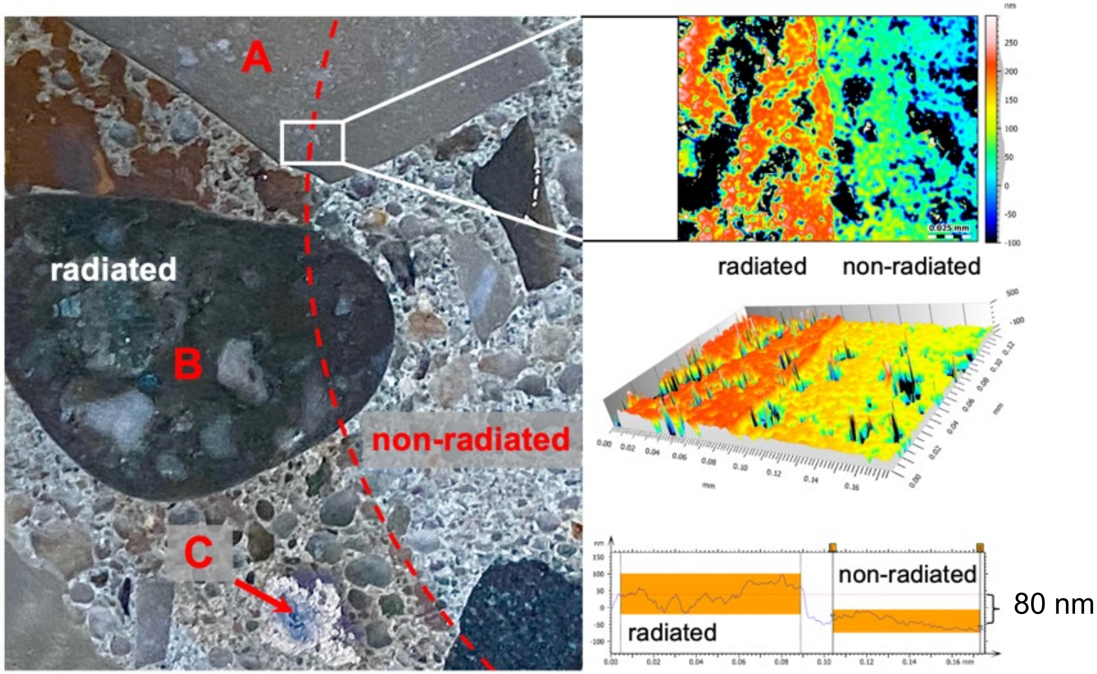

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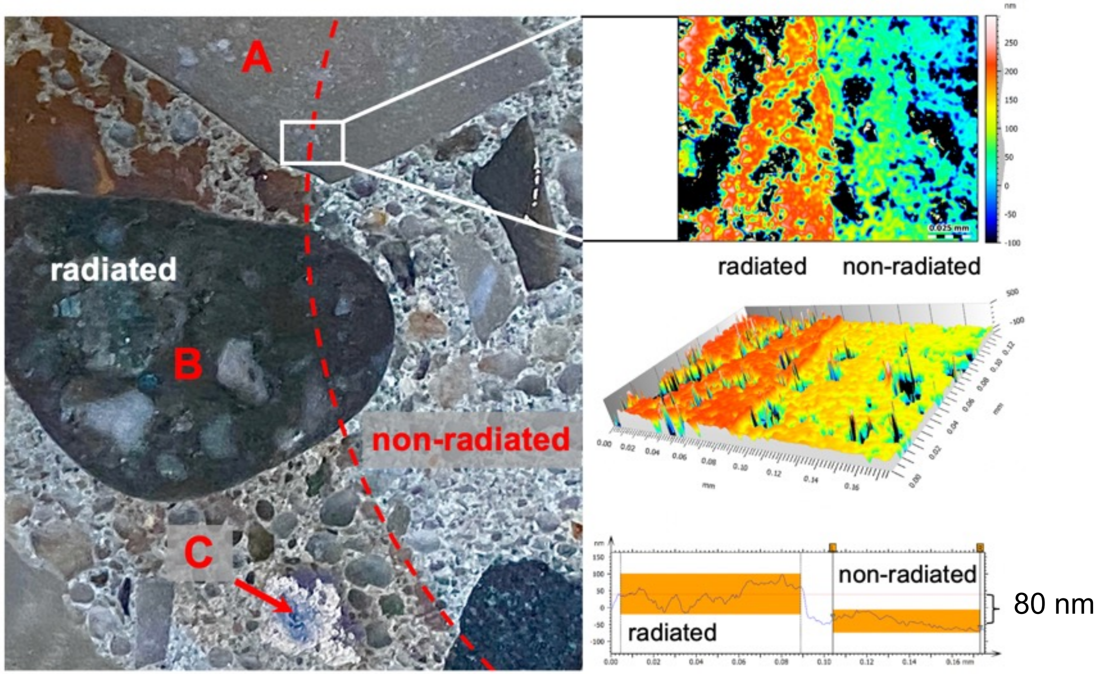
Vertical Scanning Interferometry reveals:

