Towards a New UHPC Mix Design for Nuclear Applications: A Review Study

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Abstract

This paper presents a review of academic literature regarding various additives and modifications of concrete and cementitious materials in general, and ultra-high performance concrete (UHPC) in particular, for applications in nuclear settings. The overall goal of the review is to inform future robust UHPC mix development to be specifically used for nuclear waste storage facilities. There is an urgent need for longer term storage which calls for a new generation of extremely durable spent nuclear fuel (SNF) storage facilities that could leverage UHPC. In nuclear settings, concrete is not only a structural material but it is used for radiation shielding and public safety. Research into general concrete additives and modifications for nuclear applications has shown the importance of incorporating heavy aggregates and boron compounds to increase shielding properties against gamma and neutron radiation. Over the past decades, various conventional concrete (CC) mixtures have been proposed to improve radiation shielding. However, less number of studies explored radiation attenuation performance of UHPC mixes. UHPC has significant advantages over CC in hostile environments, and it is expected that such features can manifest itself in nuclear applications with proper mix design and adjustments. The paper summarizes studies on radiation attenuation performance of UHPC with and without specific mix adjustments, identifies existing knowledge gaps, and proposes theoretical UHPC mix additives for future storage facilities of SNF.

Keywords: UHPC, spent nuclear fuel, dry storage facilities, radiation attenuation

1. Introduction

In nuclear power plants, nuclear fuel that has been used and removed from the reactor core, known as spent nuclear fuel (SNF), is stored in spent fuel pools. Since the 1980s, after reaching the pools' capacities, reactor sites started to place SNF in dry cask storage systems (DCSSs). Initially, it was anticipated that a permanent repository in Yucca Mountain, Nevada, would be available within 20 to 40 years after the first casks were loaded. However, the project encountered significant public opposition, and, eventually, lost federal funding in the early 2000s. Up to this date, there are no defined long-term disposal and reprocessing path that policymakers would agree on. None of the existing reactors have capacities to store SNF they will produce over their lifetime (Macfarlane 2001). Eventually, all the DCSSs from all the reactor sites will be shipped to the defined permanent geological repository or one or more consolidated sites which might require decades (APS 2007). It is expected that DCSSs are going to be used for fuel storage longer than it was foreseen.

Any design of a DCSS must undergo a licensing process through the Nuclear Regulatory Commission (NRC). The initial certification period is typically 20 years. Overall, the system must be designed to ensure public health and safety by maintaining radiation shielding, subcriticality control, passive cooling, confinement, and structural integrity under design-based conditions of normal and off-normal operations, including accidents. At the end of the certification period, a cask vendor must demonstrate that the loaded dry cask storage can continue to meet the technical requirements for another approval period (up to 40 years).

Many of the existing designs of DCSSs involve the use of a concrete horizontal or vertical shielding structure, which is referred to differently in the literature as a cask, module, or overpack. Because of the extended service life and environmental exposure, degradation of concrete structures is likely to happen. Since the 2000s, limited inspections performed by the NRC confirmed incidents of concrete degradation in DCSSs (Chowdhury et al. 2016). A well-known example is related to extensive damage in horizontal concrete modules located on the site of Three Mile Island Unit 2 due to cracking from freeze-thaw cycles (NRC 2012). Two examples from one of the damaged horizontal storage modules, HSM 05 pre-repair, are presented in Figure 1.

Figure 1 Two examples from north west (left) and north east (right) corner roof damage of a horizontal storage module at the site of Three Mile Island Unit 2 (NRC 2012)

New durable cask designs that can be used to extend regulated design life of storages and help reduce cumbersome and expensive license renewals are yet to be properly explored. Minimal or free maintenance is desired since monitoring activities are limited due to geometry, space limitations, and high radiation risks in dry storage systems. Thus, longevity for new designs and cost-effectiveness over the structure lifetime are key priorities for vendors and reactor operators.

Recently, big advancements have been made in the development of advanced construction materials; one of which is ultra-high performance concrete (UHPC). UHPC has a compressive strength that can exceed 150 MPa (21.8 ksi). Due to the inclusion of steel fiber reinforcement, UHPC is ductile and can reach tensile strength exceeding 15 MPa (2.2 ksi) and flexural strength exceeding 50 MPa (7.3 ksi) (Fehling et al. 2014). UHPC has higher durability than conventional concrete due to the use of steel fibers that bridge cracks as well as the dense microstructure (Akeed et al. 2022). Since UHPC exhibits superior mechanical and durability characteristics, it has a potential for new applications including nuclear structures. Thus, the idea of using new generation of extremely durable, cost-effective, and compact UHPC overpacks capable of securing all the safety functions while requiring minimal or free maintenance is what motivates this research.

