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The Impact of Neutron Irradiation on Concrete Structures

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ABSTRACT

Concrete is the most common and the predominant material used in the construction of nuclear plants, radioactive waste repositories and their shielding materials. Many of these structures are large and irreplaceable sections; therefore, studying and enhancing the durability of the concrete design over time is essential. The microstructure and properties of dry cement paste and/or aggregates of different types change over time due to slow hydration, crystallization of amorphous constituents, and reactions between cement paste and aggregates, as well as influences from the local environment such as humidity, temperature, radiation exposure, and/or chemical interactions. This paper is reflecting the current state of knowledge about the components of concrete and the behaviour under such circumstances over time.

1 INTRODUCTION

Most of the presently operational nuclear plants were built in the 1970-80s aiming 40-years of operation. The recent projects aim 60 or even 80 years operations with higher power load resulting almost one order of magnitude higher neutron fluence for the structural materials close to the core. Though concrete is a relatively cheap material and easy to be casted while being efficient shielding against neutrons and gamma-rays, their behaviour during the life span of the plant is an open issue. However, to ensure the continuous safe operation of nuclear power plants, aging management is an important duty in nuclear industry. To successfully maintain the structural properties of the aging NPPs theoretical and experimental studies are necessary to examine concrete strength and stiffness, water behaviour, volume change of cement paste etc. The largest experimental project in the present times is the double-walled containment building - referred to as VerCors (from Verification Réaliste du Confinement des Réacteurs) of EDF's research and development laboratory. The 30 m height structure was built of about 5000 tonnes of concrete and 700 sensors. This scaled-down size containment was built in 2015 by EDF [1].

To facilitate concrete degradation estimations some models have appeared beside the measurement- and experience-based techniques. Damage Evaluation for Irradiated Concrete (DEVICE) [2] [3] numerical code is composed of a cement-based material model and a one-dimensional deterministic transport code. The code couples cement hydration with moisture, heat and radiation transport. Sobol's method [4] [5] can be applied for radiation damage estimations too. This method runs Monte Carlo simulations to calculate the overstressed concrete ratio. However, our knowledge about radiation damage in concrete structures is still limited, which causes high uncertainties in the models. For this reason, further experimental work is suggested in the field to gain knowledge and support the development of models. This paper can be considered as a preliminary study, which evaluates the concrete degradation experimental possibilities and environment in the Budapest Research Reactor (BRR) [6].

2 EFFECTS OF IRRADIATION ON SHIELDING CONCRETES

Shielding concretes are exposed to three different damaging effects: neutron radiation, gamma-radiation and increased temperature [7]. Though they affect the concrete in different ways, these effects are usually present together, so it is hard to determine which damages are attributed to which effects. This problem was illustrated by Fujiwara et al. [8] (see Figure 1) highlighting the differences of the measurement conditions in the figure of Hilsdorf et al. [9]. A solution for this issue could be the setup of a highly realistic sample environment, though it has many difficulties.

