Department of Architecture
School of Engineering
The University of Tokyo
Building Material Laboratory

Summary of recent progress of JCAMP and Hamaoka project

Ippei Maruyama*1

Toshiaki Kondo *2, Shohei Sawada*2, Osamu Kontani*2,
Kiyoteru Suzuki*3, Takafumi Igari*4, Takahiro Ohkubo*5,
Kenta Murakami*1
with the aid of CVR team

Abudushalamu Aili*6, Kazuhiro Yokokura*7, Yoshito Umeki*7

*1: The University of Tokyo, *2: Kajima Corporation, *3: Mitsubishi Research Institute, Inc.,*4: MRI Research Associates, Inc., *5: Chiba University, *6: Nagoya University, *7: Chubu electric company

Background

1.NRA Project　Previous Study（until JFY2016）

Purpose	Outcome	Remaining Issue
➢ Confirmation of the scientific validity of the current reference values for irradiation doses. ➢ Securing the scientific basis for the operator's application for more than 40 years, etc.	➢ Development of Scientific Basis Data for Concrete Irradiation Deterioration ➢ Revision of reference values for concrete degradation due to neutron fluence $1\times10^{20} \rightarrow 1\times10^{19}$ n/cm^2	➢ Rational evaluation methods and scientific basis data for irradiation degradation according to the actual compositional conditions of concrete

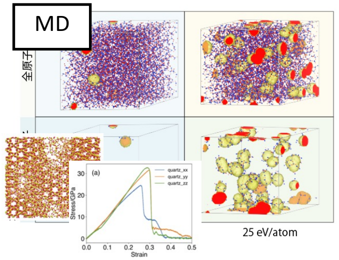
2.METI Project　（until JFY2022）

Purpose	Outcome	Remaining Issues
➢ Improvement of regulatory accountability in special inspections for operations exceeding 40yrs ➢ Expansion of knowledge and international contribution through the collaboration based on the joint research	➢ Data set of irradiated concrete aggregates in Japan ➢ Develop the procedures of integrity evaluation for irradiated concrete and RC members.	➢ Development of a method for evaluating the degradation of concrete that takes into account the irradiation rate effects and the scientific basis for this method. ➢ Achievement of international consensus on irradiation degradation of concrete

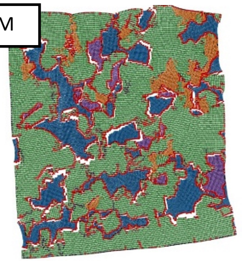
Safety / integrity evaluation process

Calculation

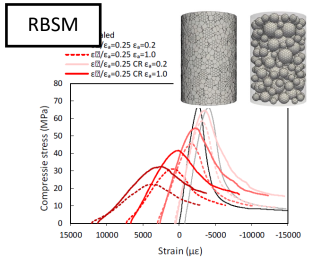
MD



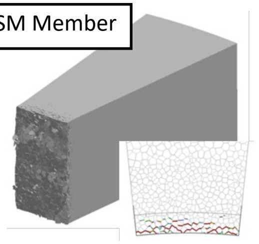
FEM

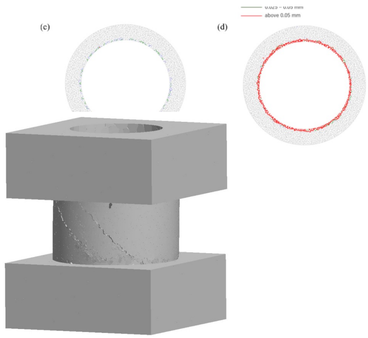


RBSM



RBSM Member





Mineral / atomic level → Aggregate level → Concrete level → Component → Structure

Experiment /Validation

XRD +TEM +Raman

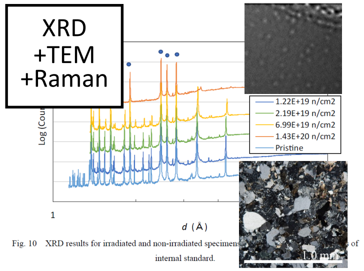
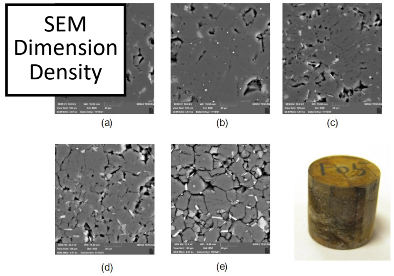
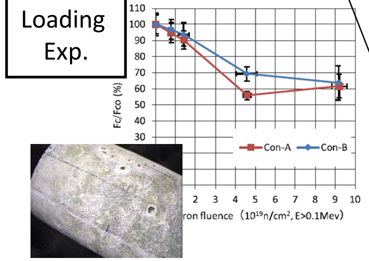


Fig. 10 XRD results for irradiated and non-irradiated specimen ... internal standard.

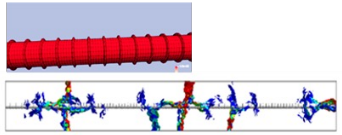
SEM Dimension Density

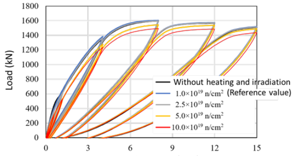


Loading Exp.

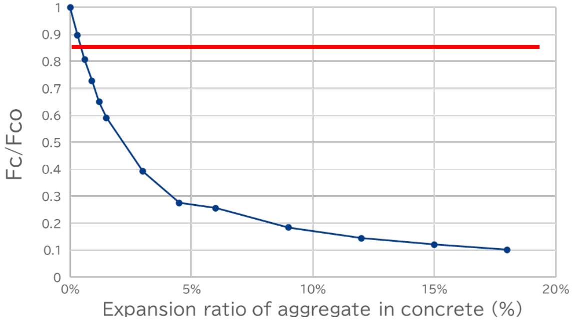


RBSM Member

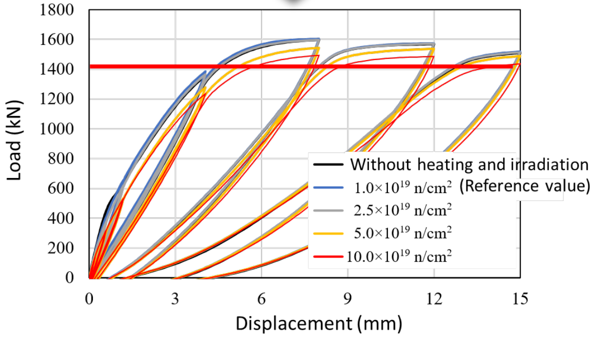




Neutron fluence threshold value



Concrete Strength Evaluation



Structural performance evaluation

Irradiation experiments

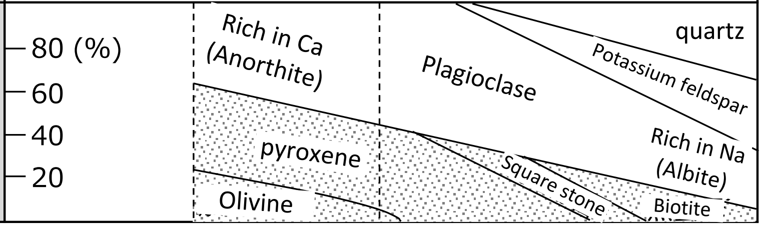
Information of concrete aggregates used in Japanese PWR were collected and representative aggregates + pure phases were selected for irradiation.

