WASTES

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Toxicity measurement techniques for building materials with wastes

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ABSTRACT: The innovative Horizon 2020 program sponsored by the European Union (EU) aims to promote and develop processes of waste integration in construction materials. However, several potential health hazards caused by building materials have been identified and, therefore, there is an ongoing need to develop new recycling methods for hazardous wastes and efficient barriers in order to prevent toxic releases from the new construction solutions with wastes. This paper presents an overview that focus on two main aspects: the identification of the health risks related to radioactivity and heavy metals present in building materials and identification of these toxic substances in new construction solutions that contain recycled wastes. Different waste materials were selected and distinct methodologies of toxicity evaluation are presented to analyse the potential hazardous, the feasibility of using those wastes and the achievement of optimal construction solutions involving wastes.

1 INTRODUCTION

The importance of evaluating the potential hazards of toxicity of building materials with wastes is the result of the rising awareness of several human living conditions, environmental consequences and massive damaging of industrial track (Hopwood et al., 2005; McDougall et al., 2001; Sullivan et al., 2014). In fact, construction materials generate about 31% of all waste in Europe. Moreover, recent investigation (Torgal et al., 2012; Fucic et al., 2010; Fucic, 2012, Kovler, 2009; Orisakwe, 2012; Prazakova et al., 2013) showed that several health problems may be caused by the chronic exposure to hazardous substances present in the building materials, such as materials that contain asbestos, heavy metals, radioactive substances or release volatile organic compounds and nanoparticles. For instances, asbestos exposure has been pointed out as a cause of health problems such as mesothelioma, lung cancer and asbestosis. Therefore, the Directive 2003/18/EC prohibited the production of asbestos-based products (Torgal et al., 2012). For all these reasons, the European Union (EU) seeks to create frameworks of policies and methodologies that will promote a substantial reduction of the waste generation in the future, as well as increase the control of the exposure to hazardous wastes.

The integration of recycled waste materials in new construction materials, as well as in engineering structures, is foreseen as a promising sustainable approach to close the material loop towards circular materials' economy, reducing hazardous emissions to air, land, water and human living conditions. Several waste materials have been integrated in construction solutions, such as: fly ash, phosphorous slag, tin slag, copper slag, residues from aluminium production, residues from steel production, textile, rubber tires, plastic, sewage sludge and foundry sands (Torgal et al., 2012). Nevertheless, the success of this approach implies the characterization of the material properties (e.g. chemical, structural and energy content) and also requires the evaluation of its toxicity level (Tiruta-Barna et al., 2012; Perrodin et al., 2000). To evaluate the toxic effects of pollutants for human and environment, standard methodologies based on classical human Health Risk Assessment (HRA), Ecological Risk Assessment (ERA) and laboratorial tools exist as measures to quantify

Table 1. List of building materials and health effects associated to the presence of radioactive substances.

Isotope:	^{226}Ra, ^{232}Th, ^{222}Rn, ^{40}K
Building materials:	Granite, gneiss, porphyries, syenite, basalt, tuff, pozzolana, lava, phosphor-gypsum, alum-shale, coal fly ash, oil shale ash, tin slag, phosphate fertilizer, oil industry, red mud from aluminium production residues from steel production, from steel production residues from steel production, concrete, clay, bricks, sand-lime, tiles, cement.
Health effects:	Cancer, cardiovascular diseases, immunological disturbances, chromosomal abnormalities and gene mutations.
References:	(Torgal et al., 2012; Nisnevich et al., 2008; Yao et al., 2015, Kovler, 2012a; Kovler, 2009; Dikstra et al., 2004; Marocchi et al., 2011; El-Thaer, 2012; Uosif, 2014; Kinsara et al., 2014)

the potential hazardous pollution in specific disposal, elimination or reuse scenarios (Tiruta-Barna et al., 2012).

There are two main tasks that are carried out in parallel in this paper. The first one is to describe two sources of toxicity (radioactivity and heavy metals), which is the core of section 2. The second task is to describe some techniques used to determine toxic substances in construction solutions with wastes materials. Finally the conclusion points out the state of the current practice in the field of toxicity evaluation of construction materials and directions for future research are considered.

2 TOXIC SOURCES AND MAIN HEALTH HAZARDS OF BUILDING MATERIALS

Radioactivity is a process by which certain naturally occurring or artificial nuclides undergo spontaneous decay or loss of energy releasing a new energy, therefore resulting in an atom "the parent nuclide" transforming to an atom "the daughter nuclide".

