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Composite materials with primary lead slag content: Application in gamma radiation shielding and waste encapsulation fields

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This paper presents a study on the valorization of primary lead slag as substituent of aggregate in concrete for gamma radiation shielding and as a component of cementitious binders with application in low level radioactive solid waste encapsulation.

The first part of the paper outlines the impact of primary lead slag on the compressive strength and gamma radiation shielding properties of concrete. The presence of the primary lead slag as aggregate substituent in concrete, along with steel shot and barite, enhances the density and shielding efficiency against gamma radiation for Ir^{192}, Cs^{137}, Co^{60} radioactive elements with average gamma energies of 0.37 MeV, 0.662 MeV and 1.25 MeV. The results showed that even the presence of primary lead slag led to a decrease in compressive strength and all the tested concretes exhibited compressive strengths higher than 50 MPa. The utilization of primary lead slag could be recommended as aggregate in heavyweight concrete.

The second part of the paper focuses on the experimental investigation of the physic-mechanical properties and low level radioactive solid waste encapsulation capacity of grout based on binders with primary lead slag content. Two binders (30% and 50% lead slag content) were used in cementitious grouts with different binder:sand ratio (1:1, 1:1.5 and 1:2), two types of sand (river sand or crushed concrete sand), different grain size distribution of sand and water/binder ratio of 0.3. It was observed that the grouts exhibit compressive strengths higher than 30 MPa, regardless of the binder:sand ratio. The increase in the binder:sand ratio influenced negatively the flow ability and positively the drying shrinkage. The low level radioactive solid waste encapsulation capacity of studied grout was lower than that of the reference grout with CRT glass waste.

1. Introduction

Waste management is a major challenge for environmental protection and natural resources conservation. Due to the increased requirements to reduce waste amounts and recycle as much as possible, the construction industry has proposed methods to achieve these goals (Arribas et al., 2014; Huang et al., 2015). Some of the advantages gained when utilizing different industrial wastes are: eliminate the costs of disposing, minimize air and soil pollution problems, conservation of natural resources, and reduce the cost of building materials and road construction materials.

The concrete is a versatile material which consists of at least three quarters aggregates. The aggregate influences the physical and mechanical properties of concrete, as well as the shielding characteristics of concrete. Heavyweight concrete has a density higher than 2600 kg/m^3 and it is widely used in nuclear facilities, radio therapy rooms, and for storing and transporting short lived low and intermediate level radioactive wastes (Waly and Bourham, 2015). To achieving high density the concrete must contains heavy aggregates (density higher than 3 g/cm^3). The most common natural aggregates used to achieve concrete with high density are minerals as magnetite, hematite, limonite and barite.

The natural aggregate can be replaced by wastes, such is primary lead slag. The density of primary lead slag (3.31 g/cm^3) is comparable to that of aggregates used in heavyweight concrete. The primary lead slag is a sub product in lead production from

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sulfide ore containing small amounts of copper, iron, zinc, precious metals, and other trace elements. The processing of lead concentrate into metallurgical lead involves three major steps: sintering, reduction, and refining (www.epa.gov). The resulting slag is usually dumped and in many cases, has not been treated in order to recover valuable metals such as silver, zinc and rare earth elements (indium, gallium) (Roesler, 2015). The quantity of lead and zinc slag produced and dumped every year in Europe is about 1 Mt (Mandin et al., 1997). Reusing such waste in heavyweight concrete is a way to reduce the environmental impact and protect natural resources. All these make primary lead slag an interesting material and environmental friendly alternative since it may have a positive influence on the shielding properties. The primary lead slag waste has not been extensively tested for valorization. In this context, the main goal of the study presented in this article is to investigate the influence of primary lead slag on the shielding and mechanical properties of heavyweight concrete. Moreover, the primary lead slag was used as addition to cement in order to obtain binders for grouts with application in low level radioactive solid waste (LLW) encapsulation, considering that cementitious systems are recommended for this process.

2. Research background

A large number of studies related to design, development and characterization of heavyweight concrete with common aggregates are available in the literature. In recent years, there has been a need to develop heavyweight concrete utilizing cheaper materials, preferably byproducts. There are several reports of experimental work on common and potential alternative aggregates influence on the heavyweight concrete properties.

Saidani et al. (2015) substituted siliceous sand for barite and observed that the addition of barite powder resulted in lower shrinkage and slightly decreased compressive strength, for about 10% for 100% sand substituted for barite.