Specimen ID, type and origin

ID	Rock type / Mineral	Origin / Manufacturer
A	Synthetic quartz	Tokiwa Tech Co., Ltd.
B	Quartz glass	Shinetsu Quartz Co., Ltd.
C	Plagioclase	Itoigawa, Niigata
D	Alkaline feldspar	India
F	Granite	Takamatsu, Kagawa
G	Altered tuff	Kasugai, Aichi
H	Andesite	Satsumasendai, Kagoshima
J	Basalt	Karatsu, Saga
K	Peridotite	Samani, Hokkaido
L	Sandstone	Tsuruga, Fukui

Classification of igneous rocks

Blue: coarse aggregate used for PWR in Japan
Red: additionally selected rocks

		Ultramafic rock	Basic rocks	Intermediate rock	Acidic rock
Mottled tissue	Volcanic rock		Basalt	Andesite	Rhyolite
Equal grain texture	Plutonic rock	Peridotite	Gabbro	Diorite	Granite
SiO_2(weight%)		45% ◀──▶ 52% ◀──▶ 66%			
color		dark ◀──▶ Whitish			
specific gravity		About 3.2 ◀──▶ About 2.7			
Mineral composition					


Synthetic quartz


Quartz glass


Plagioclase


Alkaline feldspar


Granite


Altered tuff


Andesite

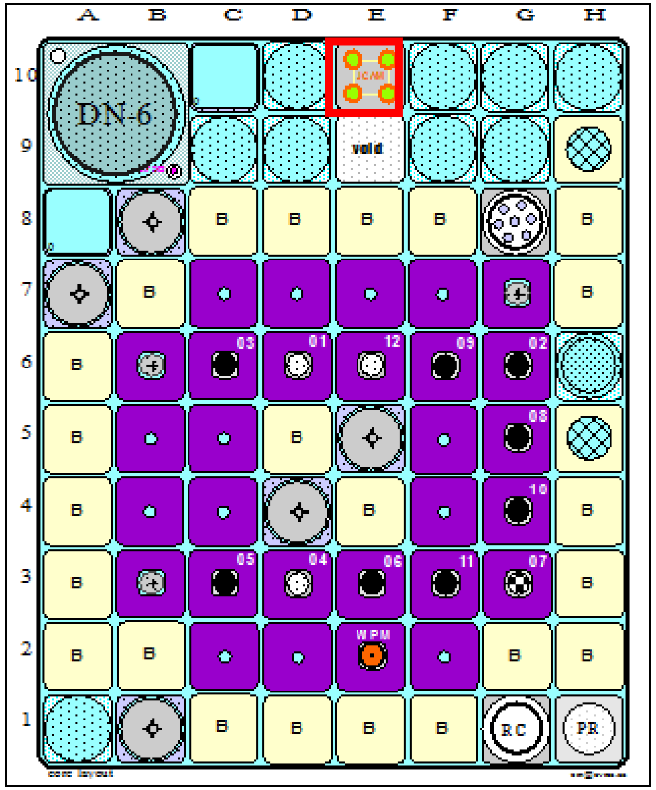

Basalt


Peridotite

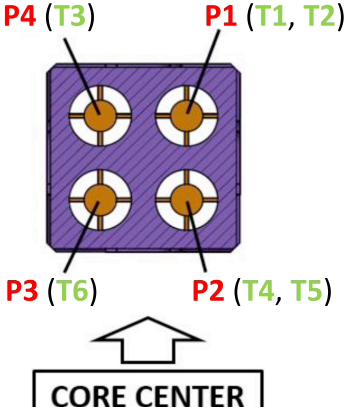

Sandstone

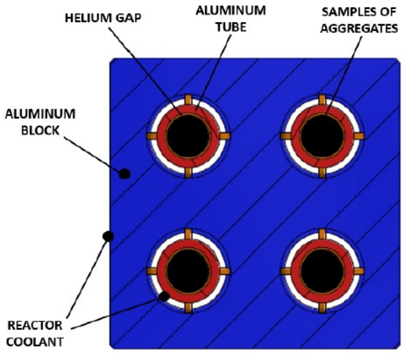
Reference: Harutaka Sakai, "Introduction to Earth Sciences, 2nd Edition, Planetary Earth and Atmosphere / Ocean Systems", Tokai University Press

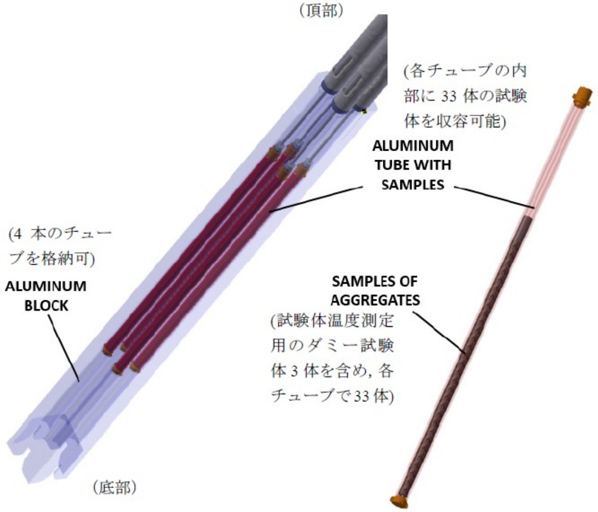
Irradiation experiment
Low flux (4.77 n/cm^2/s) + Medium flux(8.70 n/cm^2/s)

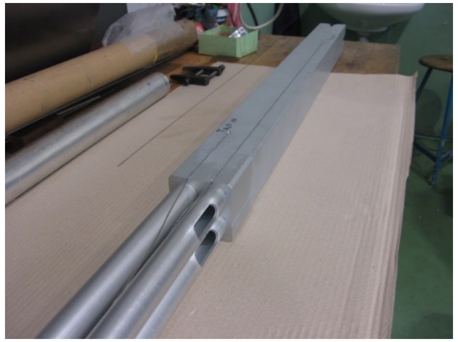


LVR-15

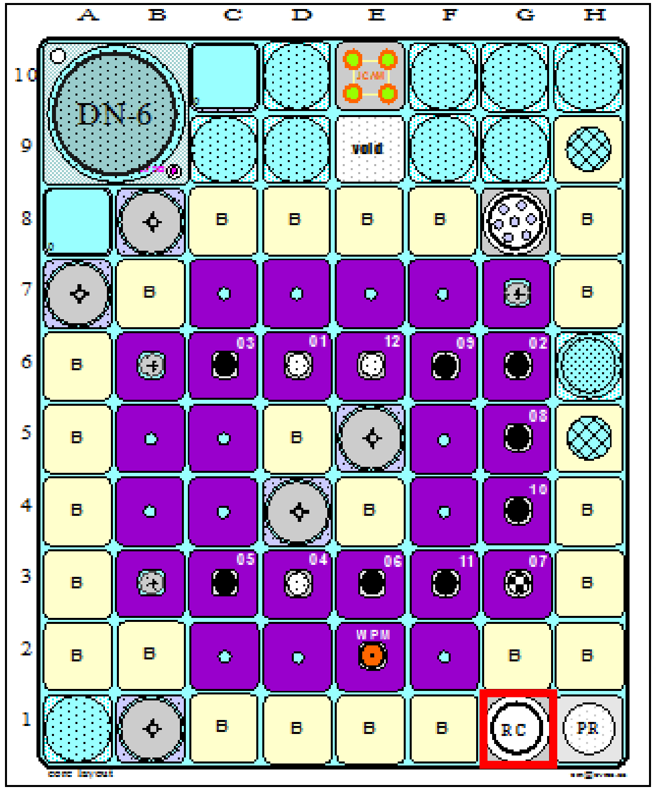


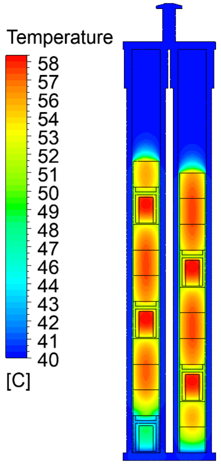




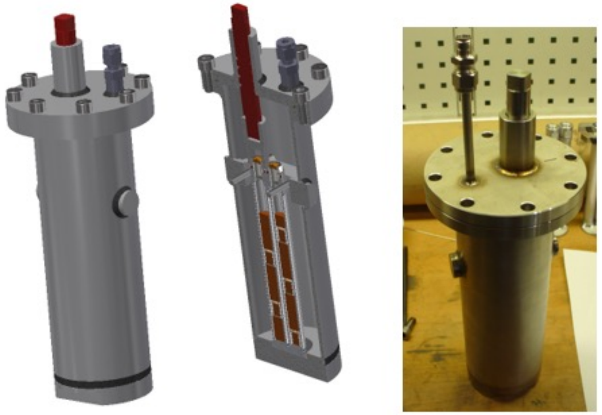


Irradiation experiment: High flux (18.7 n/cm^2/s)



