TOPIC: 3-6 Non-metallic material Ageing Management

Development of System for Evaluating Concrete Strength Deterioration Due to Radiation and Resultant Heat

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Abstract. Evaluation of the soundness of concrete exposed to irradiation has been studied within the framework of a project of the Nuclear and Industrial Safety Agency (NISA) "Japan Ageing Management Program for System Safety". This contribution presents the background to the existing evaluation processes, a review of the irradiation exposure effect on concrete and needs for irradiation testing. Based on results of this study, working assumptions for the development of an evaluation system are derived, and an overall picture of a numerical model as well as a framework for evaluating concrete soundness under irradiated conditions are proposed.

1. Introduction

Twenty nuclear power plants (NPP) in Japan have undergone an examination called PLM (Plant Life Management) by the Japanese government to determine their ability to carry on continuous operation. Japan has now entered the period of safety management of NPPs of different ages. Additionally, after the Tōhoku earthquake, an improved system evaluation method for NPPs was considered desirable by the people as well as the electric companies. Based on performance-based evaluation, time-dependent performance of structures and their components should be judged on the basis of the deterioration mechanism and a mechanism-based quantitative prediction method.

In assessing the reduction of concrete strength under radiation fields, $1x10^{20}$ n/cm^2 for fast neutrons and $2x10^{10}$ rad ($2x10^8$ Gy) for gamma rays have been adopted as the reference levels of nuclear radiation. Concrete structures have been regarded as sound as long as the radiation levels have been lower than the reference levels, even if the NPPs have operated for 30 years or more. The reference levels were estimated from experimental data summarized by Hilsdorf [1]. However, the experimental conditions of the data were very different from the actual irradiation conditions of concrete structures of light water reactor (LWR) power plants. This paper examines the applicability of these reference levels to the soundness evaluation of concrete against nuclear radiation, possible mechanisms of concrete degradation including interaction between concrete components and nuclear radiation, and a scheme to develop a system for evaluating concrete strength deterioration due to radiation and resultant heat.

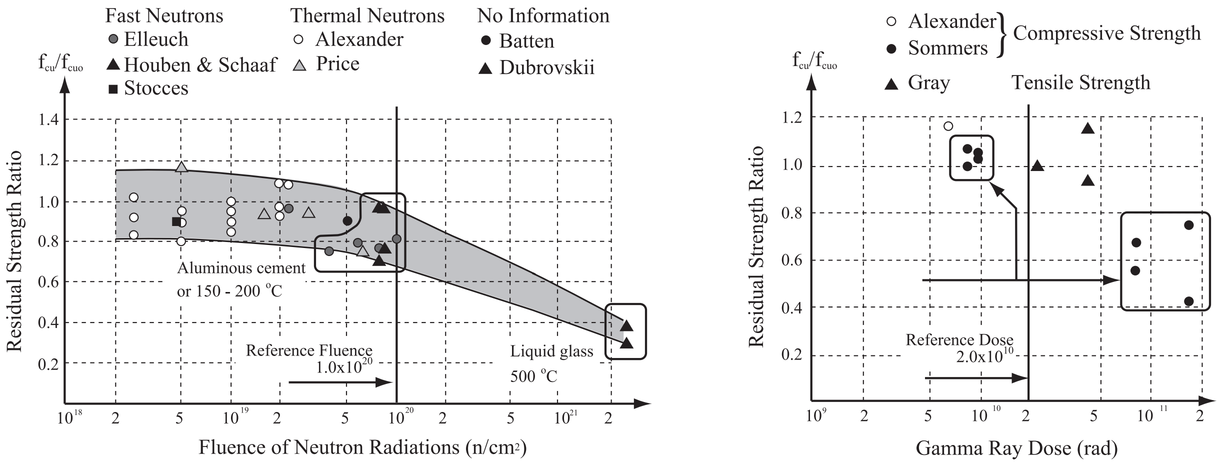
2. Backgrounds of the project

2.1 Background of the reference level

Most experimental studies on irradiation effects on concrete were carried out in the 1960's and 1970's. Although deduction of the mechanism of concrete strength deterioration was quite difficult with the available data, Hilsdorf put the original data together to illustrate the residual strength ratio of concrete as a function of fluence or dosage, as shown in Figure 1. The reference levels adopted so far seem to have been determined from these figures.