- out-of-plane expansion
  for quartz of + 80 nm
- ablation
  of epoxy resin of - 600 nm
- surface of aggregate B too rough
  height changes cannot be observed

Ion irradiation in concrete – pilot study



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Quartz

ΔV_{obs} = 18.8 % (this work)

ΔV_{obs} = 18.1 % (Luu *et al.*)

At max neutron fluence:

ΔV_{max} = 17.8 % (Le Pape *et al.*)

Calculated density change (Luu *et al.*)

$$\rho/\rho_0 \approx \frac{1/(R+\ \Delta H)}{1/R}$$

ΔH step height

R ion range determined by SRIM

Luu *et al. J. Nucl. Mat.* **545** 152734 (2021)

Le Pape *et al. J. Adv. Conc. Technol.* **16 (5)** 191-209 (2018)

Ziegler *et al. J. Nucl. Instrum. Methods Phys. Res.* **B268** 1818-1823 (2010) http://www.SRIM.org

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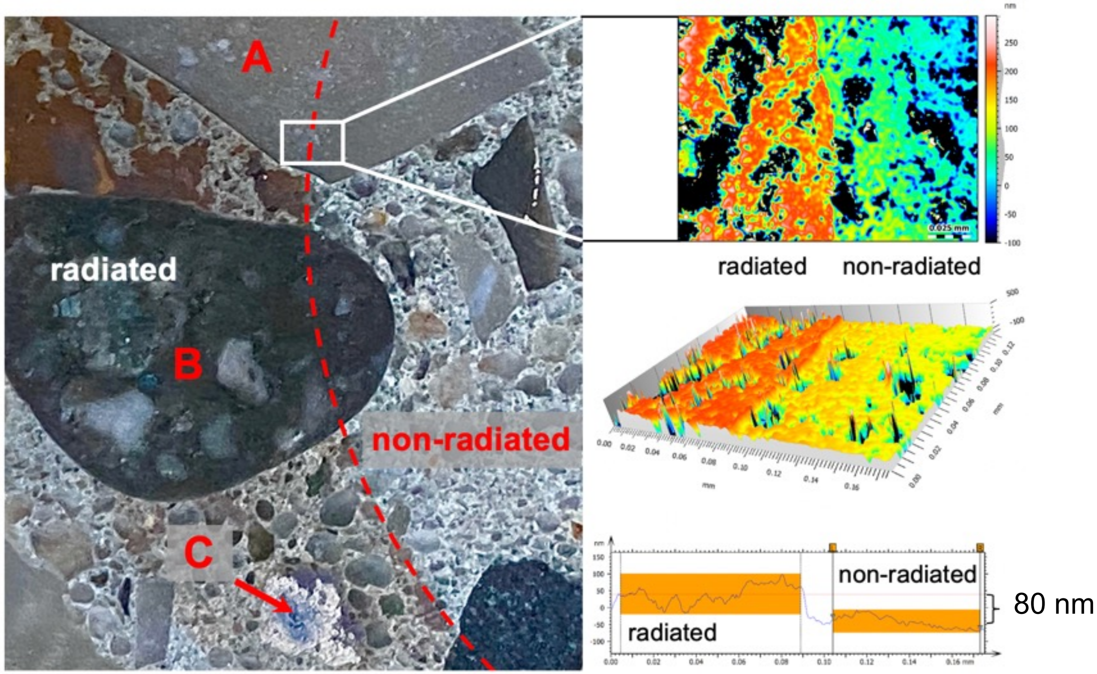