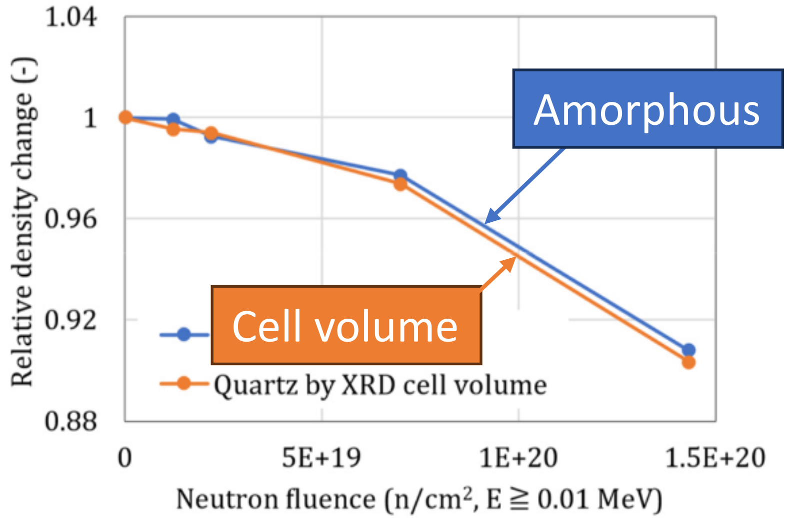


Reactor power (MW)	Cooling water temperature	Temperature – high (°C)	Temperature – low (°C)
1.1	12.2	13.3	13.0
2.0	14.8	17.3	16.9
3.5	20.4	25.3	23.5
6.9	33.1	42.2	39.7
8.8	45.1	56.7	52.8
9.0 (estimate)	44.1	56.1	52.3
9.7 (estimate)	47.1	60.0	55.9
Change of cooling circuit heat removal to full			
9.0	37.00	48.9	45.0
9.7 (estimate)	39.46	52.3	48.1



Sample capsule	Estimated temperature maximum (°C)
T8	57.22
T7	55.01
T9	53.25

Background data: IFE-irradiated sample PIE results

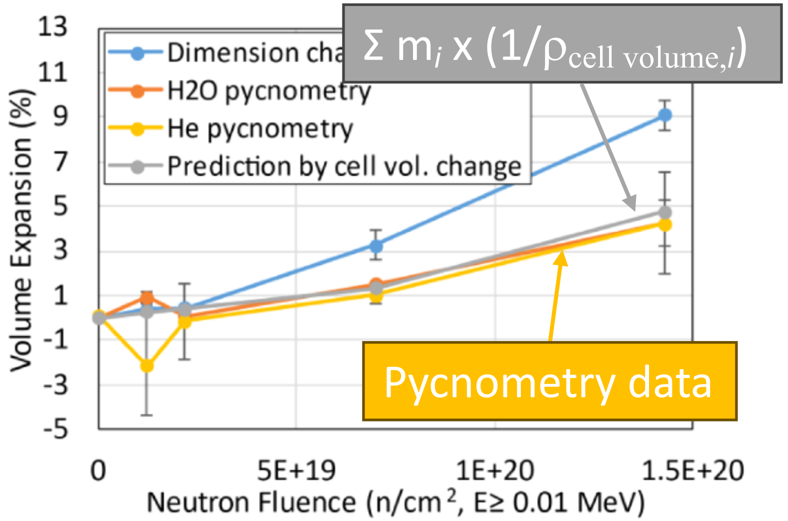


Amorphous

Cell volume

Fig. 12 Relative density change of α-quartz and amorphous phase calculated by assuming that other phases do not contribute to the volume change of aggregate.

Maruyama, I., Kondo, T., Sawada, S., Halodova, P., Fedorikova, A., Ohkubo, T., Murakami, K., Igari, T., Rodriguez, E. T., & Suzuki, K. (2022). Radiation-induced alteration of meta-chert. *Journal of Advanced Concrete Technology*, *20*(12), 760–776. https://doi.org/10.3151/jact.20.760

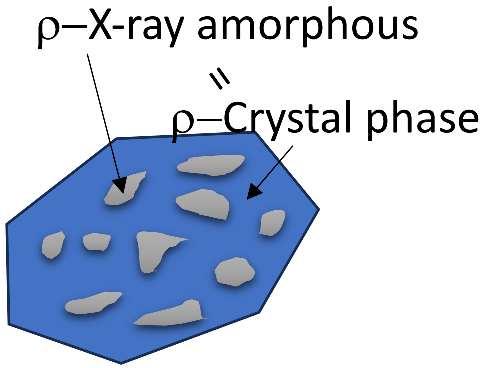


$\Sigma\ m_i \times (1/\rho_{\text{cell volume},i})$

Pycnometry data

Fig. 7. Volume expansion of sandstone based on dimensional changes, water pycnometry, and He pycnometry, and the predicted expansion based on the cell volumes of the minerals as a function of neutron fluence.

Ref: Maruyama, I., Meawad, A., Kondo, T., Sawada, S., Halodova, P., Fedorikova, A., Ohkubo, T., Murakami, K., Igari, T., Rodriguez, E. T., Maekawa, K., & Suzuki, K. (2023). Radiation-induced alteration of sandstone concrete aggregate. *Journal of Nuclear Materials*, *583*, 154547. https://doi.org/10.1016/j.jnucmat.2023.154547