When the experimental conditions of data plotted in Fig. 1 are examined, inconsistencies are found regarding measured properties [2], binder type of concrete [3,4], specimen size [5], temperature of concrete [3,4,5,6,7], and neutron energy level [2,3,7,8]. However, concrete structures for LWRs are made from Portland cement and are subjected to fast neutron radiation with the temperature kept less than 65 deg. C, and the targeted property for evaluating soundness is compressive strength. The situation is similar for evaluation of the reference level for gamma irradiation. In the experiment of Sommers [9], specimens were cured in deionized water, and Gray evaluated tensile strength [10]. More detail is shown in a recent paper [11].

This implies that additional new data are necessary to evaluate the applicability of the reference level and real structural performance of concrete members supporting reactors. New experiments should be designed to clarify the mechanism of concrete deterioration by nuclear radiation. This will require knowledge of both concrete and radiation sciences.



(a) Neutron radiation effect

(b) Gamma radiation effect

FIG.1 Residual strength ratio of concrete exposed to neutron radiation (a) and gamma radiation (b). (after [1], some notes are added together.)

2.2 Interactions between radiation and concrete components

2.2.1 Structures of concrete components

Concrete is macroscopically composed of two phases: cement paste and aggregate. Cement paste comprises hydration products of Portland cement. Aggregate is made of crushed natural rocks or gravels.

Portland cement reacts with water to form hydration products. Unhydrated portland cement generally comprises five mineral constituents of which alite and belite, which are silicates of calcium, are responsible for the strength of the cement paste. Alite and belite form calcium silicate hydrates (C-S-H), which have a complex internal pore structure with a high specific surface area (approx. 200 m^2/g by water vapor BET analysis). It can be visualized as a tobermorite with many imperfections and irregularities such that it seems amorphous.

A large number of water molecules exist in the cement paste system in different states. They can be divided into three categories according to how strongly they are bound to the hydration products, as shown in Figure 2. Free water exists in capillary pores and behaves like bulk water. Gel water is absorbed on the surface of hydration products, although crystal surfaces of C-S-H are not clear. Chemically bound water is strongly fixed to hydration products. The definition of chemically bound water is still under discussion and dehydration at 105°C under CO_2-free conditions is normally used to distinguish between chemically bound water and non-evaporable water [12]. Gel water and capillary water are dehydrated below 105 °C as evaporable water.

Unlike cement paste, aggregates are usually well-crystallized rocks originating from a large part of the earth's surface and comprise SiO_2 and Al_2O_3. Aggregates contain evaporable water and non-evaporable water, although their amounts are much less than in cement paste. Water absorption of aggregates used in NPP would be less than 2.5% and the proportion of non-evaporable water (crystallization water) in aggregates ranges from 0.5% to 1.5% [13].

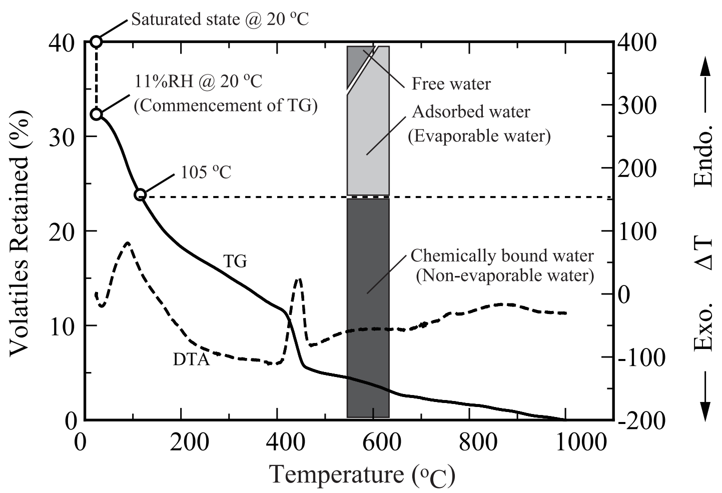


FIG.2 Types of water in cement paste with TG-DTA data of ordinary Portland cement paste.

Water to binder ratio of the cement paste is 0.55, and it was sealed and cured at 20 °C. Prior to the TG-DTA analysis, specimen was equilibrated with dry air of 11%RH at 20 °C.