Since concrete structures in DCSSs are used for both structural integrity as well as for radiation shielding, UHPC's radiation attenuation properties are of high importance. Extensive research has been done over the years on development of radiation shielding concrete by incorporating heavy aggregates and boron additives. However, only a limited number of studies have explored the effect of these modifications on UHPC's performance, which are summarized in this paper. Following the brief conducted literature review, the paper proposes a theoretical UHPC mix adjustments to enable the use of UHPC for future dry storage of SNF.

2. UHPC Radiation Attenuation

Attenuation is the process by which the number of particles or photons is reduced by absorption or scattering when passing through a body of matter. In nuclear settings, there are four principal forms of ionizing radiation that can be encountered in SNF: alpha particles, beta particles, gamma rays, and neutrons. Overall, any shield that is capable to attenuate gamma rays and neutrons will be effective for attenuating other radiations (El-Khayatt 2011). In other words, alpha and beta particles shielding is usually not a concern when compared to gamma and neutron radiations. The latter ones have also significant contribution to concrete degradation in SNF dry storage, and as such, only gamma and neutron radiations are concerned in the brief review presented herein.

2.2. Gamma-Rays Attenuation

Gamma rays are high-energy, high-penetration form of electromagnetic radiation. While passing through an object, some of the gamma-ray photons interact with the material. A linear attenuation coefficient (μ) is a factor that indicates how much attenuation will take place per a unit length (typically per cm). The intensity of photons that pass through a sample can be experimentally measured using a narrow beam of mono-energetic photons. There are several testing facilities in academic institutions as well as national laboratories that can be used for experimentally determining attenuation coefficients. One example is presented in Figure 2 for completeness. Commonly, cesium-137 (^{137}Cs) and cobalt-60 (^{60}Co) gamma-ray sources are utilized in such experiments.

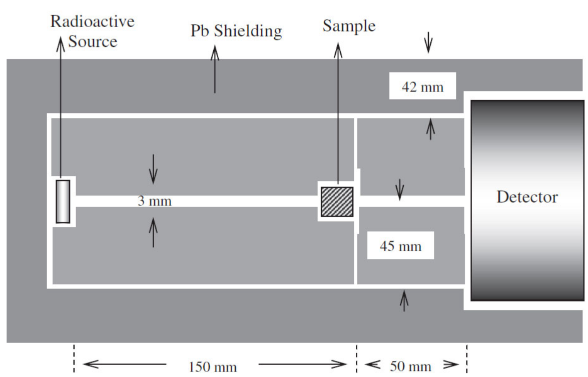

Figure 2 One example of experimental setup for measuring attenuation coefficients (Yılmaz et al. 2011)

A relationship between the incident intensity (I_0) of photons, transmitted intensity (I), and thickness of a material sample (x) can described by Equation (1).

$$I = I_0 e^{-\mu x}. \quad (1)$$

Attenuation coefficients of gamma rays through the shielding material can be attained theoretically by WinXcom or its older version, XCOM program. Several studies have looked into determining the linear attenuation coefficients of UHPC, which are summarized in Table 1.

Table 1 Summary of gamma rays linear attenuation coefficients of UHPC reported in the literature

Study	Linear attenuation coefficient, μ (cm^{-1})		
Experiments	Gamma-ray source		
	^{137}Cs	^{60}Co	
Azreen et al. (2018)	0.155	0.096	
Tufekci and Gokce (2018): Reported for different w/b ratios of: 0.18, 0.24, and 0.36	0.173 0.177 0.169	0.116 0.117 0.111	
Rashid et al. (2020)	0.146	0.091	
Khan et al. (2020)	0.187	−	
Han et al. (2022)	0.154	−	
Theoretical calculations	Decay energy (MeV)		
	0.662	1.173	1.332
Gökçe et al. (2018)	≈ 0.202	≈ 0.152	≈ 0.144

2.2. Neutron Attenuation

Neutrons radiation is a form of ionizing radiation which occurs due to a release of free neutrons; high-speed neutral particles with an exceptional ability to penetrate materials. Primarily, neutron radiation occurs inside a nuclear reactor during nuclear fission. Neutrons can be classified by their energy into several different categories. In general, there is no standard definition for such categories. Nevertheless, a popular classification is the one presented by Hilsdorf et al. (1978), which includes: thermal neutrons (Energy < 1 eV), epithermal neutrons (1 eV < Energy < 0.1 MeV), and fast neutrons (Energy > 0.1 MeV).

