The effect of ion irradiation on amorphization and volume change in model materials of concrete aggregates

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A part of this study was the results from the "Japan Concrete Aging Management Program on Irradiation Effects (JCAMP)" sponsored by METI in Japan

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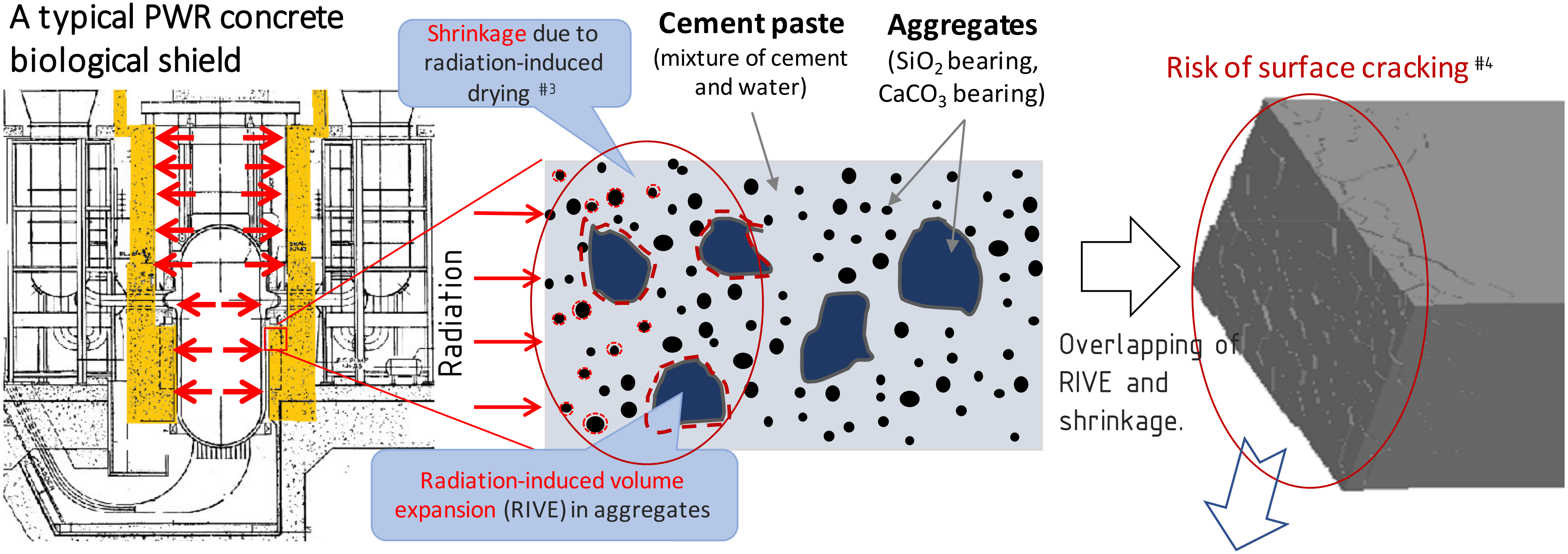
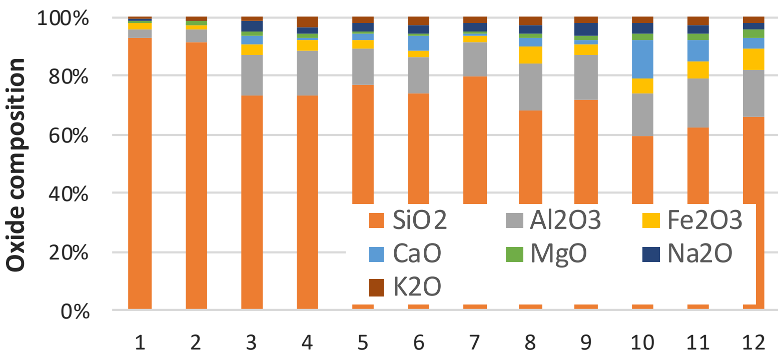
Introduction Degradation of concrete biological shield wall

A typical PWR concrete biological shield





Siliceous rock aggregates at different locations in Japan[#1]

Issues relating the degradation of concrete during long-term operation (LTO):