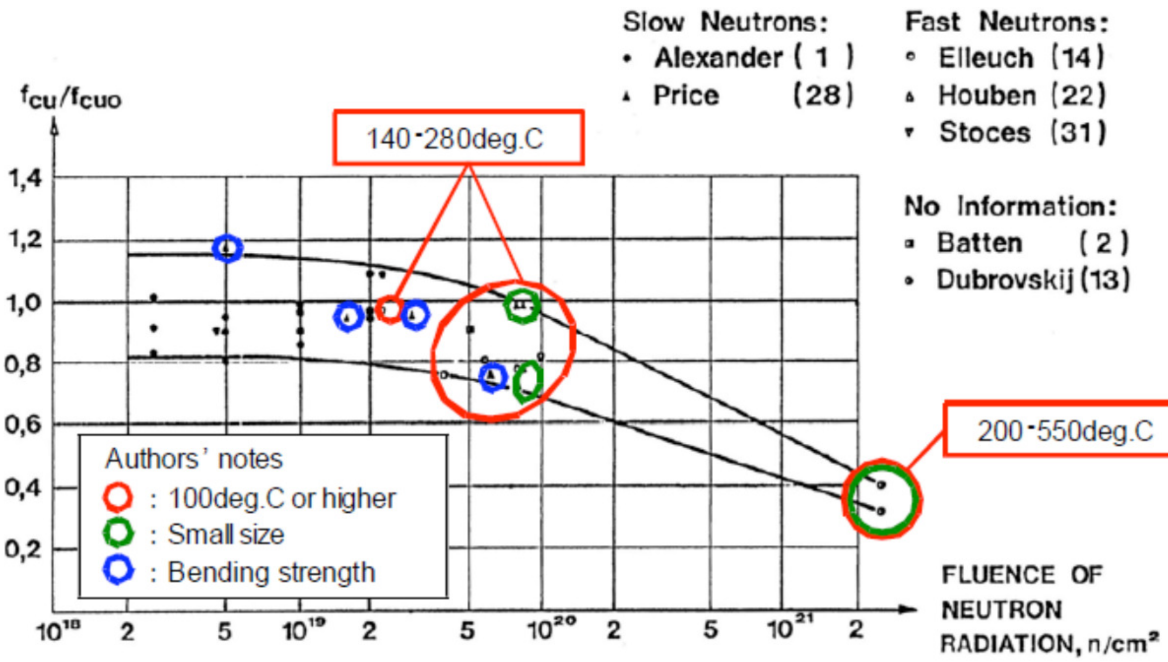

Figure 1: The weak points of the data collected by Hilsdorf et al. [9] on compressive strength measurements of irradiated concrete samples [8]. On the vertical axis f_{cu}/f_{cuo} is the compressive strength of concrete exposed to neutron irradiation (f_{cu}) related to strength of untreated concrete (f_{cuo}).

2.1 The effect of neutron irradiation

Neutrons have more significant effect on dense, well-crystallized materials compared to materials of random structures and high porosity. In concretes, the aggregates form the crystallized phase, thus neutrons cause more distortion and damage to their internal structure than to the concrete paste. These distortions are leading to increase in volume, which can cause cracks in long-term [10].

The first overview about concrete degradation by neutrons was written by Hilsdorf et al. [9]. When they collected the available measurement data, the original graph of Figure 1 was created, where f_{cu}/f_{cuo} on the vertical axis is the compressive strength of concrete exposed to neutron irradiation related to strength of untreated concrete. This study concluded that high neutron fluence above 10^{19} n/cm^2 can lead to decrease of compressive strength. Later publications [11], [12] suggest rather 10^{20}-10^{21} n/cm^2 as threshold neutron fluence for concrete degradation.

2.2 The effect of gamma-rays

Shielding concretes are exposed to gamma radiation originating from the reactor and also from the neutron interactions in the concrete itself. Gamma-rays cause radiolysis, which is the decomposition of water to hydrogen, oxygen and hydrogen peroxide. These decomposed particles can form gas molecules. As most of the water content of concretes is present in the cement paste, gamma radiation rather affects the cement paste than the aggregate minerals [11].

Based on the plotted residual compressive and tensile strength of concrete against gamma-ray dose, Kontani et al. suggested $2*10^8$ Gy as threshold value for concrete degradation caused by gamma radiation in their review [13]. Similarly to that, the report of Electric Power Research Institute suggests 10^8 Gy [14].

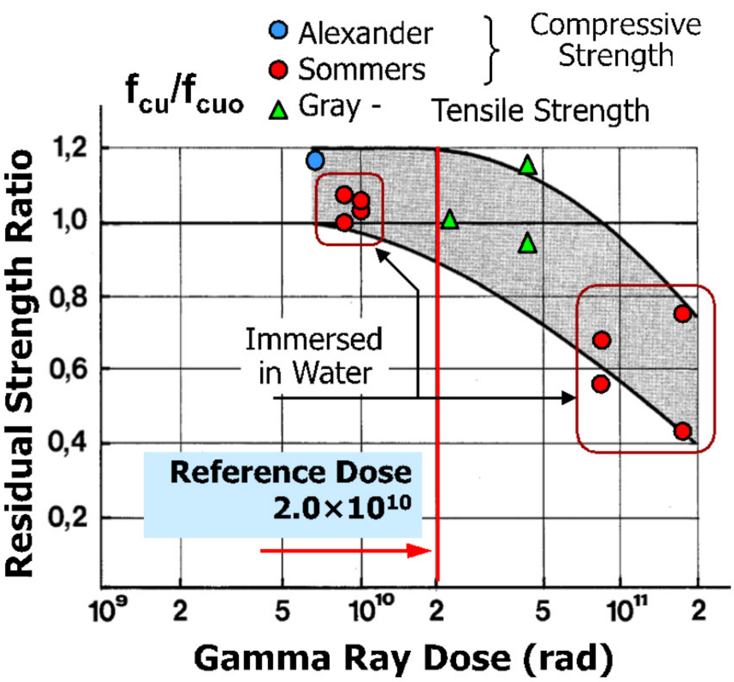

Figure 2: The effect of gamma radiation on the residual compressive strength of concrete [13]. On the vertical axis f_{cu}/f_{cuo} is the compressive strength of concrete exposed to gamma-irradiation (f_{cu}) related to strength of untreated concrete (f_{cuo}).