A typical setup for a neutron attenuation test is somewhat like the one presented in Figure 2 for gamma rays attenuation. For such experiments, two types of neutron radiation sources are commonly used: plutonium-beryllium (Pu-Be) and americium-beryllium (Am-Be) sources. To obtain a beam of thermal neutrons, a polypropylene block can be placed in front of the neutron radiation source to moderate the fast neutrons, and hence, produce slower thermal neutrons.

For a fast neutron, the probability that it undergoes a first collision is called removal macroscopic cross section (Σ_R), which is usually reported in cm^{-1}. The attenuation, or "removal" in this case, means removal from the fast group. The relationship between the emitted and transmitted intensities of fast neutrons (I_0 and I, respectively) as a function of sample thickness can be expressed as shown in Equation (2). It is noted that because of the variety of interaction processes that happen between matter and neutrons, the calculation in Equation (2) can be only considered approximate. The macroscopic cross-section for thermal neutrons can be evaluated using similar equation as for the fast neutrons.

$$I = I_0 e^{-\Sigma_R x}. \tag{2}$$

As for UHPC, there is almost no previous research or literature body on neutron attenuation performance of UHPC. Only recently, this topic has started to gain scientific attention. One study by Arfa et al. (2022) have determined the values of macroscopic cross-sections for thermal and fast neutrons in UHPC concrete samples with 3% volume of steel fibers as 0.0839 and 0.0357 cm^{-1}, respectively. Thus, for UHPC to be considered further for nuclear applications, there is an immediate knowledge gap and urgent need to investigate and characterize the neutron attenuation of different types of UHPC with different steel fibers ratio along with any future mix modifications.

3. UHPC Mix Modifications

Few preliminary studies have considered UHPC mix modifications to improve its nuclear properties and radiation shielding, mostly for gamma radiations. However, larger number of studies explored many add-on materials for conventional concrete to make it suitable for nuclear applications. Many of those studies are likely viable to extend to UHPC as well. This section provides a brief summary of mixture additives that have been considered for UHPC as well as promising ones that can likely enhance UHPC gamma and neutron shielding properties.

3.1 Gamma rays

Three principal types of gamma-ray interactions with matter can be distinguished: photoelectric effect, Compton effect, and pair production. The relative importance of these processes depends on the photon energy level and the atomic number of the material. Since each process involves interactions with electrons, the goal is to maximize the density of electrons within material. For conventional concrete, increasing the density by better compaction can be complicated because of the presence of coarse aggregates and their possible segregation. A more common approach is to use high-density materials that contain elements with high atomic numbers. Extensive research has been done over the years on development of concrete with heavyweight coarse aggregates like magnetite, barite, and hematite to improve gamma-ray attenuation performance (Abdullah et al. 2022). The goal is to produce concrete with density higher than 2600 kg/m^3 which is commonly referred to as a heavyweight concrete. Based on such studies for conventional concrete, researchers have attempted to incorporate heavy fine aggregates in UHPC.

Rashid et al. (2020) have reported linear attenuation coefficients for UHPC containing magnetite fine aggregate for ^{137}Cs and ^{60}Co sources as 0.197 and 0.122 cm^{-1}, respectively. Han et al. (2022) evaluated the effect of replacement of sand in UHPC with 20, 40, 60, 80, and 100% of magnetite. For ^{137}Cs source, the reported linear attenuation coefficients ranged from 0.167 to 0.202 cm^{-1} for different magnetite ratios. In both studies, inclusion of magnetite effectively enhanced the UHPC samples attenuation of gamma-rays. However, the compressive strengths of the UHPC mixes were affected. For specimens with full substitution of sand by magnetite, the reported compressive strength reductions were 13% (Rashid et al. 2020) and 4.5% (Han et al. 2022).

Incorporation of barite fine aggregate into UHPC was evaluated by Tufekci and Gokce (2018), Gökçe et al. (2018), and Azreen et al. (2020). All studies reported a significant improvement of gamma-ray attenuation performance. The compressive strength reductions of the UHPC samples with barite ranged from 15 to 21%. Gökçe et al. (2018) have also evaluated impact on flexural strength and modulus of elasticity and reported a reduction equal to 36 and 16%, respectively.