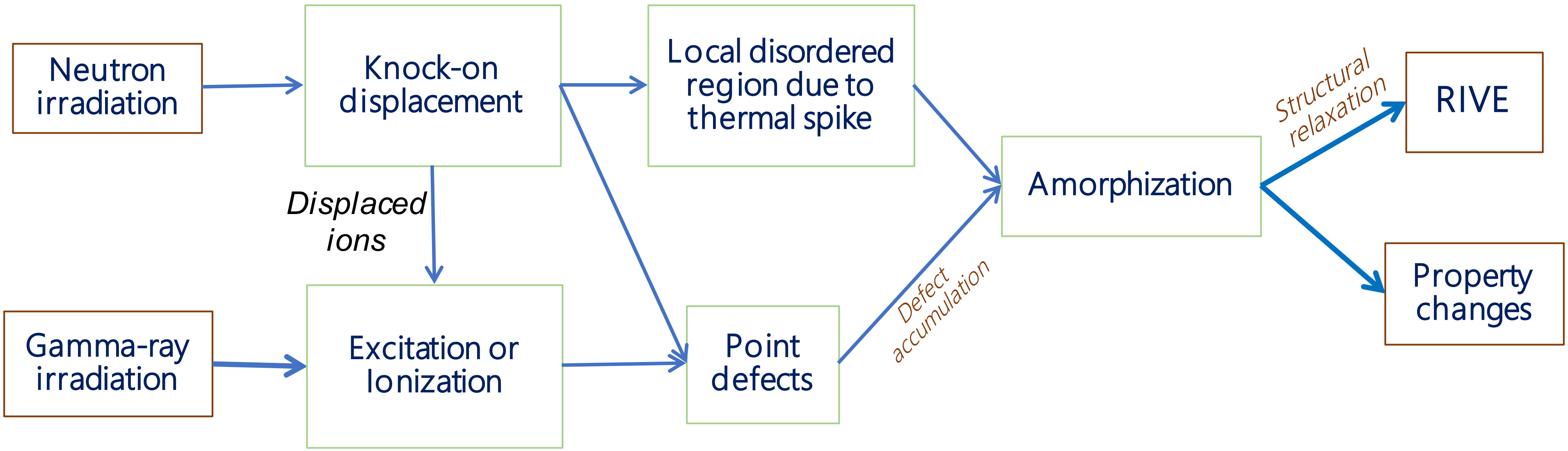
- ○ Diversity of mineral compositions
- ○ RIVE will occur during LTO?
- ○ What is maximum radiation level 10^{19} or 10^{20}(n/cm^2)?

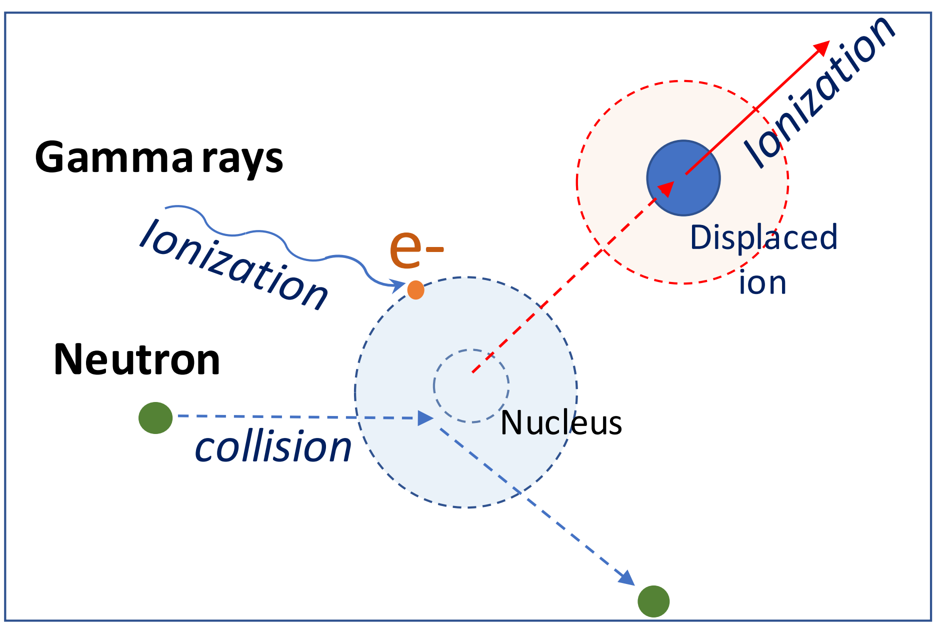
Current data remains high uncertainty due to diversity of neutron spectrum, test conditions, as well as aggregates used;