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Quartz

ΔV_{obs} = 18.8 % (this work) → $5\cdot10^{14}$ Si ions/cm^2 at 300 keV

ΔV_{obs} = 18.1 % (Luu *et al.*) → $6\cdot10^{15}$ Si ions/cm^2 at 3000 keV

At max neutron fluence:

ΔV_{max} = 17.8 % (Le Pape *et al.*)

Luu *et al. J. Nucl. Mat.* **545** 152734 (2021)

Le Pape *et al. J. Adv. Conc. Technol.* **16 (5)** 191-209 (2018)

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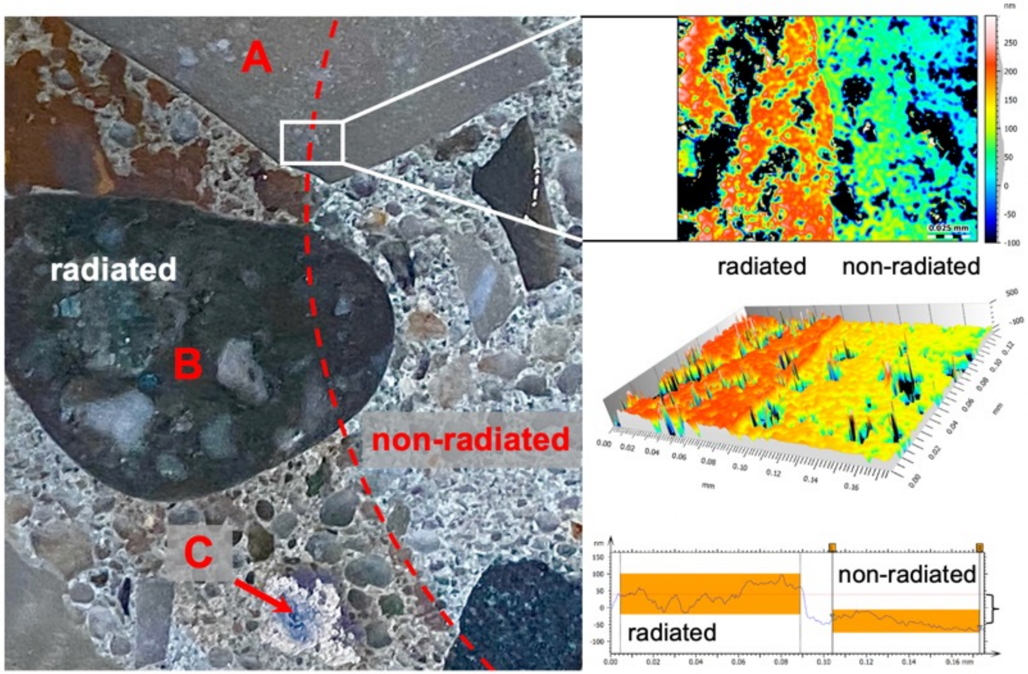

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(microcline: $KAlSi_3O_8$)
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  for quartz of + 80 nm
- ablation
  of epoxy resin of - 600 nm
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  height changes cannot be observed

Quartz

ΔV_{obs} = 18.8 % (this work) — 430 nm → $5 \cdot 10^{14}$ Si ions/cm^2 at 300 keV

ΔV_{obs} = 18.1 % (Luu *et al.*) — 2000 nm → $6 \cdot 10^{15}$ Si ions/cm^2 at 3000 keV

At max neutron fluence:

ΔV_{max} = 17.8 % (Le Pape *et al.*)

Luu *et al. J. Nucl. Mat.* **545** 152734 (2021)

Le Pape *et al. J. Adv. Conc. Technol.* **16 (5)** 191-209 (2018)

Ziegler *et al. J. Nucl. Instrum. Methods Phys. Res.* **B268** 1818-1823 (2010) http://www.SRIM.org

Conclusions

- maximum volume expansion of quartz under selected ion irradiation conditions was achieved

Outlook

- standard application for beam-time at IBC / same ion fluence and energy

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- surface of feldspar-containing grains too rough to measure height changes by VSI
- expected volume expansion for microcline / better specimen polishing
  Luu et al. 5.8 %
  Le Pape et al. 7.7 %

Outlook

- standard application for beam-time at IBC / same ion fluence and energy
- surface polishing / low energy oblique incidence Argon broadband ion beam (Ar-BIB)
- mono-phase minerals