2.2.2 Interaction between gamma rays and concrete components

Collision of gamma rays with materials, whether it results in photoelectric effects, Compton scattering, or pair production, causes electrons to be ejected and positron is occasionally involved. The ejected or produced electrons collide with materials to eject more electrons, so that the number of ejected electrons (secondary electrons) continuously increases until the energies of the secondary electrons become lower than the ionization energies of the materials. Since the mass of secondary electrons is very low, they do not directly eject atoms composing the materials. Gamma rays have therefore very little effect on the solid materials composed of rather isotropic, ionic and metallic bonds.

However, gamma rays affect materials through electronic excitations. Collision of secondary electrons with materials causes electronic excitations. The electrons in the excited states loosen their excess electronic energies to the vibrational energies of atoms composing the materials, resulting in the destruction of anisotropic chemical bonds such as covalent bonds. Siliceous minerals are therefore likely to be decomposed by gamma irradiation because the Si-O bond is covalent [14].

Water, which exists in concrete as one of its main component, is likely to be decomposed by gamma rays into hydrogen, oxygen and hydrogen peroxide [15]. Since water exists in cement paste rather than aggregate, gamma rays have more effect on cement paste than on aggregates. Gamma rays from nuclear reactors with an energy ranging from 100 KeV to 10 MeV interact with cement paste and aggregates via Compton scattering.

2.2.3 Interaction between neutron and concrete components

Interactions between neutrons and materials can be broadly classified into two categories: scattering and absorption. When a fast neutron collides with a heavier atomic nucleus, the neutron loses its energy to become an intermediate neutron by releasing a neutron and the extra energy as scattering gamma rays, which is called inelastic scattering. The intermediate neutron interacts with a lighter nucleus and neutron via elastic scattering while preserving the total kinetic energy. This scattering process accompanies the dislocation of atoms from their original positions. The intermediate neutrons produces hundreds of elastic scatterings and reduce their own energy till they become thermal neutrons. Once neutrons become thermal neutrons, their energy levels are not sufficient to eject neutrons from atomic nuclei.

The reference level of neutrons is assumed to be more than 0.1 MeV. Neutrons affect the solid phase by kicking out atoms from their original locations. According to some papers, limestone and siliceous aggregates expanded under neutron irradiation [16, 17, 18]. Since aggregates are dense and well-crystallized materials, when neutrons collide with the crystal lattice, the lattice constant increases, defects are accumulated and finally specific volume increases. Only a limited number of test results [19] of neutron irradiation on cement paste showed that the cement samples showed no changes in their appearance. Although dislocation of atoms in solid cement paste may take place, lattice defect by distortion may not be accumulated due to its amorphous nature.