ρ–X-ray amorphous
≈
ρ–Crystal phase

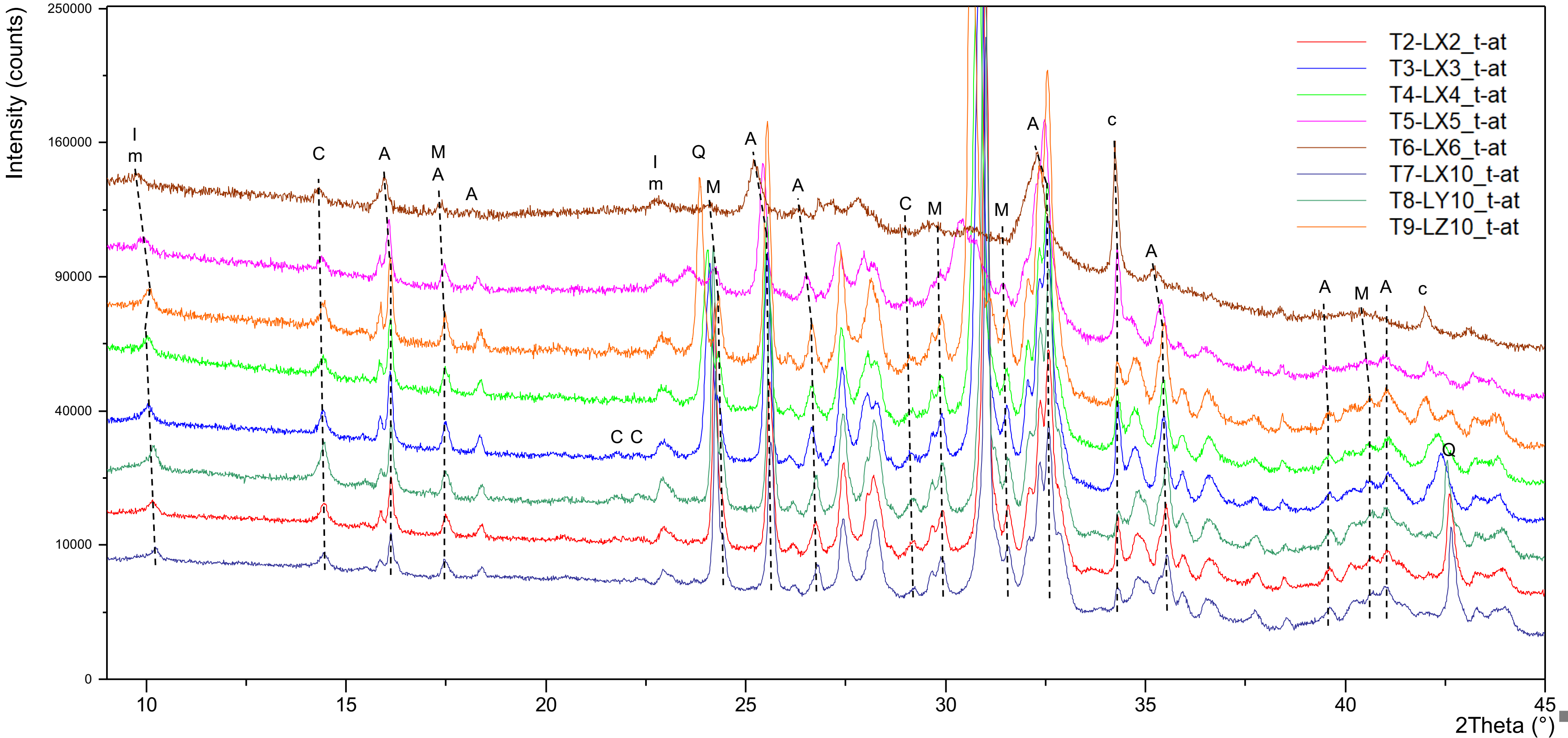
Alpha-quartz grain

- Previous IFE results proved that the density of X-ray amorphous region = density of expanded cell volume, within our irradiated experiment.

XRD/Rietveld analysis : quartz-cell volume expansion evaluation

- XRD was measured before and after irradiation

A – albite, C – clinochlore, I – illite, M – microcline, m – muscovite, Q – quartz.

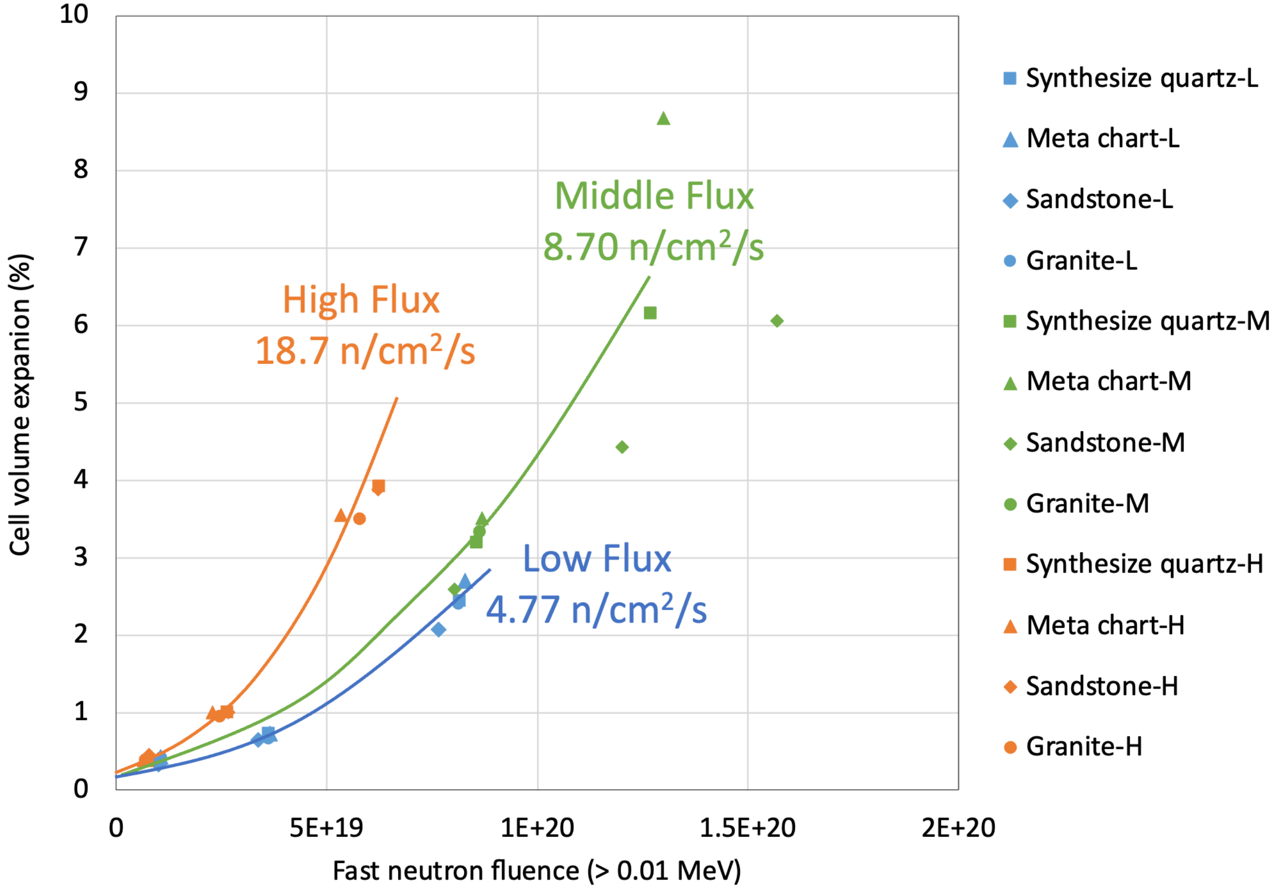


Unpublished data:
Further data validation process is needed.

Experimental results: Rietveld analysis

- α-Quartz, α-Quartz HT, β-Quartz were evaluated.
- All the cell volumes were identified.
- α-Quartz volume expansion was evaluated by the average of expansion of crystalline phases of quartz.

→ Identify the flux impact



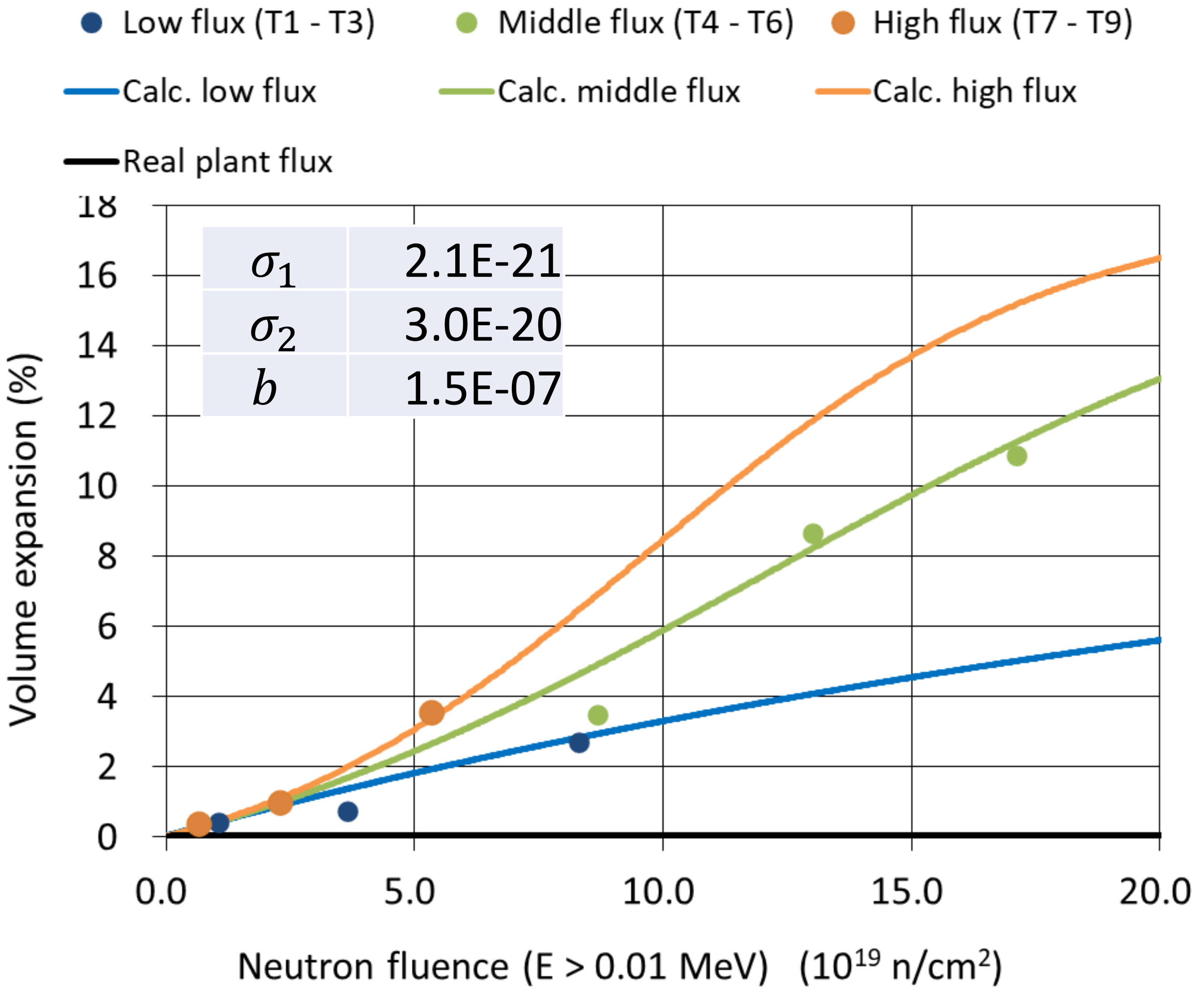
Unpublished data:
Further data validation process is needed.