Radiation sources can be natural or artificial. In case of natural, it is estimated that 85% can be cosmic (14%) and telluric radiation (71%) (Kovler, 2012b). The telluric radiation is caused by Naturally Occurring Radioactive Materials (NORM),^{235}U and ^{238}U – uranium; ^{40}K – potassium; ^{226}Ra – Radium and ^{232}Th – thorium, which represents an external gamma radiation, both inside and outside buildings (Kovler, 2011; Kovler, 2012a; Kovler, 2012b). The radionuclides ^{226}Ra, ^{232}Th and ^{40}K, which are present in the Earth's crust, are also in building materials or in additives of building materials (Table 1). Radon, ^{222}Rn, is a radioactive, colourless, odourless, tasteless noble gas, occurring naturally as the decay product of radium ^{226}Ra (external radiation). The internal radiation in the human body is caused by natural radionuclides in food, water and air. Moreover, the internal radiation exposure is due to radon gas ^{222}Rn, which belongs to the ^{238}U decay chain, and marginally to its isotope thoron ^{220}Rn, which belongs to the ^{235}U decay chain, and their short lived decay products, exhaled from building materials into the room air (Kovler, 2011). Being an inert gas, radon can move rather freely through the materials. For instance, Kovler (2012b) mentioned that radon emanates naturally from the ground and from mineral building materials, wherever traces of uranium or thorium can be found (e.g. granite or shale). Radon is also present in high concentration in granitic residual soils, rocks and fly ash (Kovler, 2012b; ECRP-112, 1999; ECRP-96, 1997). Table 1 lists examples of building materials where the radioisotopes are present and the corresponding health problems associated to radioactivity exposure.

The new Council Directive 2013/59/EURATOM, which sets out basic safety standards for protection against the hazards arising from exposure to ionising radiation, recognized the importance of protection against natural radiation sources, in particular from industries processing materials containing naturally-occurring radionuclides. Establishing reference levels for indoor radon concentrations and for indoor gamma radiation emitted from building materials and introducing requirements on the recycling of residues from industries processing naturally-occurring radioactive

Table 2. Heavy metals in building materials and health effects associated to chronic exposure.

Heavy metal	Health effects	Building materials	References
Cadmium	Lung cancer, osteomalacia, proteinuria.	Pigments, chemical stabilizers, nickel-cadmium batteries.	(Orisakwe, 2012)
Mercury	Stomatitis, nausea, nephrotic syndrome, neurasthenia tremor.	Fluorescent lamps.	(Orisakwe, 2012)
Lead	Nephropathy, anaemia, encephalopathy.	Lead connection pipe.	(Hayes, 2012)
Chromium	Pulmonary fibrosis, lung cancer.	Dyes and paints, stainless steel, metallurgy, chrome platin.	(Orisakwe, 2012)
Arsenic	Diabetes, hypopigmentation, cancer, liver disease.	Chromated copper arsenic in pressure treated wood.	(Hall, 2002)

Table 3. Estimation of heavy metals present in wastes of case studies.

Wastes	Heavy metals	Case studies	References
Coal fly ash	Cr, Cu, Ni, Pb, V, Zn, As, Hg, Cd, Sn.	Incorporation of coal fly ash in Portland cement concrete.	(Torgal et al., 2012) (Yao et al., 2015) (Azevedo et al., 2012) (EPA, 2015)
Rubber tires	Zn, Se, Cd, Pb.	Incorporation of crumb rubber in asphalt concrete.	(Peralta et al., 2012) (EHHI, 2015)

materials into building materials. Other main hazards of interior building materials are the heavy metals, which are present in pigments, electric lamps, leads pipelines and various other household wares. Heavy metals are also present in various wastes used in concrete. Table 2 lists examples of building materials that contain heavy metals (cadmium (Cd),mercury (Mg), lead (Pb), chromium (Cr), arsenic and the corresponding health effects associated to heavy metals chronic exposure (Orisakwe, 2012; Hayes, 2012).

3 INTEGRATION OF WASTE MATERIALS IN CONSTRUCTION SOLUTIONS

3.1 *Case studies of waste incorporation in building materials*

Integrating waste materials in building materials requires a good knowledge of the properties and characteristics of those waste materials. Basic characterization constitutes a full characterization of the waste materials by gathering all the necessary information for a safe management of the waste in the short and long term, such as: type and origin, composition, consistency, leachability, among other (EN 14735). The Technical Committee CEN/TC 351 'Construction products: Assessment of release of dangerous substances' and CEN/TR 16098:2010 (Concept of horizontal testing procedures in support of requirements under the CPD) work underpinned of standards in the field of the assessment of building materials for their reuse.