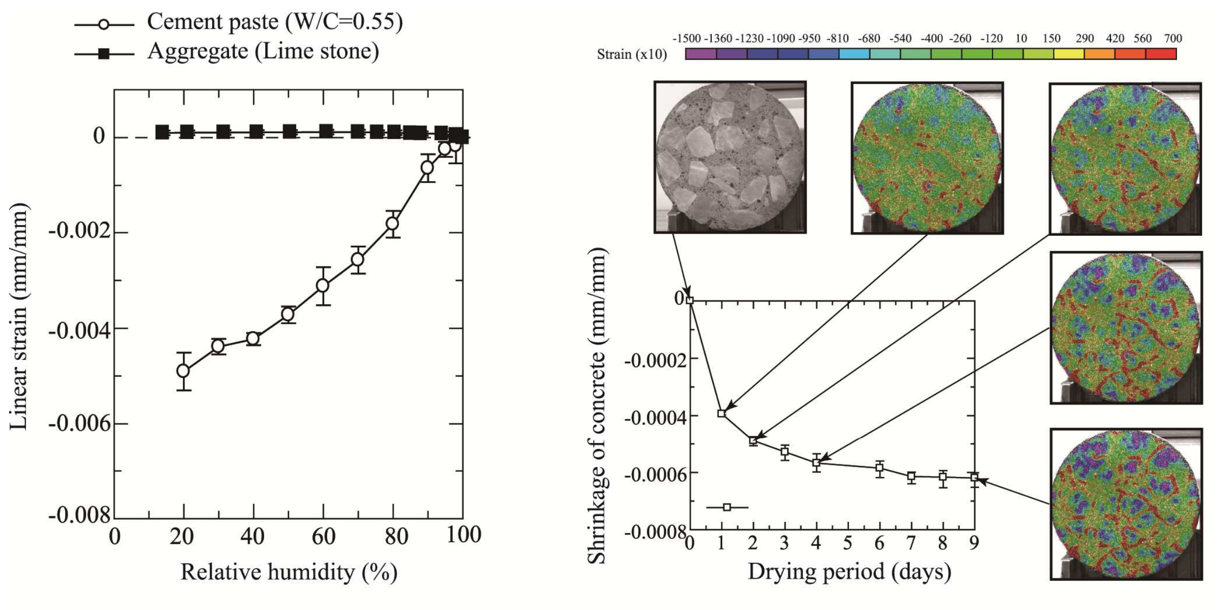
2.3 Mechanism of concrete deterioration

Even under drying and constant temperature conditions, volume changes of cement paste and aggregate are different, and this leads to cracking. Linear deformation during drying of cement paste and aggregate, and resulting cracking, are shown in Figure 3.

Cement paste and aggregates behave differently under radiation exposure conditions. Cement paste shrinks under drying conditions. Even at higher temperatures, this shrinkage exceeds thermal expansion. Under gamma-ray irradiation, water in the cement paste matrix is easily decomposed, and in addition, temperature increase of the matrix due to gamma heating causes water evaporation. Therefore, considerable shrinkage in cement paste is expected. On the other hand, aggregates, which have well-crystallized materials, expand due to accumulation of defects in their crystal structures.

Concrete strength as well as stiffness is most likely degraded in accordance with the volume change of cement paste and aggregate. These interactions are summarized in Table I.

To understand time-dependent concrete properties and real structural performance of reinforced concrete structures, it is necessary to examine water behavior in cement paste, volume change of cement paste and aggregate, and concrete strength and stiffness under irradiated conditions.



(a) Shrinkage strain of cement paste and aggregate

(b) Shrinkage strain and internal strain distribution of concrete

FIG.3 Shrinkage strains of concrete components (a) and average shrinkage strain of concrete cured under 60% RH and 20 ^{o}C and internal strain distribution in concrete (b).

Table I. Effect of gamma rays and neutrons on concrete and its components.

	Gamma Rays	Neutrons
Cement Paste	**[Water]** 1) Water is decomposed by radiolysis to generate hydrogen and hydrogen peroxide which in turn decomposes into water and oxygen. 2) Gamma heating cause additional hydration of unhydrated cement and phase change of hydrates. 3) Gamma heating causes severe drying in cement paste. **[Solids]** 4) Si-O bond of C-S-H may be slightly decomposed due to the covalent nature. 5) Electrons are ejected by scattering of gamma rays and collide with the solid phases.	**[Water]** 8) Molecular products from water may be the same as those for gamma rays, but the yields are different due to the difference of linear energy transfer. **[Solids]** 9) Dislocation of atoms in solid phase of cement paste takes place, the lattice defect will not be accumulated due to its original crystalline imperfections.
Aggregate	**[Water]** 6) Small amount of water will be released by the radiolysis and gamma heating. **[Solids]** 7) Siliceous aggregate may be slightly decomposed.	**[Water]** 10) Same as that of cement paste. **[Solids]** 11) Lattice constants are increased due to the dislocation of atoms, and lattice defects are accumulated.
Concrete	12) *Drying cause the large shrinkage, lose stiffness, and strength change of cement paste. This results cracks around aggregate, and stiffness and strength of concrete will be deteriorated.* 13) Additional reaction of unhydrated cement may increase the strength of concrete. 14) Hydrogen peroxide generated in radiolysis process may react with cement paste, and may affect on the strength of concrete.	15) *Increase of lattice constant and accumulation of defects in aggregate cause expansion. This expansion produces cracking around aggregate and stiffness and strength of concrete will be deteriorated.* 17) Siliceous aggregate may show larger expansion than non-siliceous aggregate.

