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Cement-waste interactions: Hardening self-compacting mortar exposed to gamma radiation

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ABSTRACT

For the disposal of high level radioactive waste and for attenuation of the emitted radiation, the Belgian supercontainer concept considers the use of cylindrical concrete containers: the radwaste (encapsulated in a canister and stainless steel overpack) is embedded in a hardened self-compacting concrete buffer, and for closure of the supercontainer the remaining gap is filled by casting a self-compacting mortar (filler and lid). As a consequence, this cementitious layer, surrounding the radwaste, will be exposed immediately to the heat-emitting radioactive waste and gamma radiation with dose rates up to 20 Gy/h during hardening and hydration of the cementitious matrix.

In this research study, the effect of gamma radiation on the mechanical properties (e.g. compressive strength) and the microstructure of the cementitious samples is investigated thoroughly. By means of compressive strength determination and by analysing the microstructure of the cementitious samples, the effect of gamma radiation during the hardening process of the samples is identified. Small self-compacting mortar cubes were cast and irradiated immediately by gamma rays during hardening. The effect of the total absorbed dose (Gy) and the applied dose rate (Gy/h), in combination with different hardening times at first exposure and total irradiation times is determined. Furthermore, the impact of the composition of the cementitious mortar (e.g. by changing the cement type and the water-to-cement ratio (W/C-ratio)) is investigated.

Throughout the test program it was found that a strength loss due to gamma irradiation can be expected, influenced by the total received dose and by the applied dose rate. Furthermore, the age at which irradiation starts (hardening time at first exposure), plays a role in the effect of the gamma irradiation. A correlation between the strength of the mortar samples and its microstructure is found by means of fluorescence microscopy on thin sections and nitrogen adsorption tests: by applying gamma radiation the capillary porosity, the pore volume distribution and the specific surface of the pores is affected. Scanning electron microscopy (SEM) also revealed a change in microstructure due to gamma radiation.

1. Introduction

Worldwide cementitious barriers are being considered for shielding structures for nuclear applications: (i) shielding structures incorporated into the design of Nuclear Power Plants (NPP's, Ouda, 2015), (ii) shielding structures for temporary storage and long-term disposal of vitrified waste and spent fuel assemblies, (iii) shielding structures for medical applications and facilities or (iv) in case of sudden radioactive hazard (e.g. Fukushima, March 2011).

The disposal of radioactive waste nowadays is a very hot topic, and is being investigated worldwide for more than five decades. For the disposal of highly active, heat-emitting C-level waste (High Level Waste, HLW) and the spent fuel (SF) assemblies, the use of cylindrical concrete supercontainers (Fig. 1), deeply disposed in clayey host rock layers, is considered as the reference design in Belgium (Bel et al., 2005). Understanding the effect of irradiation on the concrete, and identifying the interactions between the waste

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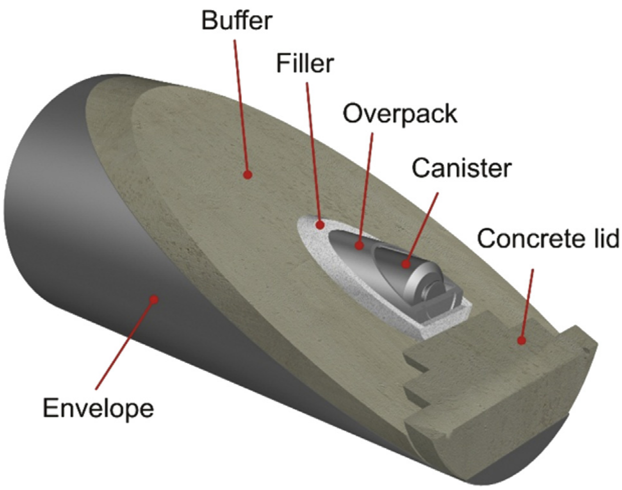

Fig. 1. The Belgian supercontainer concept – An engineered barrier system using cementitious layers.

and the cementitious barriers, is one of the key aspects that has to be clarified in order to increase the predictability and confidence of the Belgian radwaste-disposal concept. The supercontainer is a stepwise fabricated engineered barrier system (Fig. 2), based on the use of an integrated vitrified waste package composed of a carbon steel overpack surrounded by a concrete buffer based on Ordinary Portland Cement (OPC) (Craeye, 2010). Self-Compacting Concrete (SCC) (De Schutter et al., 2008) is considered for the buffer. Once the buffer is cast and sufficiently hardened (Fig. 2, Stage 1), the radwaste is placed inside the buffer (Fig. 2, Stage 2), and the remaining gap is filled by casting a fresh self-compacting mortar (Fig. 2, Stage 3 and Stage 4). Fabrication stages 3&4 need to be conducted in a shielded environment, called 'hot cell', and the cementitious barriers cast in those two stages are irradiated during hardening, whereas the concrete buffer is already in a hardened state. Only gamma irradiation has to be taken into account as alpha and beta radiation is blocked by the overpack, and the impact of the neutrons can be neglected (Wickham et al., 2004). According to Poyet (2007) the cementitious barriers, in direct contact with the overpack, are subjected to a dose rate inferior to 23 Gy/h.

