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Alkali-activated concrete with Serbian fly ash and its radiological impact

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The present paper reports the results of a study on different types of fly ash from Serbian coal burning power plants and their potential use as a binder in alkali-activated concrete (AAC) depending on their radiological and mechanical properties. Five AAC mixtures with different types of coal burning fly ash and one type of blast furnace slag were designed. Measurements of the activity concentrations of ^{40}K, ^{226}Ra and ^{232}Th were done both on concrete constituents (fly ash, blast furnace slag and aggregate) and on the five solid AAC samples. Experimental results were compared by using the activity concentration assessment tool for building materials - the activity concentration index I, as introduced by the EU Basic Safety Standards (CE, 2014). All five designed alkali-activated concretes comply with EU BSS screening requirements for indoor building materials. Finally, index I values were compared with the results of the application of a more accurate index - I(ρd), which accounts for thickness and density of building materials (Nuccetelli et al., 2015a). Considering the actual density and thickness of each concrete sample index - I(ρd) values are lower than index I values.

As an appendix, a synthesis of main results concerning mechanical and chemical properties is provided.

1. Introduction

The construction industry is facing great challenges in directing its activities towards sustainable development. Environmental protection, use of waste and recycled materials and reducing the use of non-renewable resources have become the focus of current research in the field of building materials. However, the European Commission with the 2013/59/EURATOM directive (CE, 2014) considered building materials emitting gamma radiation as within the scope of the directive: so, it introduced a reference level for indoor gamma radiation emitted from building materials and requirements on the recycling of residues from industries processing naturally-occurring radioactive materials into building materials. In particular, a reference level of 1 mSv per year, as an effective dose from indoor external exposure to gamma radiation has been fixed. At this scope, for building materials being of concern from a radiation protection point of view (see the indicative list in the Annex XIII of the directive) (CE, 2014), the activity concentration of natural radionuclides (^{40}K, ^{226}Ra and ^{232}Th) has to be determined, as required in Article 75(2). Moreover, to identify “materials that may cause the reference level laid down in Article 75(1) to be exceeded”, the activity concentration index I -as conservative screening tool- has been introduced. The activity concentration index I is given by the following formula:

$$I = \frac{C_{226Ra}}{300} + \frac{C_{232Th}}{200} + \frac{C_{40K}}{3000} \quad (1)$$

where C_{226Ra}, C_{232Th} and C_{40K} are the activity concentrations in Bq/kg of natural radionuclides in building material.

It has to be noted that building materials, being of concern from a radiation protection point of view, should be also regarded as “construction products” as defined in Regulation (EU) No 305/2011 (CE, 2011). Indeed, building materials - among several basic requirements - have not to be a threat for inhabitants and

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construction workers concerning “the emission of dangerous radiation”. Therefore, it is important to account for the provisions of both two legislations when the building materials are placed on the market. Largely used bulk building materials - as concrete – can be made of many constituents, so the activity concentrations of natural radionuclides in the final building material derive from the contribution -in terms of radioactivity- of each constituent.

Furthermore, about materials incorporating residues from industries processing naturally-occurring radioactive materials (NORM), the radiological contribution of NORM residues (in terms of the activity concentration of natural radionuclides) could be assessed separately or in the final building material considering appropriate partitioning factors.

There is a big effort to develop tailor made building materials -as concrete- which takes into account environmental protection and reducing the use of non-renewable resources with health protection issues. This global aim can be reached with the development of inorganic alumino-silicate polymers synthesized starting from by-product materials rich in silicon and aluminium, which can be used instead of cement for concrete production. There are several NORM residues that have potentials to fulfill these requirements such as fly ash (FA), different types of industrial slag, red mud etc.: some of them are already utilized for such purposes (Payá et al., 2015; Shi et al., 2006).

In Serbia, about 70% of total electric energy is produced by five coal burning power plants. Consequently, 6 million tons of fly ash, or approximately 1 ton per capita, are produced per year and about 200 million tons of fly ash are already landfilled (Dragaš et al., 2014). At the moment, only 2.7% of the total fly ash production in Serbia is utilized by the construction industry (Dragaš et al., 2016). The characterization of fly ash and its products will open the possibility for a wide use both outdoors and indoors, as soil stabilization, road sub-base construction and as binder in alkali-activated concrete.