3. Evaluation system for concrete structures under radiation and resultant heat

3.1 Diagram for numerical analysis

It is helpful in evaluating the soundness of concrete structures affected by irradiation to develop a numerical simulation that predicts changes in concrete strength and stiffness of concrete members based on a scientific background. The possible mechanism of concrete deterioration as a result of differences between volume changes of cement paste and aggregate is shown in 2.3. Besides this, modeling of cement hydration, phase composition, volume change of cement paste, stiffness and strength of cement paste, and response of cement paste and aggregate to a given environment would enable prediction of concrete strength and stiffness. When a concrete member is assumed to be a target field, governing equations are solved as unsteady state systems under given boundary conditions such as heat flux, temperature, humidity, neutron fluence and gamma-ray dose rate. An overall picture for the numerical simulation is proposed as shown in FIG.4.

Experimental results of control test pieces in construction, core drilled specimen, and non-destructive testing could contribute to simulation accuracy.

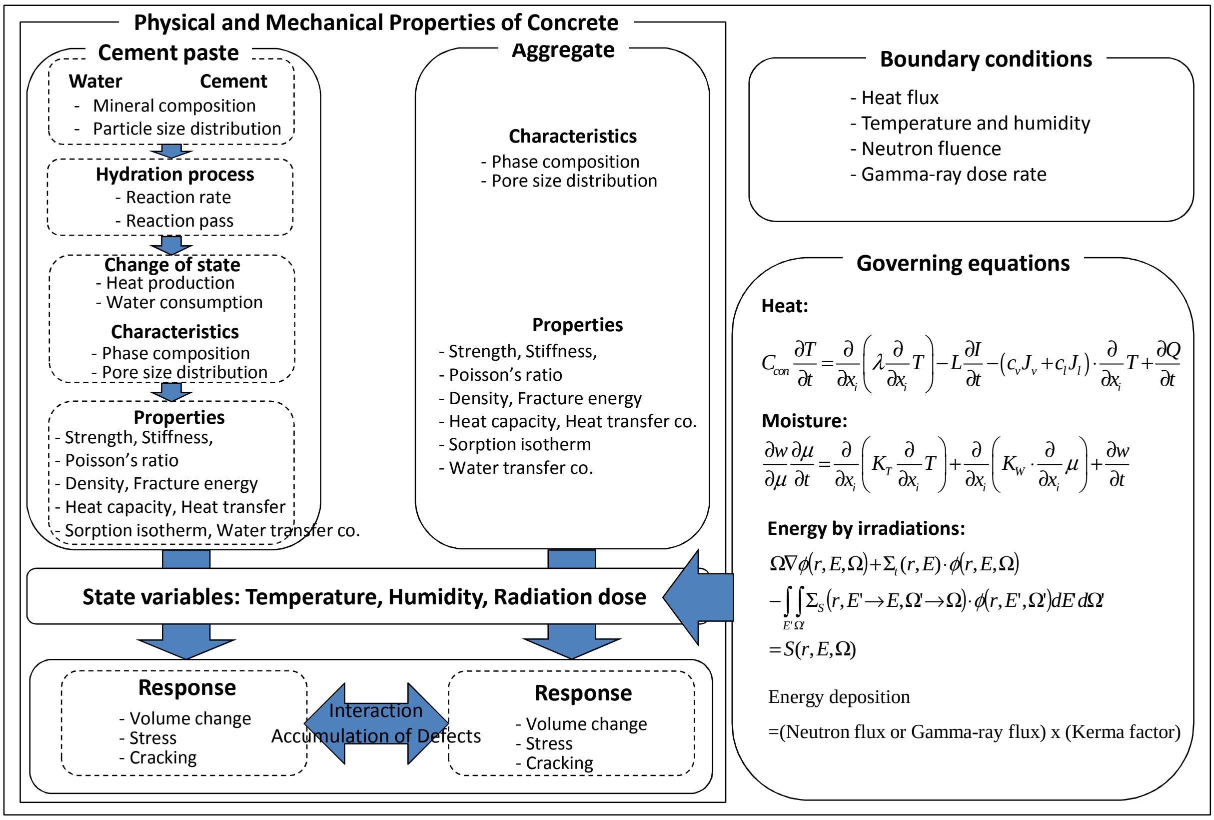


FIG.4 Diagram of numerical simulation for concrete deterioration under irradiated conditions.