Mostofinejad et al. (2012) investigated the effect of water/cementitious materials ratio, type of aggregate (limestone or barite), content of aggregate, cement and pozzolanic material (silica fume) on the gamma radiation absorption and compressive strength of concrete. They concluded that the concrete with limestone aggregates had higher compressive strength than concretes with barite aggregate at water/cementitious material ratio of 0.35, 0.40 because of lower strength of barite. Moreover, γ-rays attenuation coefficient of concrete with barite is nearly 30% higher than that of concrete with limestone aggregate.

Esen and Yilmazer (2010, 2011) observed a decreased compressive strength and an increased shielding characteristics, unit weight, ultrasound pulse velocity and modulus of elasticity of concrete with barite content.

Gencel et al. (2011) studied the influence of hematite addition on gamma and neutron shielding characteristics and mechanical properties of concretes by experimentally investigation compared to Monte Carlo simulation. The results showed that compressive strength increase with hematite content. Moreover, studied concretes containing hematite exhibited gamma-ray shielding capacity.

Esen and Doğan (2017) concluded that heavyweight concrete with siderite aggregate can be successfully used as a radiation shielding material for all industrial and sanitary infrastructures subjected to radioactive radiation.

Tufekci and Gokce (2017) investigated the mechanical properties of high performance heavyweight concrete with content of novel and inexpensive byproduct called granulated ferrous waste. The studied concrete contained three different aggregates (silica sand and quartz powder blend, steel fiber, granulated ferrous waste) and steel fibers. The results showed that concrete with granulated ferrous waste content is highly flowable, denser and had increased compressive and flexural strengths.

González-Ortega et al. (2014) studied radiological and mechanical properties of concrete with EAF steel slag in comparison with the concrete with limestone aggregate and barite concrete. The authors concluded that the EAF steel slag increased the density of concrete compared to concrete with limestone aggregate, exhibit compressive strength similar to conventional concrete but higher than barite concrete. The attenuation coefficient of gamma rays was higher than that of conventional concrete and smaller than value corresponding to barite concrete.

Maslehuddin et al. (2013) observed that the concrete with 50% EAF steel slag and 50% steel shot meet the requirements related to weight and shielding properties.

Ling and Poon (2012) found that it is technically feasible to use barite and treated funnel glass as replacement of natural aggregates in heavyweight concrete for radiation shielding construction.

Cullu and Ertas (2016) studied the effect of the lead mine waste on the radiation shielding properties and compressive strength. The results revealed that the concrete with lead mine aggregate do not absorb gamma radiation as 100% barite concrete.

Alwaeli (2013, 2017) investigated the effect of scale and granulated lead-zinc slag wastes as substitutes for sand on the gamma radiation shielding and compressive strength of hybrid concrete. The results demonstrated that the hybrid concrete with 100% substituted sand by lead-zinc slag (50% scale lead-zinc slag and 50% granulated lead-zinc slag) exhibit a higher linear attenuation coefficient of gamma radiation in comparison with conventional concrete.

Ajorloo et al. (2016) examined the influence of lead slag produced in the battery recycling process as substituent of natural aggregates in concrete. The results obtained by replacing 40–60% natural aggregate by lead slag showed that increasing the amount of secondary lead slag rose the density, mechanical strength and gamma-ray attenuation coefficient.

Regarding primary lead slag, the research carried out so far relates mainly to characterization of the leaching behavior of waste and waste-containing materials. The stability of the primary lead slag in conditions that simulate acidic and alkaline environments was studied by Lima and Bernardez (de Andrade Lima and Bernardez, 2013). The results lead to the next conclusions: in acidic environment – Ca, Fe and Si were strongly solubilized; Pb and Zn solubilization was significant below $pH = 3$ while in purified water – Ca and Si were solubilized, unlike Pb, Fe, and Zn which were not detected in the solution.

Barna et al. (2004) introduced primary lead slag as a partial substitute for sand in two road materials (namely sand-cement and sand-bitumen). The road materials were subjected to monolithic leaching test under intermittent wetting conditions and saturated scenario.

Primary lead slag can be used as aggregate in heavyweight concrete as well as component in geopolymers. Onisei et al. (Onisei et al., 2012) reported on the synthesis and properties of inorganic polymers with 100% fly ash from lignite's combustion, 100% primary lead slag and mixtures of the two. The results showed that compressive and flexural strength increased up to 70% primary lead slag content. Increasing the primary lead slag content had a positive effect on the bulk density and water absorption of inorganic polymers.

Cementitious systems can be used for encapsulation of radioactive wastes. The solid nuclear wastes are classified in three types in terms of its radioactivity: low (LLW), intermediate (ILW) and high-level wastes (HLW). HLW produces important quantities of heat during storage. The ILW and LLW can be encapsulated in