Alkali-activated fly ash concrete (AAFAC) has attracted much interest in academic and commercial spheres over the past decade. Many studies have shown a great potential for use of AAFAC in the construction industry (Shi et al., 2006; Van Deventer et al., 2012) and that physical and mechanical characteristics of AAFAC are influenced by many factors: fly ash particle size and chemical composition, fly ash loss on ignition (LOI), type and concentration of alkali activators (Na_2O concentration and SiO_2/Na_2O ratio), temperature and duration of curing (Fernandez-Jimenez et al., 2005; Criado et al., 2005, 2010; Kovalchuk et al., 2007).

Furthermore the utilization of FA in AAFAC has to be carefully studied from a radiation protection point of view because of the enhanced content of natural radionuclides compared to Portland cement. Considering the data about the average content of radionuclides - ^{226}Ra, ^{232}Th and ^{40}K - in raw and final building materials used in the European Union (i.e. cement, fly ash and concrete) (Trevisi et al., 2012; Nuccetelli et al., 2015b), a large scatter of published values is observed. Radioactivity content in FA varies a lot not only between different regions and countries but even within the same power plant during time. Obviously, it is highly dependent on the type and features of the raw material (coal) from which FA originates. This leads to the conclusion that a detailed radiological characterization of residues, concrete costituents and concrete itself is necessary to assure that these materials are not of radiological concern. Although there is a variety of methods for screening building materials from the radiological point of view (Trevisi et al., 2013), the activity concentration index I has been widely accepted and adopted as a screening tool in the EU legislation, the 2013/59/EURATOM directive (CE, 2014).

The aim of this paper is to describe the results of the radiological characterization of FA from five Serbian coal burning power plants, concrete costituents and AAFAC in order to assess their possible application as construction materials. Moreover, about the potential classification of alkali-activated fly ash concrete (as indoor-structural e.g., slabs, beams, walls, columns) or non-structural (partitions), many parameters in terms of basic physical and mechanical properties have been considered: details about chemical, physical and mechanical properties are given in Appendix.

2. Materials and methods

2.1. Materials and preparation of alkali-activated fly ash paste mixtures (AAFAP)

Fly ash was obtained from five coal burning power plants in Serbia, namely, “Kolubara”, “Nikola Tesla - B”, “Kostolac”, “Morava” and “Nikola Tesla - A”. FA samples were marked with numbers (ID) from 1 to 5 following the above mentioned order of the power plants. These plants were chosen as they have or will have a system for direct transportation of dry fly ash to the final user. Design and preparation of the samples was performed in two steps. The first step consisted of the preparation of alkali-activated pastes with different types of fly ash and alkali activator and the testing of basic material properties. In this step several alkali-activated paste mixtures (alkali-activated fly ash paste -AAFAP) containing fly ash and blast furnace slag were also prepared. Blast furnace slag was supplied from a local steel factory (specific density 2880 kg/m^3). ID of each sample was given in the form P_FA-N or P_FA-N + S, where P stands for paste, FA-N for fly ash type and S for slag.

2.2. Alkali-activated fly ash and slag concrete mixtures (AAFASC)

Based on previous results, in the second step, alkali-activated fly ash and slag concrete was designed with the addition of other necessary components such as plasticizer and aggregate. The aggregate was a mix of natural river sand (0/4 mm) and coarse aggregate (4/8 mm and 8/16 mm) with a specific density of 2600 kg/m^3 (see Fig. 1 and Table 1).

In particular, 5 AAFASC with 50% of FA and 50% of slag were designed and different mixture proportions were used. ID of each sample was given in the form C_FA-N, where C stands for concrete and FA-N for the used fly ash type.

2.3. Radiological characterization

The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K were measured in each of the concrete solid constituents - 5 types of FA, slag, aggregates, as well as in the final products – 5 types of alkali-activated concrete made with these constituents. The measurement of radionuclides content in different AAFAC and solid constituent samples was performed by gamma spectrometry. In particular two HPGe detectors with efficiency of 30% and 70% with

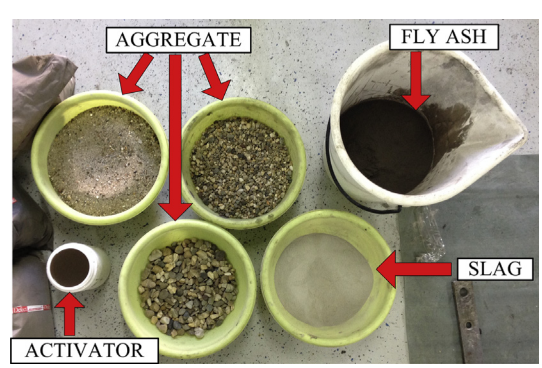

Fig. 1. Alkali-activated concrete components.