The effect of gamma radiation on the strength properties and microstructure development of hardening cementitious materials is unknown at this time, and needs to be investigated to increase the reliability of the supercontainer during its lifetime. In this original paper, the effect of gamma radiation on the strength and the microstructure of the filler barrier (Figs. 1 and 2, Stage 3) is studied, both for low as for high level dose rates. The goal is to identify and quantify the interaction of gamma radiation with the microstructure of self-compacting the mortar during hydration of the cement and hardening of the samples. By means of compressive strength determination and by analysing the microstructure of the cementitious samples, the effect of gamma radiation during the hardening process of the samples is identified. Small self-compacting mortar cubes were cast and irradiated immediately by gamma rays during hardening. The effect of the total absorbed dose (Gy) and the applied dose rate (Gy/h), in combination with different hardening times at first exposure and total irradiation times is determined. Furthermore, the impact of the composition of the cementitious mortar (e.g. by changing the cement type and the water-to-cement ratio (W/C-ratio)) is investigated.

2. Cement-waste interactions: a literature review

In the past, research related to cement-waste interactions was conducted, especially regarding computational and predictive modelling of radiation damage to concrete and assessment of the effects of radiation on concrete durability for concrete for nuclear purposes (William et al., 2013). Concrete, intended for nuclear purposes, is subjected to two types of radiation: gamma rays and neutrons which are electrically neutral. These two types of radiation interfere differently with concrete structures, and some concrete types are more vulnerable to one type or the other, depending on the constituent materials used in the composition (William et al., 2013). For the supercontainer concept, only gamma-rays have to be considered, as mentioned by Wickham et al. (2004): they are energetic photons with an energy level >0.1 MeV, with electrically non-charged particles. In this study, the focus lies on interactions and the effects of gamma radiation on the mechanical properties, physical properties and microstructure of the subjected mortar. According to William et al. (2013) radiation has an effect on the shielding material, and for radiation shielding, a high transfer of energy corresponds to a more effective radiation shield. Due to this effective radiation shielding capacity of concrete and similar cementitious materials, a change of mechanical and physical properties of the shielding material, is likely to occur.

According to Bouniol (2004) the most important effect of irradiation of cementitious materials by gamma rays is the contribution to the gas production in the cement matrix due to the radiolysis of the pore water, which can lead towards a detrimental internal gas-pressure build-up, leading towards cracking of the buffer of the supercontainer. Furthermore, a redistribution or reorganization of the pores of irradiated hardened paste samples is found by means of mercury porosimetric measurements.

Vodák et al. (2005) found a strength loss of about 10% due to the application of gamma-radiation, starting from a total received dose of $3 \cdot 10^5$ Gy. The main mechanism leading towards the devaluation of the mechanical properties is the formation of calcite ($CaCO_3$), decreasing both the strength of the cementitious material and the size of the pore space. With increasing dose of radiation, the porosity and specific pore surface area of the microstructure are reduced and due to the formation of calcite micro-cracking occurs near the pores which may contribute to the observed strength reduction. Furthermore, the gamma radiation accelerates the

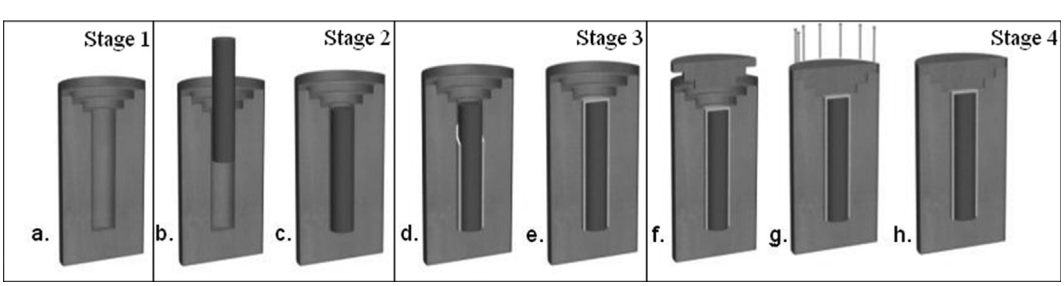

Fig. 2. Stepwise fabrication of the supercontainer – hardening cementitious layers in steps d to h are in direct contact with the gamma radiation.