Results

Here, we propose a crystalline – amorphous 2 phase model which takes into account the recovery at the interface of 2 phases:

$$R = R_1C_1 + R_2C_2$$
$$1 = C_1 + C_2$$

$$\begin{cases} \dfrac{dC_1}{dt} = -\emptyset\sigma_1C_1 - \emptyset\sigma_2C_1C_2 + bC_1C_2 \\ \dfrac{dC_2}{dt} = +\emptyset\sigma_1C_1 + \emptyset\sigma_2C_1C_2 - bC_1C_2 \end{cases}$$



σ_1	2.1E-21
σ_2	3.0E-20
b	1.5E-07

Simple model predicts that irradiation of existing reactor’s flux is less harmful.

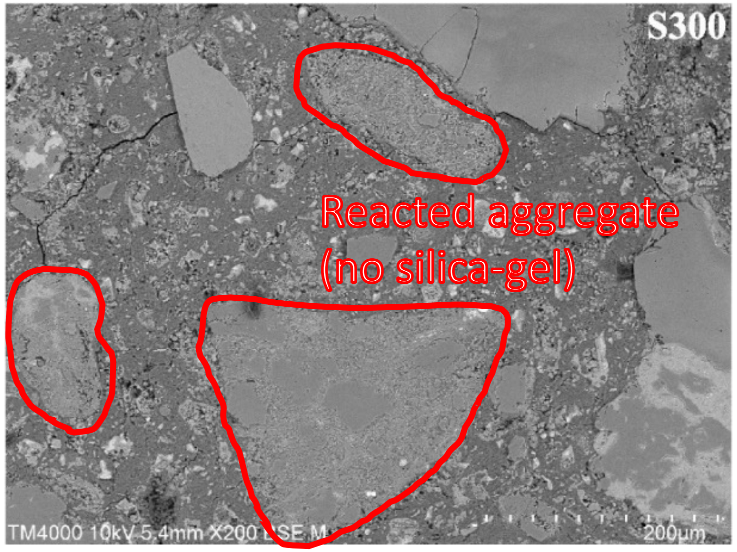
Summary

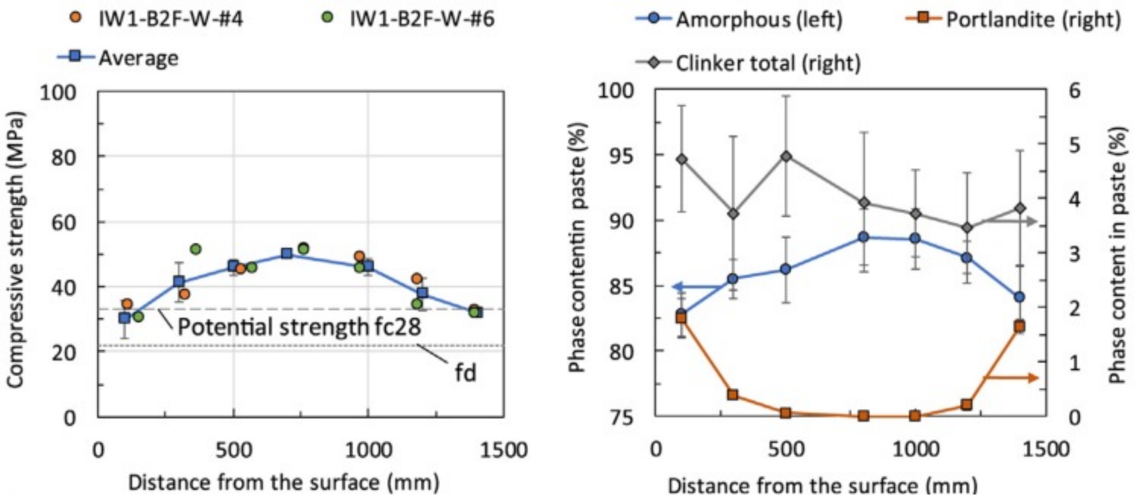
- JCAMP team identified the flux impact on the rate of expansion of alpha-quartz. This is the first evidence that the realistic neutron flux irradiation may cause less impact than those drawn by the accelerated experiments.
- Further evidences are needed. Taking cores from the real plants is meaningful.
- JCAMP team are preparing the evaluation methods for the cored samples which may have the damage distribution with steep gradient and the depth of potential damage area is very narrow.

Hamaoka Project

Main findings of Project phase I

- Strength increase of concrete in inner region of thick concrete wall.
- Reaction between aggregate and cement paste
    - hcp: Portlandite, Calcium silicate hydrates
    - Agg.: silica, alumina, alkali and other oxides
    - Reaction path: dissolution-precipitation
    - Confirmed by: Portlandite depletion, C-A-S-H increase in XRD
    - Characterized by aggregate reaction degree from ICP-AES





Strength (left) and content of portlandite and C-S-H (right) along the wall thickness, in H1-IW1

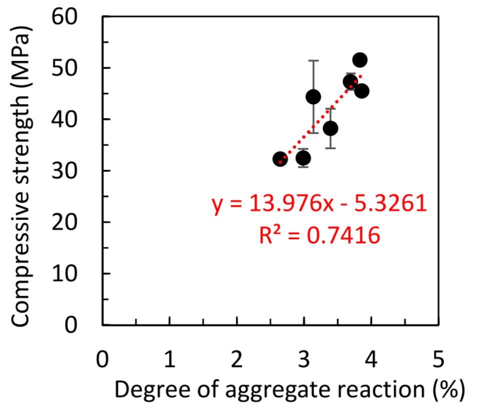


Fig. 33 Compressive strength (W) as a function of degree of aggregate reaction calculated from ICP-AES results.