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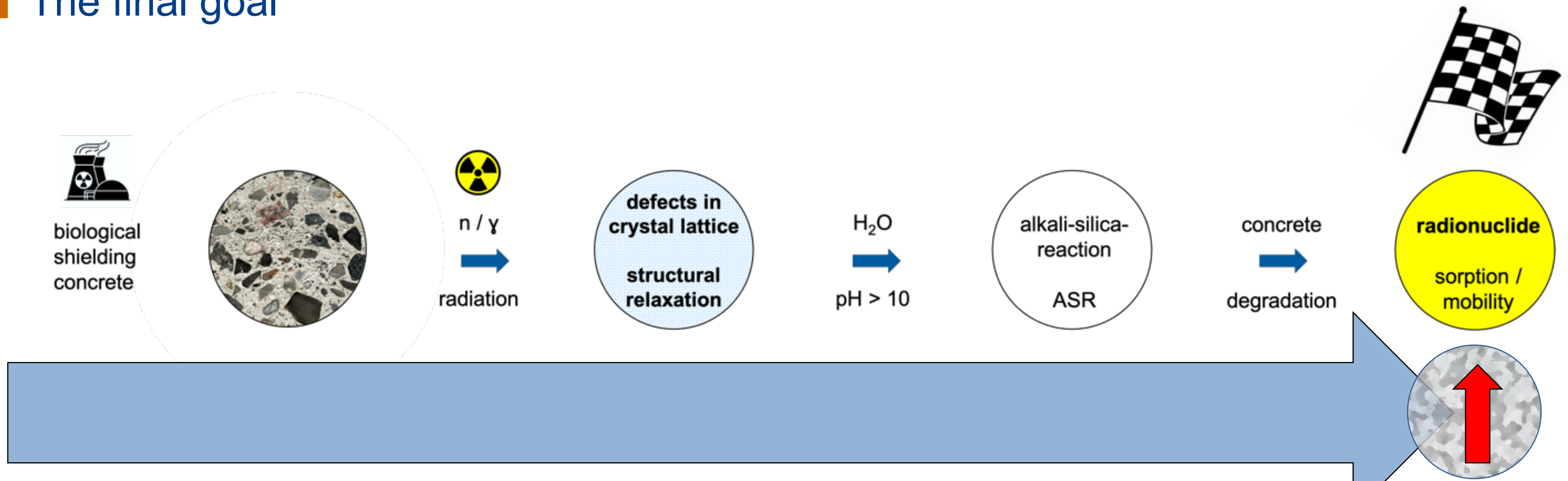
Outlook

- standard application for beam-time at IBC / same ion fluence and energy
- surface polishing / low energy oblique incidence Argon broadband ion beam (Ar-BIB)
- mono-phase minerals
- radiation damage as a function of temperature (< 300 °C)
- structural changes / electron backscatter diffraction (EBSD) and FIB-SEM
- *in situ* dissolution studies / VSI and SEM

The final goal



Non-radioactive samples / enabled the evaluation of sample preparation and measurement logistics

- pore structure evaluation for reactive transport investigated
- ion-irradiation studies (pilot study completed / beamtime application submitted)
- silicate dissolution kinetics (in progress)

Radioactive samples / controlled area laboratories

- concrete and aggregate minerals / neutron irradiated at various NPPs (*via* EMPRADO project)
- concrete drill cores from Greifswald (arrives 15 June 2021)
- radionuclide inventory
- radiation damage to mineral phases

❖ Mobility of radionuclides in concrete
under various leaching environments

Acknowledgments

Federal Ministry of Education and Research

Concrete reference material

Sample provided from DeBeKon project
by Hans-Jürgen Friedrich (Fraunhofer Institute for Ceramic Technologies and Systems, IKTS)

Quantitative phase composition:
Elsa Qoku (TUBA Freiberg, Institute of Ceramics, Refractories and Composite Materials)

Sample preparation

Polished sections: Ina Noack (TU Dresden, Institute of Construction Materials)

Ion beam radiation

Shavkat Akhmadaliev (HZDR, Ion Beam Centre)

Preliminary VSI assessment for sample preparation

Holger Lippold (HZDR, Institute of Resource Ecology, Leipzig)

PhD mentoring support

Nina Huittinen (HZDR, Institute of Resource Ecology, Rossendorf)
Cornelius Fischer (HZDR, Institute of Resource Ecology, Leipzig)

DRESDEN concept

HZDR