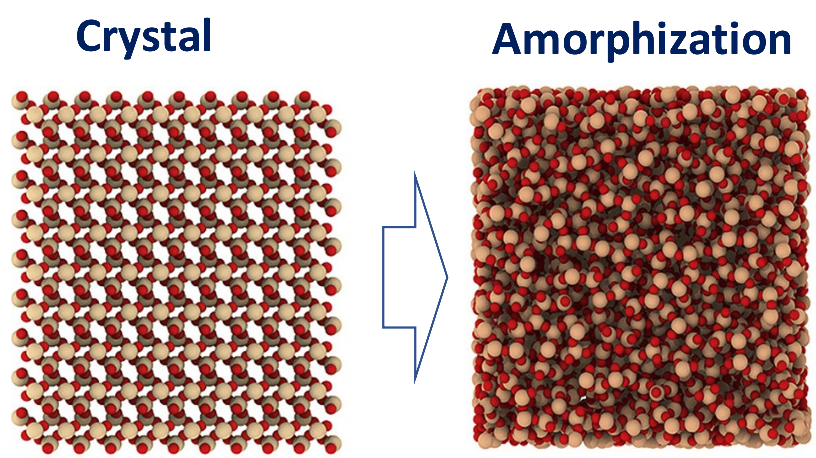
- Diversity of composition of siliceous rock aggregates [#1]
- Quartz (SiO_2) and aluminosilicate minerals (i.e., albite($NaAlSi_3O_8$), microcline($KAlSi_3O_8$)) are common minerals and have high susceptibility to amorphization [#2]

[#1] *G. Igarashi et al, Constr. Build. Mater (2015);* [#2] *Y. Le Pape et al, J. Adv. Concr. Technol (2018);* [#3] *Maruyama et al, J. Adv. Concr. Technol (2017);* [#4] *T.M. Rosseel et al, J. Adv. Concr. Technol (2016)*

Hypothesis on amorphization and RIVE







Krishnan et al., (2019)