2.3 The effect of heat

Increased temperature can cause damage in concretes in two ways [15]:

a. If the cement paste and the aggregates have different expansion coefficient, the difference in their expansion can lead to cracks between the two phases.
b. Increased evaporation due to heat exposure leads to gas release, which increase tension inside the concrete. As RIVE (Radiation Induced Volumetric Expansion) causes anisotropic expansion and cracks, increased temperature contributes to the growth of cracks inside the aggregates.

As primary sources of heat neutron- and gamma interactions in the concrete are usually marked. However, Abdo and Amin [16] revealed that gamma heating increased the temperature in the top layer of the concrete with only 1.7 ℃ degree in their experiments, which has no effect on dehydration. Effects of increased temperature generally appear above 70-80 ℃, which barely appears in shielding concretes even with the absorption some of the heat radiation from the reactor. Thus, the effect of heat is usually a side-effect of irradiation experiments in research reactors, where the temperature can increase up to several hundreds of Celsius degrees, though this effect would be better to be eliminated by the control of sample environment.

3 IRRADIATION POSSIBILITIES IN THE BUDAPEST RESEARCH REACTOR

Due to the inhomogeneous structure of concretes, larger sample size is required for degradation tests than in case of homogeneous materials e.g., metals. Partly this is the reason why the effects of irradiation are better studied and described for other reactor materials (reactor pressure vessel (RPV), fuel rods…). Another difficulty of radioactive concrete testing is the arising dust during mechanical tests, which makes it necessary to have a dedicated handling environment including a hot cell and a set of equipment solely for these tests. Furthermore, the necessary cooling time is long too in case of such large and highly radioactive materials. Finally, irradiation characteristics are key factors as well. In this study, focus was set on the irradiation characteristics of the Budapest Research Reactor (BRR) [6] as a neutron source.

Since sample size is a limiting issue, the biggest irradiation options were investigated first. The biggest sample that can be currently irradiated is 50x100 mm. Though certain tests could be performed on this sample size, the standard cracking tests require 150x150 mm concrete samples. One top of that, this irradiation channel is located far from the core which results only a moderate neutron flux: approximately $1.1*10^9$ n/cm^{2}s. With this neutron flux, the reasonably available neutron fluence is around 10^{16} n/cm^2 with 3-4 months of irradiation, which is much lower than the 10^{19} n/cm^2 value - suggested by the literature.

To achieve higher neutron fluxes, samples must be placed closer to the core. There are channels in the reactor core itself, where the maximal sample size could be 32x60 mm. Neutron flux in the 'Fast channel', which is used for Neutron Activation Analysis (NAA) is around $2.5*10^{14}$ neutron/cm^{2}s. Some channels are located around the reactor core with lower, but still high neutron flux. In the 'Thermal channel' of the NAA station the neutron flux is around $3.5*10^{13}$ n/cm^{2}s. With these neutron fluxes, 10^{19} n/cm^2 fluence can be achieved in less than a week [17].

Beside the fluence, the energy distribution of neutron spectrum is important as well, because the energy content of the fluence is directly related to its degradation capability. For this reason, neutron energy distribution in the Fast channel, in the Thermal channel and in a shielding concrete were compared (Figure 3). To generate neutron energy distribution in a shielding concrete MCNP6.2 [18] simulations were carried out. The initial neutron source was an RPV ¼ neutron spectrum from the FISPACT library [19]. After crossing a 15 cm thick steel unit in the model - which is a typical RPV ¾ thickness - neutrons were tallied in the top 10 cm of the shielding, composed of serpentine concrete (composition available: [20]).