- Homogenous hydration
- No evident carbonation

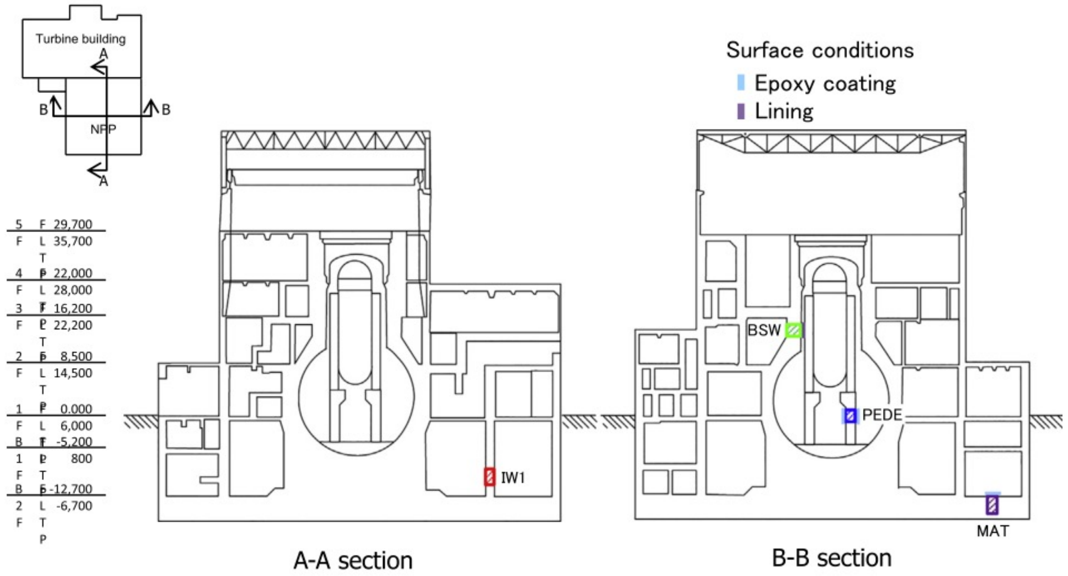
Strength increase is due to Agg.-hcp reaction

Objectives:

- Reaction, its rate, involving factors
- Mechanism of strength increase

Project Phase II

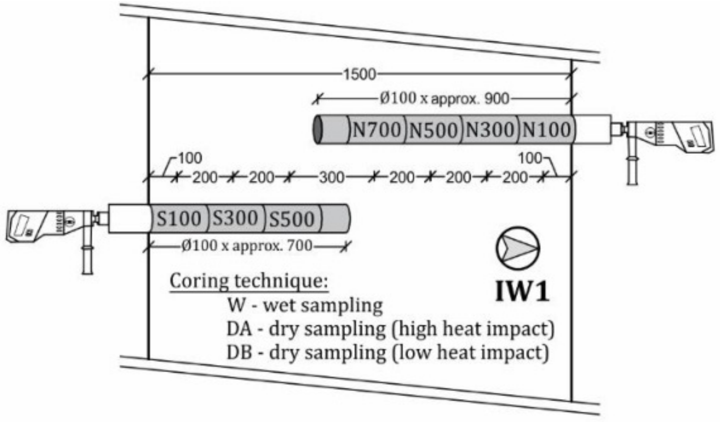
- Hamaoka power plants
  - Unit 1&2 (H1&H2) under Decom.:
  - Unit 3 (H3):
  - Unit 5 (H4):
- Members from each unit
  - Internal wall (IW1)
  - Biological shielding wall (BSW)
  - Pedestal (PEDE)
  - Mat slab (MAT)



Unit/ Member	Age of construction (years)	Cement type	Design strength (MPa)	Water to cement ratio (%)	Sand/Agg. volume ratio (%)
H1-IW1	47	OPC	22	48.3	38.5
H1-BSW	47	MPC	22	48.0	39.7
H1-PEDE	47	MPC	22	49.0	42.0
H2	47	MPC	24	48.0	43.0
H3	36	MPC	24	52.0	45.2
H5	16	MPC	32	49.0	45.5

Experimental data

- Temperature history of the members:
  - IW1: 20-30°C
  - BSW: 30-38/50-55°C during operation, 20-30°C afterwards
  - PEDE: 20-30/50-55°C during operation, 20-30°C afterwards
- Cored samples from various thick walls for:
  - Mechanical properties such as strength and elasticity, etc.
  - Physical properties such as water content, RH, porosity, etc.
  - Chemical composition by TG, XRD, ICP-AES



Schematic representation of coring, example of H1-IW1

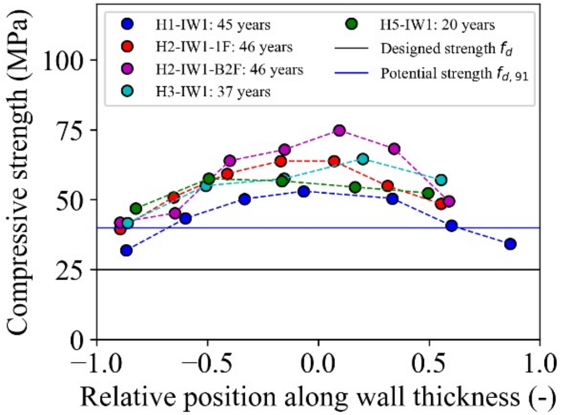
Target wall	Wall thickness (mm)	Number of samples per core	Surface condition[a]	Temperature during operation (°C)	Temperature after operation (°C)	Duration of operation (years)
H1-IW1	1500	7	N/N	20-30	20-30	16.5
H1-BSW	2200	7	N/L	30-38/50-55	20-30	16.5
H1-PEDE	1220	5	E/E	20-30/50-55	20-30	16.5
H2-IW1-1F	1700	7	N/N	20-30	20-30	18.4
H2-IW1-B2F	1700	7	N/N	20-30	20-30	18.4
H2-BSW	2200	7	N/L	30-38/50-55	20-30	18.4
H2-PEDE	1380	5	E/P	50-55	20-30	18.4
H3-IW1	1300	5	N/N	20-30	20-30	18.4
H5-IW1	1000	5	N/N	20-30	20-30	3.1

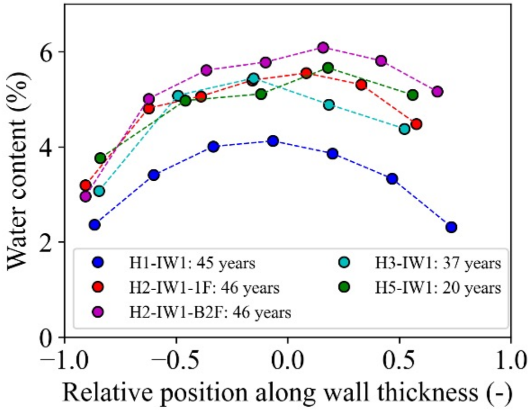
[a] N: bare surface; L: steel liner; E: epoxy resin coating; P: steel plate.

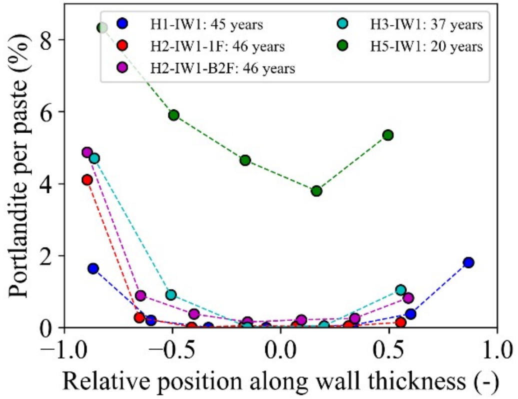
Results

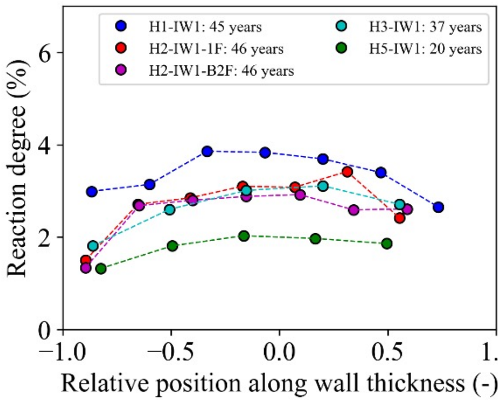
Strength | Water content | CH amount | Agg. reaction