The final rule of Environmental Protection Agency (EPA), EPA "Hazardous and Solid Waste Management System; Disposal of Coal Combustion Residuals from Electric Utilities." address serious human health and environmental risks from unsafe coal ash disposal (EPA, 2015). Table 3 shows two case studies of incorporation of wastes (coal fly ash and rubber tires) into concrete (both for Portland cement and/or asphalt concrete) and shows the heavy metals associated to each waste. In accordance with research (Torgal et al., 2012; Yao et al., 2015; EPA, 2015), the coal fly ashes contain: chromium (Cr), copper (Cu), nickel (Ni), lead (Pb), zinc (Zi), arsenic (As), mercury (Hg), cadmium (Cd), tin (Sn) and selenium (Se). A study on the properties of recycled tire rubber (Cao, 2007) shows that its chemical composition is acetone extract (15.5%), ash content (6.0%), carbon black (29.5%) and rubber hydrocarbon (49.0%).

In the Waste Framework Directive 2008/98/EC, (European Commission, 2008), waste should be classified in accordance with the European List of Waste (Commission Decision 2000/532/EC), where 405 waste types are defined as hazardous waste material. Other 200 waste types are also listed as "mirror entries", what means "Waste with potential to be either hazardous or non-hazardous depending on their composition and the concentration of dangerous substances" (Römbke, 2009). Therefore, fly ash can be classified as19 01 13 and considered with hazardous properties if containing dangerous substances (H13 and H14 "Ecotoxic"). On the other hand, it can be classified as 19 01 14 fly ash is non-hazardous. The classification to end-of-life tyres is 16 01 03 ("end-of-life tyres"), non-hazardous.

3.2 *Toxicity measurement techniques of buildings materials: radioactivity and heavy metals*

The release of soluble constituents upon contact with water is regarded as a main mechanism of release which results in a potential risk to the environment during the disposal of waste materials (Stiernström et al., 2014). Therefore, the heavy metals can be determined through an assessment of leaching potential of materials with laboratory prepared samples and on site samples. These tests aim to identify the leaching properties of waste materials. Therefore, surfacing materials are measured under controlled laboratory conditions in accordance with technical specification CEN/TS 15862: 2012 – "Characterization of waste. Compliance leaching test. One stage batch leaching test for monoliths at fixed liquid to surface area ratio (L/A) for test portions with fixed minimum dimension". The main factors to control leachability are: pH, redox potential and physical parameters.

Laboratorial determination of radioactive properties can be through gamma spectrometry and exhalation with representative site samples. The first determination of ^{232}Th, ^{226}Ra and ^{40}K on material is made in order to estimate the potential hazard of it from a radiotoxicity perspective. The radionuclide measurements can be carried out by high resolution gamma-ray spectrometry. The HPGe detectors can be lead shielded with cooper and tin lining. The Genie 2000 software can be used for data acquisition and analysis. The detection efficiency is determined using NIST-traceable radioactive standards, to reproduce the exact geometry of samples, in a water-equivalent epoxy resin matrix. The measurement of radon aims to determine the material potential to generate high indoor radon concentrations and establish a classification based on its potential risk. The composition of samples can be analysed when placed inside cylindrical sealed containers. The radon exhalation rate is measured by using an active continuous radon monitor, in order to follow the ^{222}Rn activity growth as a function of time and determining the average equilibrium of the gas concentration during the exposure period (Madruga et al., 2011).

Some simple actions such as sealing around loft-hatches, sealing large openings in floors and extra ventilation do not reduce radon levels on their own. To lower radon levels, homeowners can use passive or active mitigation methods. The first are associated to radon levels above the 4 pCi/L limit (Henschel, 1993). Such methods include sealing foundation openings and around areas where pipes enter the home, reducing 15% of radon level (UkRadon, 2014). Other methods include applying coatings or membranes on walls to reduce permeability and installing a passive soil ventilation system and also active systems with mechanical and electrical devices to reverse air flow and ventilate soil beneath the foundation (Koff et al., 2007; UkRadon, 2014).

4 CONCLUSIONS

Two main sources of toxicity have been identified in building materials as causes of health problems: radioactivity and heavy metals. Two examples of incorporation of waste solutions were shown to estimate the potential hazards. Fly ash and tire rubbers contain different chemicals which need to be tested accurately in laboratory to determine how much substances contaminate the indoor air and the environment.

The lack of safety procedures related to the process of integration of waste materials in new construction solutions constitutes a current problem in terms of toxic compounds. Moreover, when evaluating the influence of the radioactivity and heavy metals release from construction materials with wastes it is fundamental to understand which are the main factors to consider. Such studies should include both laboratory tests of those material specimens of the same geometry and also boundary conditions under well-controlled ventilation (radioactivity). Moreover, they should include assessment of the toxicity potential of the solutions with wastes in their utilization scenarios, especially via emissions in water and dispersion in the surrounding natural environment (e.g., heavy metals).

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