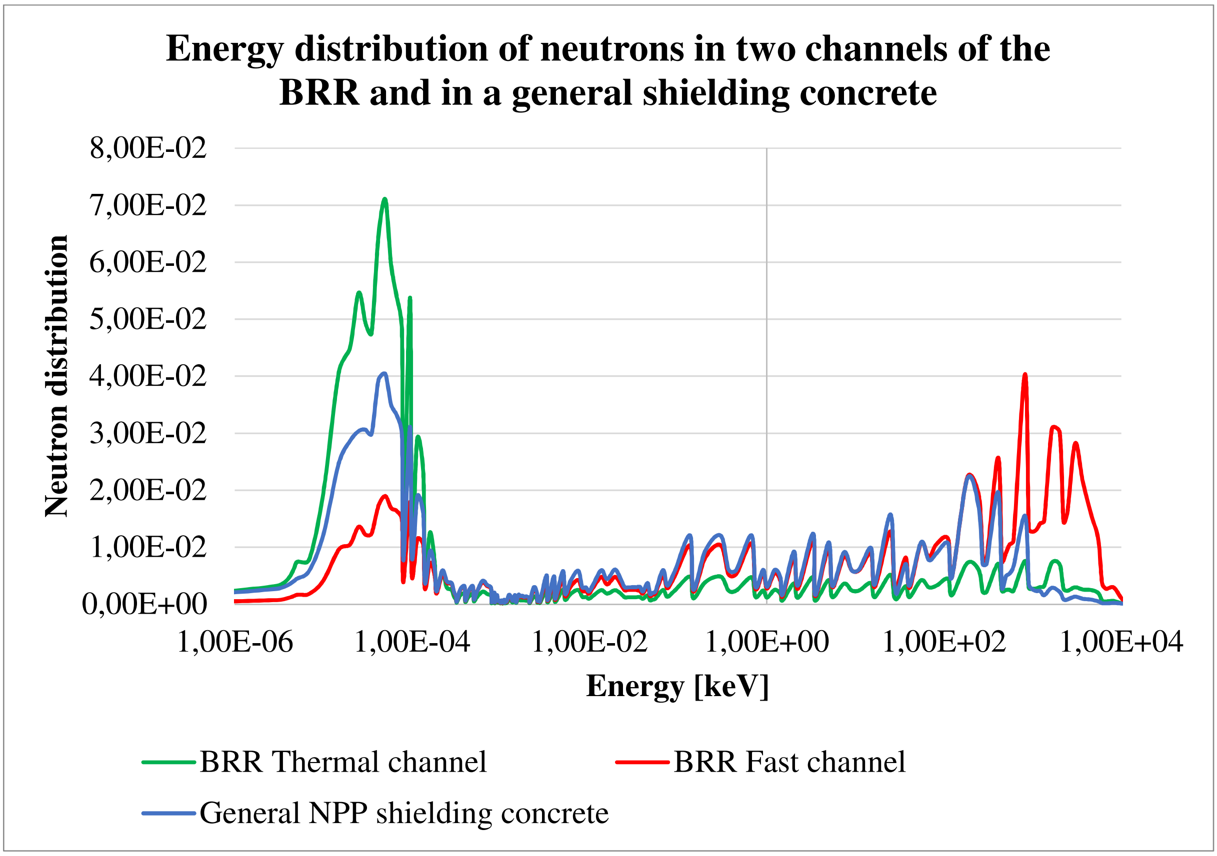

Figure 3: Neutron spectra in the Fast- and Thermal channels of the Budapest Research Reactor (BRR) and in a serpentine shielding concrete of a hypothetical NPP. Latter is a result of an MNCP [18] simulation with an initial neutron spectrum from FISPACT [19] data base.

Analysing the spectra of Figure 3, a maximum can be found in all of the curves around the 25 meV thermal energy. This peak is the highest in case of the Thermal channel, lowest in the Fast one and the peak of the shielding concrete is in between. After that energy range until around 500 keV the neutron spectrum of the shielding concrete is quite similar to that of the Fast channel. Finally, in the highest energies up to around 20 MeV there are only significant amount of neutrons in the Fast channel.

Based on these results, future irradiation degradation experiments are rather suggested to be carried out in the thermalized area of the reactor to avoid overestimations and misleading consequences of the significant portion of high energy neutrons in the reactor core. The presented curves also pose the question if the different energy content of the neutron fluences in shielding concretes and in irradiation channels were taken into consideration (correctly) in the published studies in the field or not.

4 CONCLUSIONS

Since the new nuclear power plants are designed for longer operation periods (60-80 years) than current ones, shielding concretes around the reactor pressure vessel can receive critical neutron fluence, which may cause structural damages to them during their lifetime. Though it is hard to draw strong conclusions based on the currently available scientific literature - because it is almost impossible to distinguish experimentally between the simultaneous effects of neutron radiation, gamma radiation and increased temperature of the sample environment -, very few new scientific results were published on the topic in the past few decades. New measurement results would be important to develop and validate simulation tools, which may open up reliable simulation-based solution for this problem in long-term.

In this study, the Budapest Research Reactor was investigated as a possible location for radiation damage measurements. Preliminary results show that sufficient neutron fluence can be achieved with similar neutron spectrum than that of the shielding concrete of an NPP. However, the standard sample sizes of mechanical property measurements usually exceed the current dimensions of the irradiation channels. Cooling of the sample environment during irradiation would also be advantageous to avoid the inconvenient effects of high temperature. Handling of the highly activated sample is also a challenge in such measurements both technically and financially, because a dedicated hot cell is required, installed with concrete mechanical property (such as compressive and tensile strength) measurement device and remote handling devices.

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