Assessing the Effects of Gamma Irradiation in Concrete

Elena Tajuelo Rodriguez, a William A. Hunnicutt, b Paramita Mondal, b,c Yann Le Pape a

a ORNL, One Bethel Valley Rd, Oak Ridge, TN 37831-6148, tajuelorodr@ornl.gov, lepapeym@ornl.gov
b University of Illinois at Urbana-Champaign, Urbana, IL 61801, hunnic2@illinois.edu, pmondal@illinois.edu

c University of Delaware, 301 Du Pont Hall, Newark, DE 19716, mondal@udel.edu

INTRODUCTION

The US fleet of light water reactors is under consideration for a license renewal that will prolong its lifetime to 80 years. This prolonged operation must be accompanied by proper support to ensure the safety and reliability of all the components in the power plants: reactor pressure vessels, cables and the concrete biological shield. In the case of the concrete bio-shield, a protective wall that also holds structural functions, the exposures to gamma and neutron irradiation will be in the order of 50-200 MGY [1] and 10^{19} n/cm^{2} [1,2] after 80 years of operation. Neutrons are known to affect the aggregates in concrete, causing expansion of the minerals (Radiation Induced Volumetric expansion) [1], and hence the appearance of cracks in both the aggregates and the cement paste. Gamma rays, on the other hand, cause dehydration of the cement paste due to hydrolysis [3].

Within the DOE Light Water Reactor Sustainability Program, much effort has been made to implement models to predict irradiation damage in concrete with neutron fluence [4], but the effect of gamma rays has not been considered yet, due to the lack of available data. Recently developed models have shown that creep (permanent deformation under mechanical stress) delays the onset of radiation damage in concrete [4]. This indicates that the viscous response of cement paste serves to mitigate irradiation damage. Creep was independent of the fluence in these models. However, if gamma rays are considered, it may be possible that the viscous response of the paste changes with exposure, since gamma rays cause dehydration and all the mechanical properties in cement strongly depend on its water content.

When referring to mechanical properties of cement, one of its hydration phases outweights: calcium silicate hydrates, also known as C-S-H in cement chemistry notation. Considered as the glue of cement, these semi-crystalline phases give all the strength to the paste and occupy about 40-50% in volume in hydrated Portland cement. The creep response of C-S-H outlines any other responses from the rest of hydration phases. For all these reasons, C-S-H is a good model system for cement paste.

C-S-H consists of layers with calcium and oxygen with silicate dreierketten type chains attached. Water occupies the interlayer space and other nanometric pores that are the result of the space between different blocks of stacked layers. The silicate species present in these materials are end-chains (Q^1), middle-chains (Q^2) and crosslinks (Q^3).

Two different mechanisms are proposed to account for creep in C-S-H: sliding of particles/sheets with respect to each other [5-8] and stress-induced dissolution-precipitation [9]. The stress relaxation of C-S-H is highly dependent on its water content and the partial removal of interlayer water decreases the capability of the material to relax stresses [5]. Previous studies of stress relaxation in C-(A)-S-H (Aluminum substituted C-S-H) and C-S-H suggested that the stress relaxation depends on the degree of crosslinking between the chains (%Q^1) and/or the mean (alumino)-silicate chain length [10].

To advance the understanding of the possible change in viscous response of C-S-H after gamma irradiation, an experimental program was carried out to irradiate C-S-H pellets of three different Ca/Si ratios: 0.75, 1 and 1.33. The pellets were made compressing 2g of material under 250kN using a 25.4 mm diameter die after having passed the material through a 75μm sieve and conditioned it to 11%RH. At this relative humidity, the material is considered to have only a monolayer of water on the surfaces apart from the interlayer water. The pellets were irradiated in a Co^{60} gamma reactor (J.L. Shepherd Model 109-68 Co-60 unit) to gamma-adsorbed doses of 0.39, 0.77, 1.39 and 2.24 MGY (irradiation times of 2, 4, 7.5 and 12 months). They were contained in stainless steel vials constantly purged with Ar at a pressure of 206 kPa to prevent carbonation. Control samples were placed in the same kind of containers and connected to the Ar flow but out of radiation reach.

The silicate anion structure of the samples was studied with 29Si NMR to detect any changes in silicate mean chain length. If water was removed from the interlayer, this could promote and increase in the degree of crosslinking, and eventually affect the capability of the material to creep due to sliding of its building blocks. The interlayer spacing was obtained by the position of the basal peak with XRD and the total water content with TG. The viscous and elastic responses were tested with a Hysitron nanoindenter to gain information on stress relaxation and Young’s modulus for all doses, as well as creep for the last dose. Results on the first three doses are reported elsewhere [11]. They indicated no significant changes in mechanical behavior, chemical composition, morphology, or water content. Preliminary results on the last dose are discussed in this summary.
RESULTS

The XRD results on the C-S-H samples irradiated to 2.24 MGy showed that the basal spacing of both control and irradiated samples was very similar; ~11.5 Å, ~10.9 Å and ~9.9 Å for Ca/Si ratios of 0.75, 1 and 1.33 respectively. This indicates that no water was removed from the interlayer due to gamma ray adsorption.

The water content of the samples given by TG was even slightly higher for the irradiated samples being 15.7%, 16.8% and 17.1% for Ca/Si ratios of 0.75, 1 and 1.33 respectively. The control samples showed a water weight content of 15.4%, 15.5% and 15.6%.

The silicate anion structure obtained by the deconvolution of the NMR spectra was very similar for irradiated and dosed samples too. The mean silicate chain lengths were in the order of 2 and 4 for Ca/Si ratios of 0.75 and 1 respectively for both dosed and control samples. The samples with Ca/Si ratio of 1.33 exhibited a slightly more significant change in mean silicate chain length that was estimated to a value of 13 for the dosed sample and 9 for the control sample. In general, these results indicate that the adsorbed gamma dose did not affect the chemical structure and stability of the phases, since the water in the interlayer remained after irradiation, and the silicate anion structure was comparable for both irradiated and control samples.

The creep compliance for the irradiated and dosed samples with Ca/Si = 0.75 is shown in Fig. 1. The dashed lines are the 95% confidence intervals. Both irradiated and control samples show creep compliance in the same range and the mean curves are within the confidence intervals of the other sample. This was also the case for the other Ca/Si ratios. This means there is no significant change in creep behavior between irradiated and dosed samples. The same occurred for the stress relaxation and Young’s modulus.

![Creep Compliance of C-S-H Sample](image)

Fig. 1. Creep compliance of the C-S-H sample with Ca/Si ratio of 0.75 irradiated to a gamma dose of 2.24 MGy (12 months) and the corresponding control sample.

Therefore, the adsorbed dose of gamma irradiation up to 2.24 MGy showed no effect on mechanical properties of C-S-H. Further experiments are being planned at the GIF facility at Sandia National Lab to irradiate C-S-H to a dose of 200 MGy (comparable to the expected dose at 80 years).

REFERENCES


ACKNOWLEDGMENTS

Efforts were supported by the U.S. DOE, Office of Nuclear Energy, Light Water Reactor Sustainability Program, NSUF RTE proposals and by an appointment to the Oak Ridge National Laboratory ASTRO Program, sponsored by the U.S. Department of Energy and administered by the Oak Ridge Institute for Science and Education.