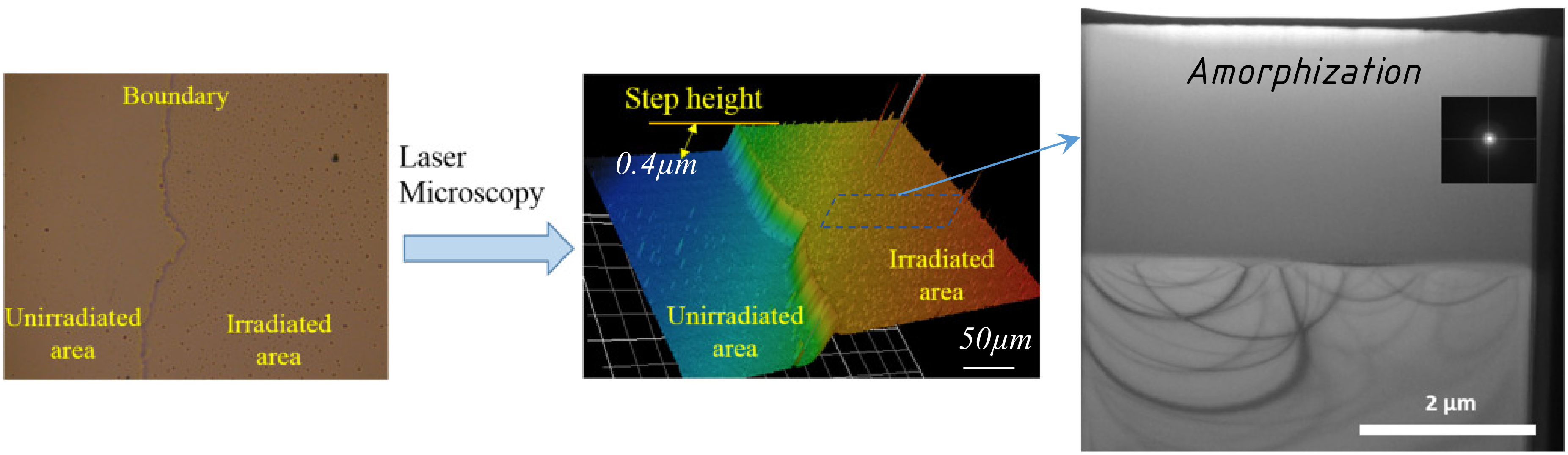
Research methodology

Crystalline samples → Ion irradiation using Tandem Acceleration, 3MeV Si ion → Microstructure / Change in properties / Evaluation of RIVE

- Microstructure: *TEM/FIB, In-situ TEM*
- Change in properties: *Nano-indentation (Hardness and Young's modulus)*
- Evaluation of RIVE: *Laser Microscopy, Profilometry*

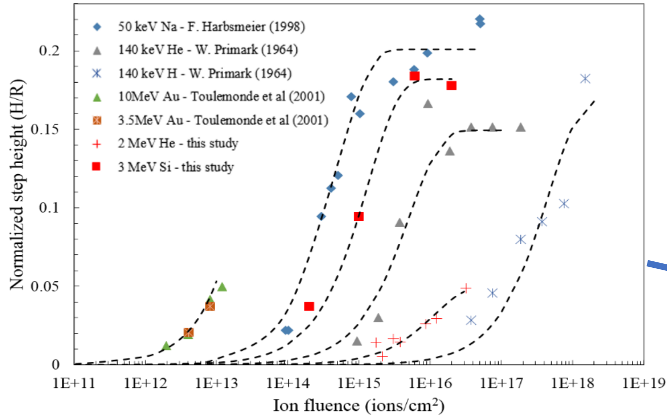
Quartz (SiO_2)
Albite ($NaAlSi_3O_8$)
Microcline ($KAlSi_3O_8$)

- ✓ *Irradiation at different temperature*
- ✓ *Max. irradiation dose is higher than amorphization level*



Result and Discussion: main cause of RIVE in case of quartz

Evaluation of RIVE in quartz

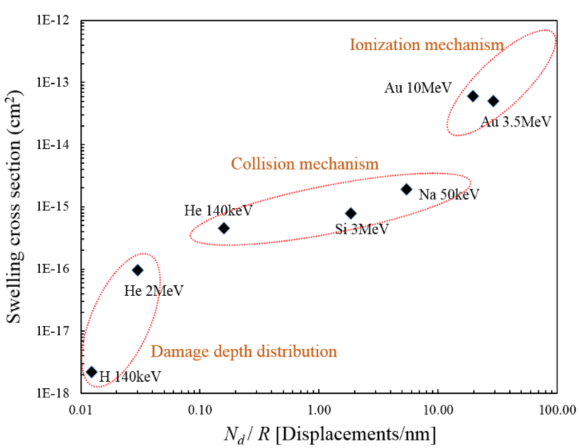


"Empirical model" for change of step height H using ion fluence Φ:

$$H = H_{\infty}[1 - exp(-\sigma_s \phi)]$$

Swelling cross section, σ_s (*the magnitude of the volume expansion due to a single radiation*)