For hematite UHPC specimens, attained linear attenuation coefficients were 0.165 and 0.108 cm^{-1} for ^{137}Cs and ^{60}Co sources, respectively, as reported by Azreen et al. (2020). The samples reached compressive strength of 149 MPa (21.6 ksi) which was 10% lower than compressive strength of UHPC containing sand as fine aggregate. Khan et al. (2020) studied the heavyweight UHPC incorporating hematite powder. Five dosages of hematite powder were considered for the experiment as a partial replacement of sand (10, 20, 30, 40, and 50%). It was reported that addition of hematite powder improved the compressive and flexural strengths, increased the dry density, and did not adversely impact the modulus of elasticity of UHPC mixes. For the radiation shielding tests, the specimens were irradiated with ^{137}Cs gamma-ray source. By incorporating regression analysis, an empirical equation for 99% attenuation of gamma-ray photons and required thickness in terms of dry density was obtained. The authors compared the required thickness of UHPC and heavyweight conventional concrete at the same dry densities (from 2600 to 3000 kg/m^3) using the developed equation along versus the equation proposed by Azeez et al. (2019) for conventional concrete. As shown in Figure 3, the radiation attenuation performance is 40% higher for UHPC when compared to conventional concrete (indicated as CC in the figure), which is presented by the smaller required thickness for case of UHPC.

In summary, the inclusion of hematite fine aggregate into UHPC mix improves its gamma-ray attenuation properties and has smaller or almost no adverse effects on mechanical properties than inclusion of barite and magnetite. As such, hematite is recommended as a promising additive for UHPC to specifically use in future nuclear waste storage facilities.

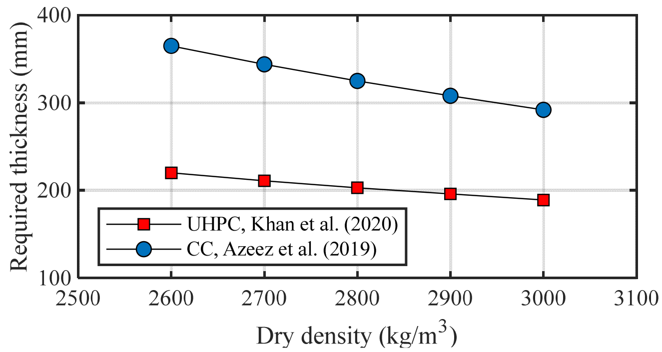

Figure 3 Required thicknesses for UHPC and conventional concrete (CC) for 99% attenuation of gamma rays (figure adopted from Khan et al. 2020)

3.2 Neutrons

Fast neutrons are not easy to attenuate, as they should be slowed down first. To slow down fast as well as intermediate energy neutrons, inelastic scattering by heavy elements (e.g. iron) and elastic scattering by hydrogen is considered to be effective (Akkurt and El-Khayatt 2013). The resultant slower neutrons should be then captured. During radiative capture, a neutron is captured by the nucleus, and as a result, one or more gamma rays, namely capture gamma rays, are emitted. To reduce or suppress the production of the capture gamma radiation, elements with high neutron capture cross section maybe introduced. One of the common materials with high capture cross-section is boron. Accordingly, and in a general sense, effective neutron attenuation can be achieved by having a neutron moderator and a neutron absorber.

Serpentine and limonite are natural aggregates containing hydrogen. These aggregates were found to be effective neutron moderators in conventional concrete applications (Kansouh 2012; Oto et al. 2015). Meanwhile, boron works as a suppressor of capture gamma-rays, and as such, boron has become a common additive in conventional concrete to improve its neutron absorption performance (Kharita et al. 2011; Rajadesingu and Arunachalam 2020; Zayed et al. 2021). Such modifications are expected to improve UHPC neutron shielding. However, future work is needed to first confirm the effectiveness of such modifications on UHPC neutron shielding, and next to evaluate the effects of including the discussed additives on UHPC properties.

4. Concluding Remarks

The brief literature review presented in this paper serves as an initial step towards rethinking of the existing designs of the nuclear waste storage facilities by incorporating UHPC. Since radiation attenuation properties are important for materials used in nuclear settings, studies on radiation attenuation performance of UHPC are summarized, which mostly considered only gamma radiations. Several knowledge gaps have been identified and mentioned in the different sections.

For future UHPC mix improvement, and based on the existing research, UHPC with hematite inclusion is concluded to be a promising solution for attenuation of gamma radiation. Meanwhile, and due to the lack of research on modification of UHPC for neutron attenuation, theoretical mix adjustment using mostly boron is suggested based on the works for conventional concrete.

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