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A study on natural radioactivity and radon exhalation rate in building materials containing norm residues: preliminary results

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HIGHLIGHTS

- Natural radionuclides content in building materials added with NORM.
- Radon exhalation in concrete added with NORM.
- Intercomparison of radon exhalation rate results.
- Effect of the concrete microstructure on radon exhalation rate.

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ABSTRACT

This paper contains preliminary results of a study on the physical and radiological characterization of concrete samples containing NORM. The natural radionuclides content in term of ^{226}Ra, ^{228}Ra, ^{232}Th, ^{40}K and the radon exhalation rate of the samples were determined. Two series of samples of concrete were prepared: samples belonging to both series were made of the same basic components (Portland cement CEM I 42,5R as binder, aggregate and plasticizer) but different amount of NORM residues and mineral additives were used.

The concrete samples were characterize in term of absolute density, permeability, total and open porosity. The radiological content was evaluated by using gamma spectrometry and the radon exhalation rate measurements were performed using the dynamic method. Moreover, the Activity Concentration Index (I), introduced by the 2013/59/Euratom Directive, has been used in order to evaluate if concrete samples could exceed the reference level for effective dose due to gamma radiation in building materials.

1. Introduction

The European Commission in 2011, launched its "Roadmap for Resource-efficient Europe" [1] in which three action lines are stated. In particular, in the first one, named "Transforming the economy" turning waste into a resource is clearly indicated. This is intended that the European Commission will stimulate secondary materials markets in order to move towards an economy based on re-use and recycling. In this framework, the use of raw materials for building material production is discouraged while the use of secondary or residue-derived materials is promoted.

In this particular frame, the attention of regulators and researchers was focused on large amount of residues produced by NORM (Naturally Occurring Radioactive Material) industries, which can be recycled as secondary raw materials in building products. Indeed, although recycling NORM residue into building materials brings significant beneficial effect on environment and sustainability, the consequent enhanced radionuclide content may have an impact on human health due to a potential increase of gamma exposure and of indoor radon activity concentration.

As consequence, the Council Directive 59/2013/Euratom (EU BSS in the following) [2] considers the protection of the population from gamma radiation emitted from building materials in its scope. Moreover, the EU BSS takes into account the possible contribution of building materials as indoor radon source among the

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items listed in the annex XVIII of EU BSS [2] to be considered in the preparation of the national action plans.

Several studies have addressed the radiological consequences of recycling industrial by products in building materials considering their impact both in terms of gamma [3–5] and radon exposures [6–9]. Nevertheless, data about radon exhalation rate in building materials containing NORM residues are not so many: in particular, in the ISS-INAIL database [10] on natural radioactivity in building materials, which collects radiological data about more than 24,000 samples, only 1100 data of 14 European Countries on radon emanation/exhalation rate are present.

The analysis of these data [10] put in evidence problems due to the use of different units and lack of information especially about the radium content.

Starting from this experience, INAIL (*National Institute for Insurance against Accidents at Work*, Italy), GIG (*Główny Instytut Górnictwa*, Poland) and ISS (*National Institute of Health*, Italy) planned a research to collect more data about radon exhalation rate from building materials (in particular on concrete samples) containing NORM residues and also to investigate the feasibility of NORM residues immobilization in concrete. The present paper contains preliminary results of this project, in particular:

- natural radionuclides content in concrete samples obtained by gamma spectrometry;
- radon exhalation measurement results performed using the dynamic method;
- intercomparison of radon exhalation rate results achieved by the three Institutes;
- evaluation of the effect of concrete microstructure and of the different percentages of industrial residues added to the concrete on radon exhalation rate.

Measurement of radon exhalation rates on building materials can be performed by different methods based on three basic approaches: the radon accumulation inside a closed chamber (CCM) [11,12], flow through systems [11,13] and adsorption [11].

The radon accumulation inside a closed chamber is the most common method (CCM) [14]; it is based on the principle of radon accumulation in an closed container mounted on the surface of the sample (soil or building material) or in which a sample is put [15–17]. The measurements of radon accumulated inside the chamber can be performed either with passive devices (SSNTD, electrets, activated charcoal etc. [11,14,15,18,19]) or with active monitors [15,16,18] as detection technique.

In the framework of the ERRICCA project (European Research into Radon In Construction Concerted Action), the performances of different radon exhalation rate methods were evaluated through an international intercomparison exercise [15] at which 20 participants from 13 countries took part. The intercomparison exercise results put in evidence that enclosed sample method together with an active radon monitoring is the most used.

For the objectives of the present study, radon exhalation rate measurements in concrete samples were carried out by using closed chamber method and active monitoring techniques. More details about the measurements are reported in paragraph 2.2.4.

2. Materials and methods

2.1. Concrete samples preparation

Two series of samples (named GIG1 and GIG2) were prepared by GIG and casted in different moulds (16 cm cubic mould according to the ISO/FDIS 11665-9 standard requirements [19] and cylindrical mould of 16 cm diameter). The cylindrical shape was chosen considering the possibility to perform tensile strength test and to evaluate the shape effect on the exhalation rate. Moreover, also smaller cylindrical samples (diameters 4.5 cm and height 7 cm) were made. All samples were prepared according to the European Standard EN 206-1 Concrete – Part 1: Specification, performance, production and conformity [20]. The samples were compacted by vibration, demolded after 48 h from preparation and later on, stored for 26 days in a water bath at 20 ± 1 °C for curing. Finally, they were dried to the constant mass in the laboratory dryer at 105 °C.

Tables 1 and 2 summarize details about the composition of GIG1 series and GIG2 series samples, respectively. The NORM residue added in GIG1 series samples is spent activated charcoal from underground water purification installation, used as admixture in different mass percentage (0.2%, 1.3% and 2.4%). In GIG2 samples series, NORM residues used as binder are bottom ashes from coal combustion, while bottom sediments from coal mine settling ponds are used as coarse aggregates.

2.2. Samples characterization

Samples were characterized in terms of absolute density, total porosity and open porosity, permeability, radionuclides content and radon exhalation rate. Indeed, many factors can influence the radon exhalation rate such as: ^{226}Ra activity concentration, its distribution within grains, material density, solid grain properties, volume of pore space, water content of the building materials, and ambient temperature [11,21–23]. This physical characterization has had the dual purposes: firstly, from an engineering point of view to provide materials that can be effectively used as building materials. Secondly, to investigate a possible correlation between the microstructure, physical and radiological properties.

2.2.1. Measurement methodologies of absolute density, total porosity and open porosity

These set of measurements were conducted in Laboratory of Mining and Environmental Hydrogeology at GIG. Measurements of absolute density were performed using a high-pressure helium pycnometer AccuPyc II 1340 (Micromeritics Instrument Corp.).

Table 1
Composition of concretes from GIG1 series.

	Binder	Aggregates	Admixture	Plasticizer	H_2O
GIG1/01	22.1% Cement CEM I 2,5R	66.4% Sand 66.7% + aglophoryt 33.3%	0.2% Spent activated charcoal 1% of the binder mass	0.2% Beto-plast	11.1% H_2O/binder ratio=0.5
GIG1/06	21.9% Cement CEM I 42,5R	65.4% Sand 66.7% + aglophoryt 33.3%	1.3% Spent activated charcoal 6% of the binder mass	0.2% Beto-plast	10.9% H_2O/binder ratio=0.5
GIG1/11	21.6% Cement CEM I 42,5R	64.9% Sand 66.7% + aglophoryt 33.3%	2.4% Spent activated charcoal 11% of the binder mass	0.2% Beto-plast	10.8% H_2O/binder ratio=0.5