$$\sigma_s = \sigma_{so}\, exp(-0.13/kT)$$

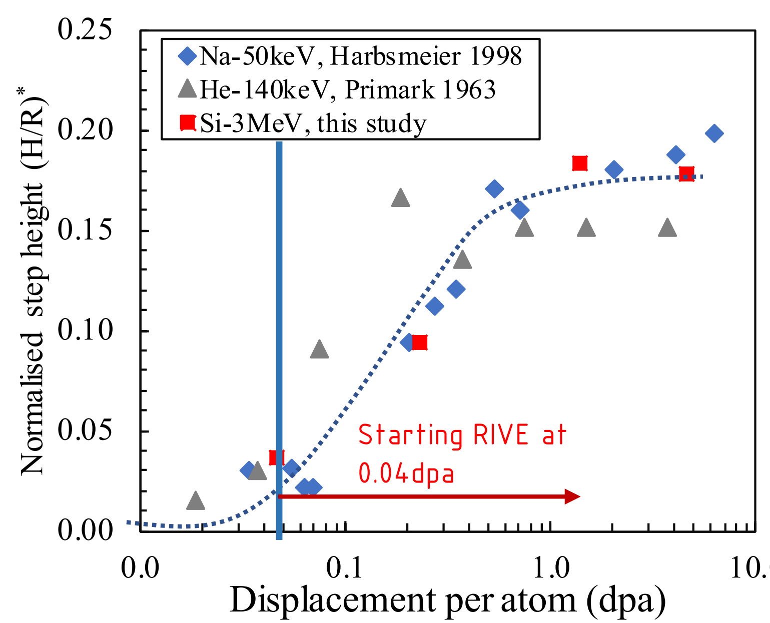


- Quartz exists in most of aggregates, easy to be amorphized, and highest RIVE, thus it can be used as an indicator of RIVE effects in concrete
- It is found that both knock-on and ionization process contribute to RIVE:

knock-ons *Ionization*

$$\sigma_s[10^{-14} cm^2] = \left[0.0072\left(\frac{N_d}{R}\right)\right] + \left[0.13\left(\frac{E_i}{R} - 1.6\right)\right]$$

- However, in neutron irradiation, kinetic energy by displaced ions is small (bellow 10^5 eV), thus RIVE is mainly related to number of displacement, dpa.
- Thus, the onset of RIVE can be determined as:



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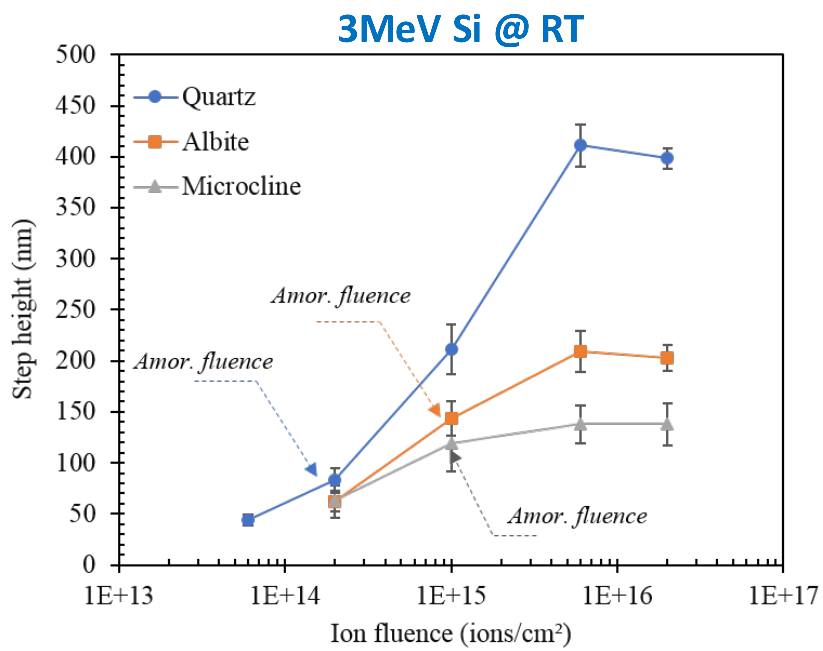
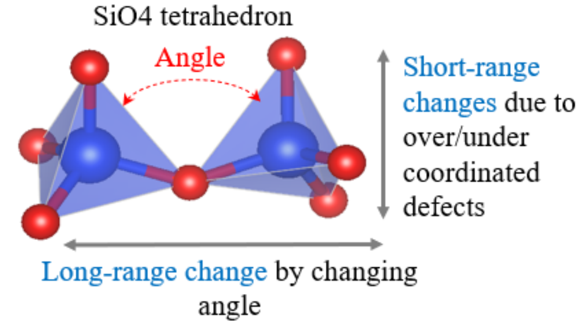
*Normalised step height is step height H divided by ion range R

Result and Discussion: RIVE mechanism in aluminosilicate minerals



RIVE is due to short range and long range change of tetrahedra



Quartz: RIVE continues after amorphization.