Internal walls

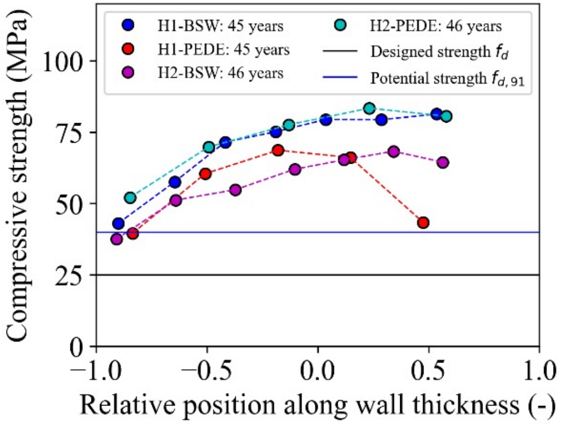


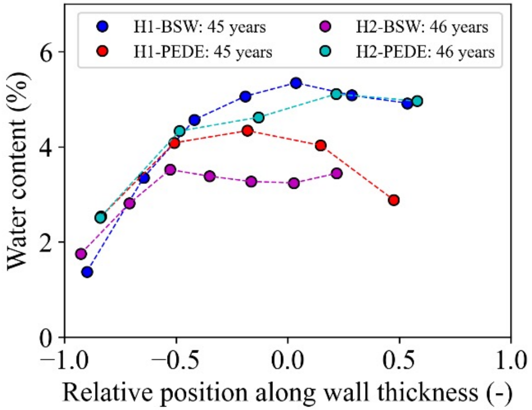


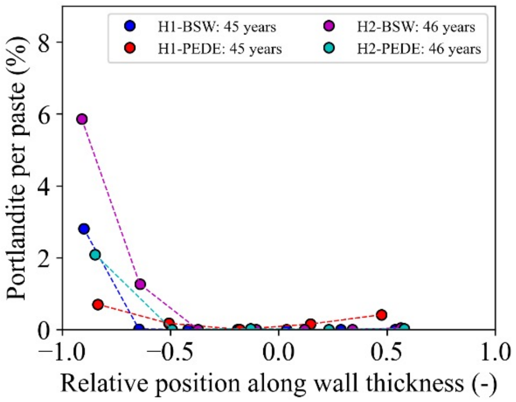


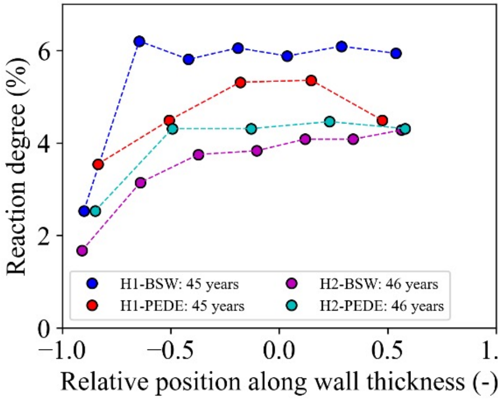


Irradiated walls
(PEDE / BSW)

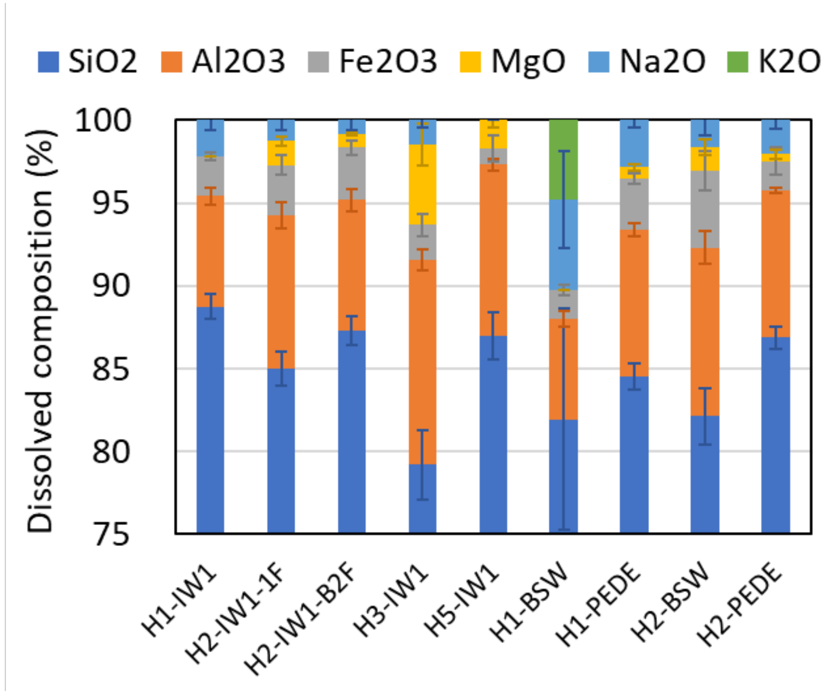


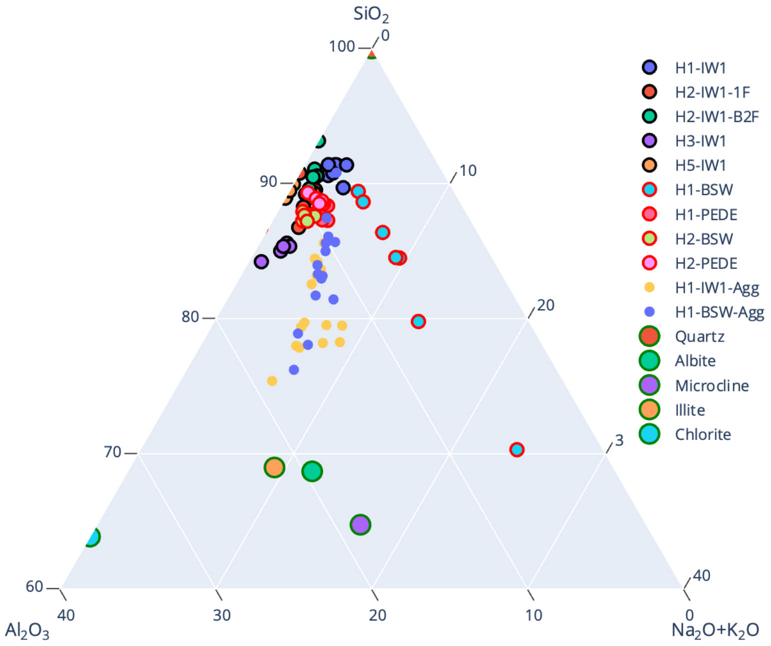






What were reacted?