3.2 Assessment methods.

Based on the mechanism of concrete deterioration due to irradiation, several assessment methods can be proposed. Firstly, the most basic assessment method is to use a limit value. This value can be a function of mineral composition, particle shape, and particle size distribution of aggregate. In this procedure, concrete strength will not be required.

Secondly, assessment by compressive concrete strength is also possible. Even if concrete deterioration is observed, concrete strength could still exceed the required design strength. Generally, real concrete strength in concrete members exceeds design strength because the concrete mix proportions consider variations of concrete quality and therefore include some margin. For this procedure, several core-drilled samples are needed. Alternatively, combinations of numerical simulation and control specimens or non-destructive tests are reasonable.

Thirdly, an assessment based on structural performance is important. It is likely that the surface concrete subjected to irradiation deteriorates and becomes lower than the required design strength, while the inner concrete is still sound. In this case, the structural performance should be examined taking account of the distribution of concrete strength. These three options are summarized in the assessment flow chart shown in Figure 5.

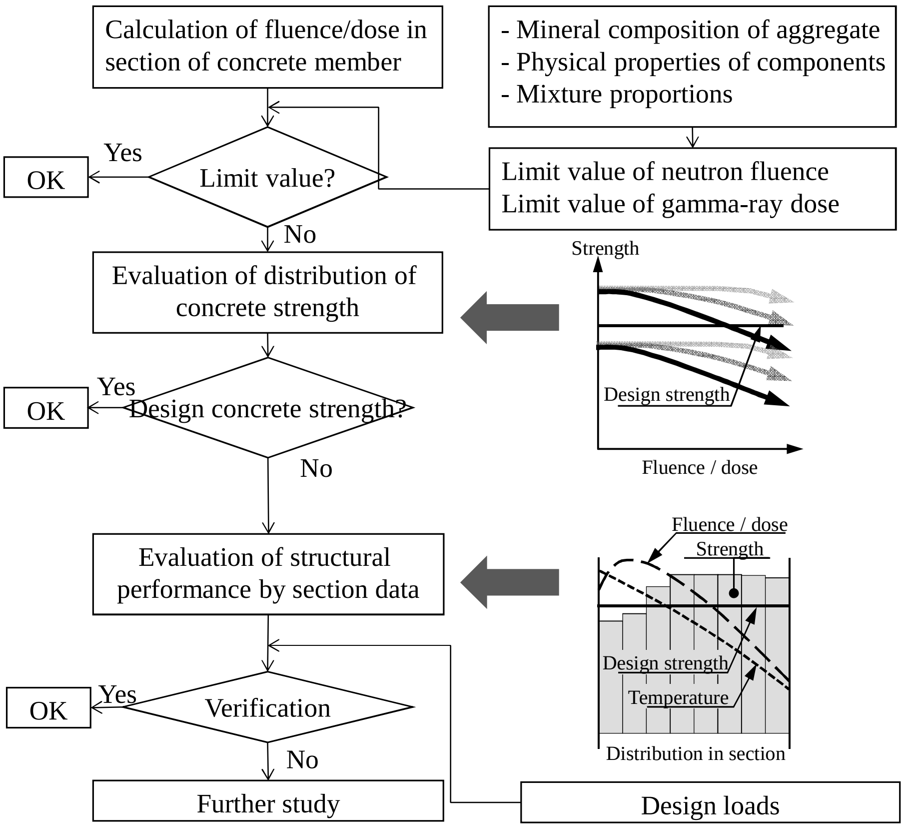


FIG.5 Assessment flow chart for soundness of concrete member exposed to radiation.

4. Schedule

The present project will be concluded by the end of Mar. 2016, and the irradiation experiments will be completed by mid 2015.

5. Conclusions

1) Technical information necessary for determining appropriate limit value of nuclear radiation in assessing the soundness of concrete structures under irradiation conditions was found to be very limited. Consequently, further experimental studies are needed.

2) Interaction between radiation and concrete components was investigated, and it was noted that neutrons and gamma rays have different effects on cement paste and aggregate.

3) Based on the interaction mechanism of concrete components with radiation, a deterioration mechanism of concrete was proposed. An overall picture of a numerical simulation capable of evaluating changes in concrete strength in concrete member was presented.

4) According to the possible mechanisms of concrete deterioration due to radiation, several assessment methods were proposed and their flow charts were presented.

Acknowledgements

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