- Long-range and short-range change contribute to RIVE

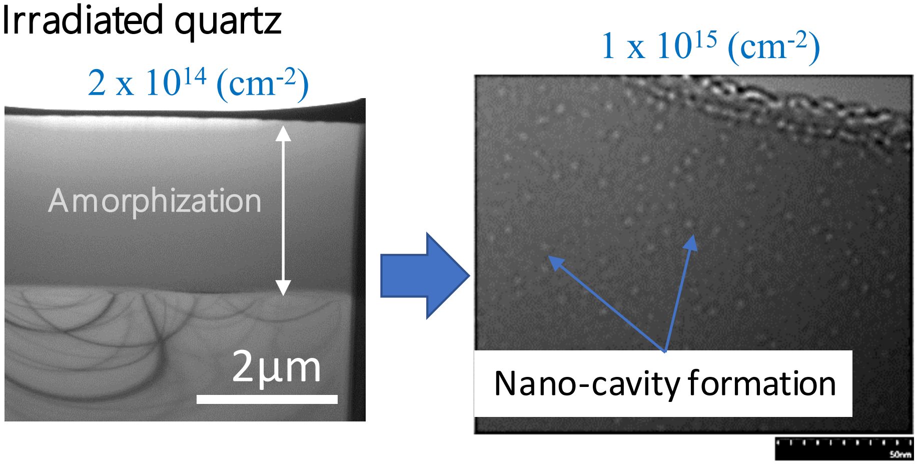
Albite: small additional RIVE

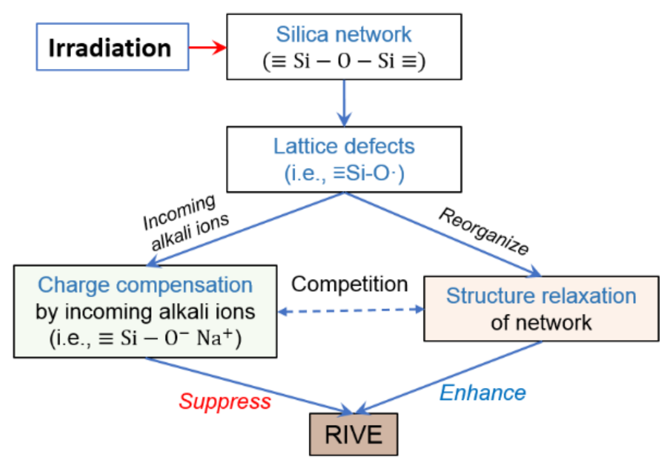
- Mainly long-range contribute to RIVE

Microcline: no additional RIVE

- Only short-range

Irradiated quartz





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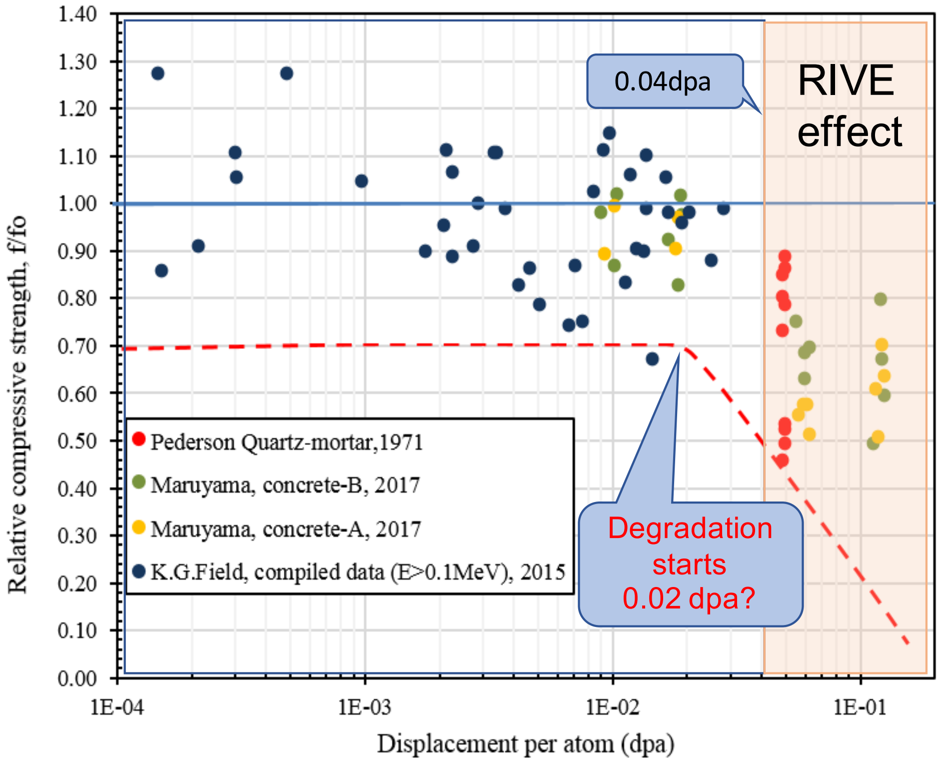
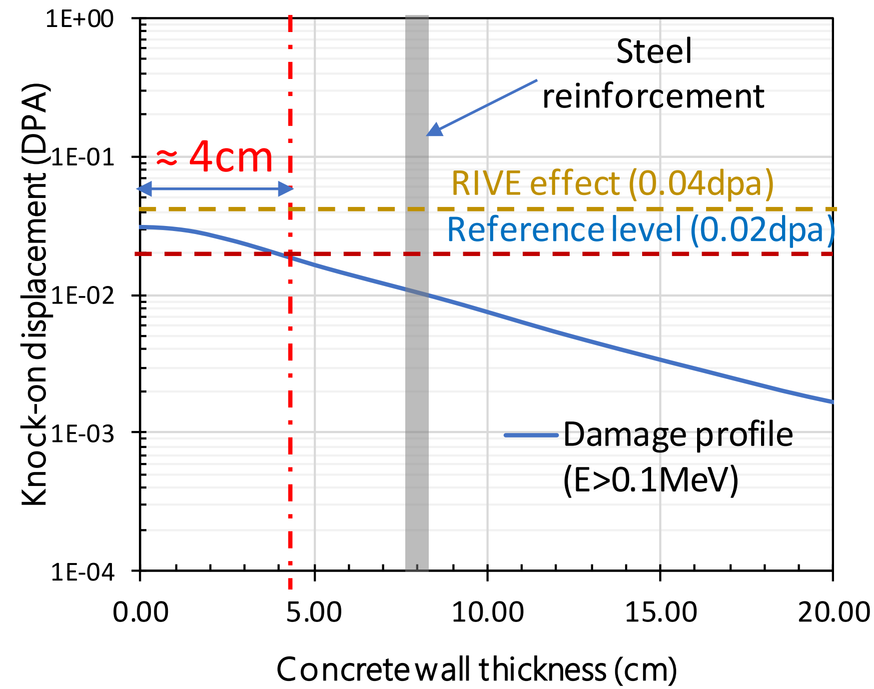
Result and Discussion: Reference level for concrete degradation

Indicator of degradation: reduction in (1) compressive strength, (2) shielding performance, and (3) seismic resistance.

Re-evaluate existing concrete data



Damage estimation after 60 years



- The RIVE in quartz starts at around 0.04dpa, but concrete's strength could reduce beyond 0.02dpa, this may be related to the radiation-induced drying effect.
- The estimated maximum damage in term of dpa for 60years is below 0.04dpa, suggesting RIVE will not occur during LTO. However, the degradation probably caused by drying is about 4cm in thickness.

Summary of results

- Though both ionization and knock-on process contribute to RIVE, number of knock-on atoms is found to be the main cause of RIVE in LWR condition.
- The mechanism of RIVE is found to be different in silicate minerals which is related to alkali contents (K^+, Na^+).
- A new reference level of 0.02dpa, which is considered the diversity of neutron spectrum, is proposed for concrete degradation

Significance of the study

- ✓ The degradation mechanism caused by irradiation in concrete during LTO was investigated.
- ✓ Importantly, in current modelling codes, RIVE is considered to be saturated at amorphization, but we found RIVE may continue after amorphization. The obtained property changes are important input for meso-scale modelling code to evaluate the performance of concrete during LTO.

Thank you very much for your attention!

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