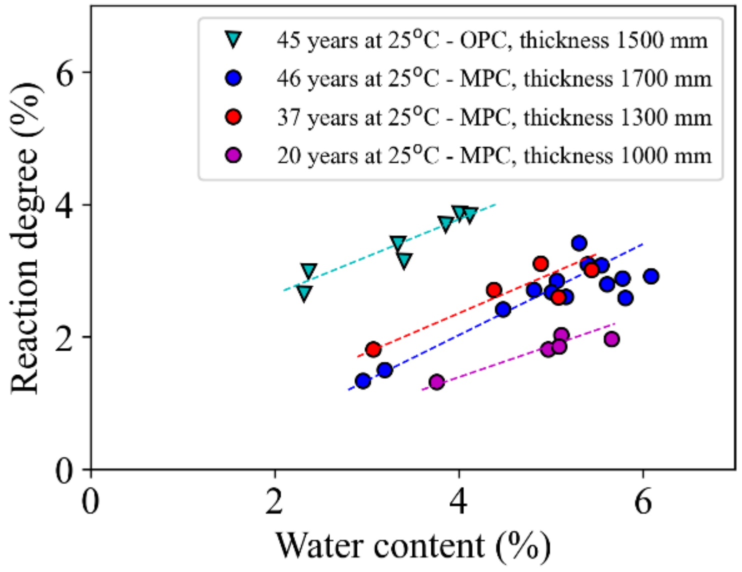




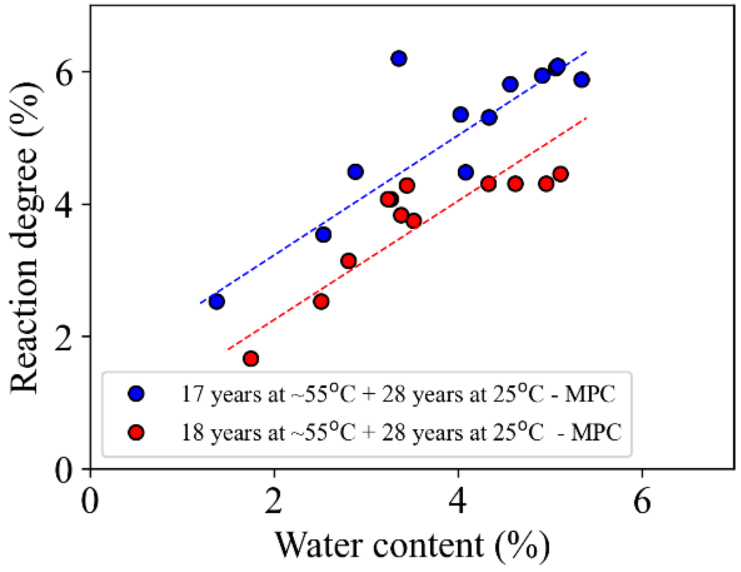
- quartz, chlorite, albite, and illite can be reacted in the system.

Rate of reaction

Internal walls



Irradiated walls
(PEDE / BSW)

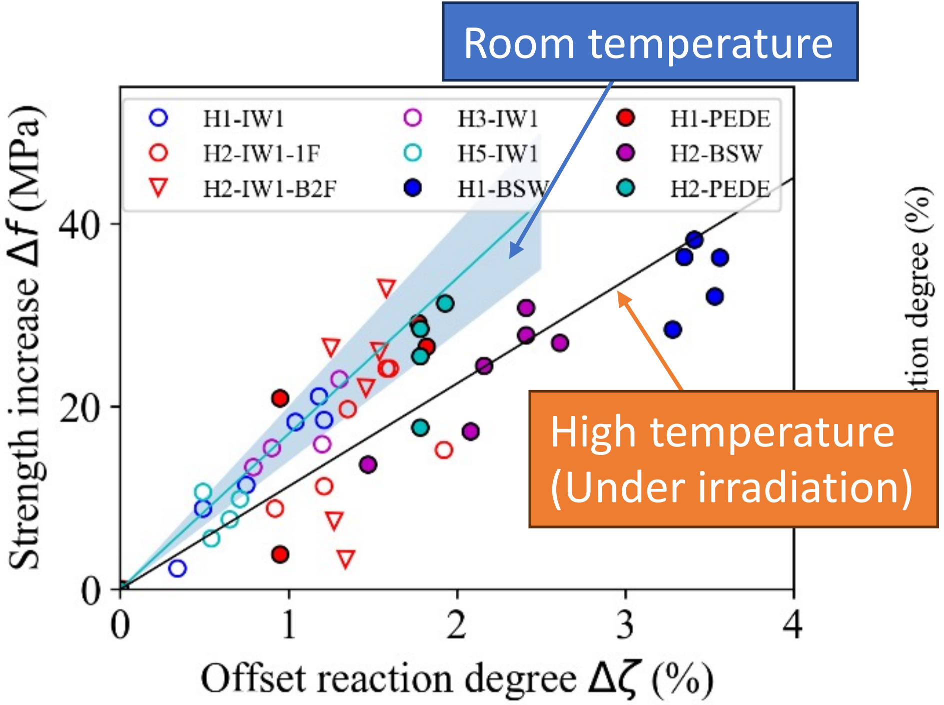


Without high-temperature history			With high-temperature history		
Walls	β (-)	$k(20°C)$[a] (%/year)	Walls	β (-)	$k(20°C$ (%/year)
α_0=0.1, θ=0.1, E_a=28 kJ/mol					
H1-IW1	10	0.086	H1-BSW	20	0.094
H2-IW1	100	0.056	H1-PEDE	20	0.108
H3-IW1	100	0.079	H2-BSW	20	0.064
H5-IW1	100	0.108	H2-PEDE	50	0.075

[a] For ease of understanding physical meaning, reaction rate $k(20°C)$ is given instead of A. The relation between A and $k(20°C)$ is shown in Eq.(4).

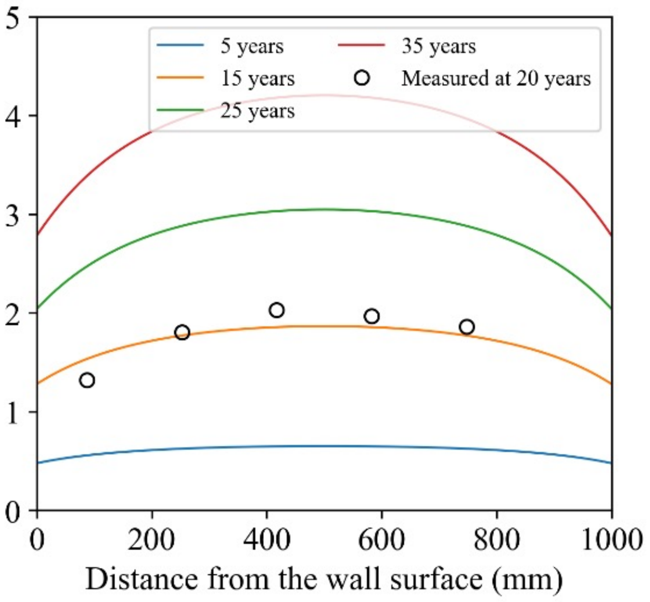
- Rate of reaction (mass%/year) and Activation of energy were obtained.

Strength prediction

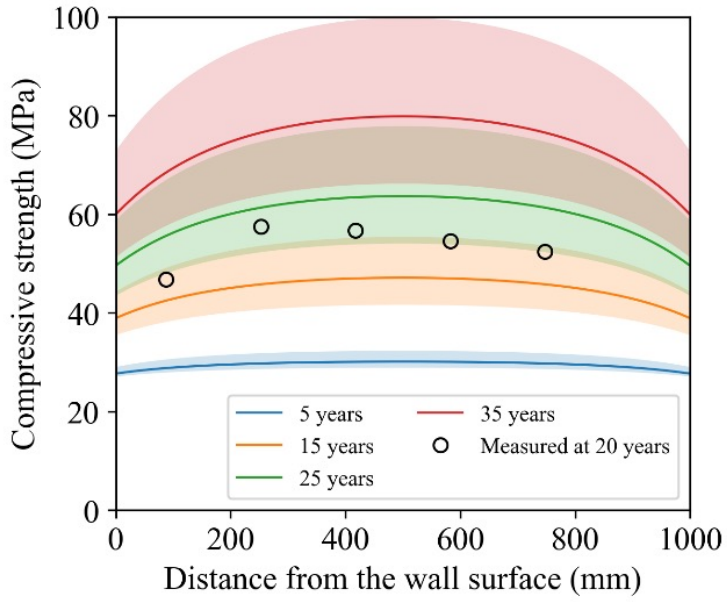


Room temperature

High temperature (Under irradiation)



Reaction degree of Agg.



Strength development

- FDM → water content + temperature distribution → Rate of reaction degree.
  → Microstructure change, Diffusion coeff. + water consumption → FDM
- Strength development of thick concrete wall can be predicted.

Summary and comments

- General sandstone fine aggregate may be reactive for long-period.
- But aggregate did not show the ASR. The dissolution rate vs Ca movement is the key. (Another paper is in preparation.)
- Slow reaction of aggregate enhance the strength, which contributes to the high performance of shear wall.
- Temperature (Gamma-ray induced) has accelerated this phenomenon.
- Neutron also may influence on increasing in dissolution rate of minerals by metamictication (neutron-irradiated amorphization)
- This influence should have also an important role in the integrity evaluation of RC member exposed to irradiation.

Thank you for your attention.

Free to ask:
Ippei Maruyama
i.maruyama@bme.arch.t.u-tokyo.ac.jp