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Review

Concrete and cement composites used for radioactive waste deposition

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A B S T R A C T

This review article presents the current state-of-knowledge of the use of cementitious materials for radioactive waste disposal. An overview of radwaste management processes with respect to the classification of the waste type is given. The application of cementitious materials for waste disposal is divided into two main lines: i) as a matrix for direct immobilization of treated waste form; and ii) as an engineered barrier of secondary protection in the form of concrete or grout. In the first part the immobilization mechanisms of the waste by cement hydration products is briefly described and an up-to date knowledge about the performance of different cementitious materials is given, including both traditional cements and alternative binder systems. The advantages, disadvantages as well as gaps in the base of information in relation to individual materials are stated. The following part of the article is aimed at description of multi-barrier systems for intermediate level waste repositories. It provides examples of proposed concepts by countries with advanced waste management programmes. In the paper summary, the good knowledge of the material durability due to its vast experience from civil engineering is highlighted however with the urge for specific approach during design and construction of a repository in terms of stringent safety requirements.

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1. Introduction

Sources of radiation are of beneficial use in many fields ranging from nuclear power, to medicine, scientific research, industry and

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agriculture. In all these applications radioactive waste with more or less radioactivity, in several forms and concentrations is produced. To ensure safe disposal of such hazardous waste a number of variables need to be considered, which lead to finding the most proper way for disposal of the particular radwaste type. There are a variety of the waste conditioning procedures and the final repositories systems. Concrete and cement composites, as one of these alternatives, are cheap and well goal-fulfilling materials used for radwaste immobilization or serving as the secondary protection barrier between waste and the surrounding environment in the near-surface repository. The experience of many years from use of cement and concrete in civil engineering provides broad knowledge about its technology, durability and performance in wide range of environments. The article is oriented to review the utilization of cement composites and concrete for radwaste solidification and final disposal with respect to the specifics of particular radwaste type management.

2. Radioactive waste classification

There are several schemes for radioactive waste classification, however the method of conditioning and the end point (i.e. transient storage or underground repository) rely on the level of radioactivity and lifetime of the radwaste. Therefore, the International Atomic Energy Agency (IAEA) has established a generally recognised classification scheme based on two parameters: radionuclide half-life and radioactivity content (IAEA, 2009). There are two categories considering the lifetime of radionuclides: long-lived and short-lived radioactive waste, while the threshold level is established to be the half-life of ^{137}Cs (30.17 years). The general term “radioactivity content” includes more parameters: activity concentration, specific activity and total activity. The higher the radioactivity content, the higher the requirements on multi-barrier protection system. A special category is the exempt waste (EW), which contains such small concentrations of radionuclides, that it does not need any protection barrier and can be disposed in conventional landfills. Exemption levels were established for both concentration and total amount of radionuclides based on the individual and collective dose and can be found in the International Basic Safety Standards for Protection against Ionizing Radiation and for the Safety of Radiation Sources (IAEA et al., 1996). Besides exempt waste there are five categories of radwaste according to IAEA:

a) *very short-lived waste (VSLW)* – usually radionuclides with very short half-lives from medicine and research
b) *very low level waste (VLLW)* – typically soil and rubble with low levels of activity concentration
c) *low level waste (LLW)* – radionuclides of higher activity than VLLW, but with limited concentrations of long-lived ones, requires engineered barriers for isolation in near-surface repositories for periods up to a few hundreds of years
d) *intermediate level waste (ILW)* – higher amounts of long-lived radionuclides, requires more advanced isolation from the environment in higher depths (tens to a few hundreds of meters) but with no or limited provisions to heat dissipation
e) *high level waste (HLW)* – waste with very high concentrations and/or high amount of long-lived radionuclides, typically spent nuclear fuel (SNF) and facilities and activated components from decommissioning of nuclear power plants, needs disposal in deep stable geological formations in depths of hundreds of meters.

The IAEA classification scheme does not give exact limits of concentrations for particular radionuclides to classify them into the appropriate classes; these are set in the domain of national regulatory bodies.

The process of radwaste management relies on its characterization, which includes the category and its form (solid or liquid). After characterization, the radwaste is processed, stored and ultimately disposed. Processing is composed of pre-treatment, treatment technologies (such as volume reduction, chemical modification, evaporation etc.) and conditioning. Conditioning transforms the waste into a form suitable for handling and storage including immobilization. Immobilization ensures safe embedding of radwaste into a matrix, preventing the leakage of radionuclides into the environment. Such solidified waste is further packaged in a system of barriers according to its class. A general scheme of radioactive waste management is presented in Fig. 1 and an example of radwaste treatment in Slovakia is given in Fig. 2. Cement matrix is well used for immobilization of VLLW, LLW and ILW waste thanks to the combination of its low cost and the suitability for radionuclide encapsulation due to physical and chemical bonds. Concrete serves as secondary barrier for LLW and ILW in the form of packages, which are further disposed in the appropriate repository.

3. Immobilization of LLW/ILW by cement composites

3.1. Cement suitability for radwaste immobilization

The understanding of the immobilization mechanisms of radioactive waste in cement matrices is complicated due to the variety of alternatives of cementing agents, radionuclide nature and service environments. However, the retention potential of cementitious materials is very high thanks to its high sorption or uptake capacity (Chen et al., 2009; Mallants et al., 2016; McCulloch, 1985). The choice of the right matrix for immobilization derives from the chemistry of both radioactive and inactive compounds present in the waste. For example, certain ion exchange resins are not compatible with the Ordinary Portland cement (OPC) environment, but also the presence of non-hazardous waste, such as cellulose may be problematic, as it degrades with time and thus generates gases, which may complex and solubilise actinides (McCulloch, 1985). These factors have to be considered in making prediction of changes in physical parameters, especially pH and Eh, due to the cement-waste interactions.

3.2. Immobilization mechanisms of cement

Cement in the immobilization process acts both chemically and physically. Physical action is mainly realized by surface sorption. The chemical mechanisms have not been up to now completely understood, but some analogues can be derived from experience of hazardous metal waste immobilization. The most published data relating the retention mechanisms is referred to Portland cement and can be summarized as follows (Ojovan, 2011):

a) *surface and bulk sorption* – prevailing mechanism for low species concentrations with low capacity, dependant on the surface area of hydration minerals, mainly of C-S-H gel
b) *ion exchange* – crystalline phases of hydrated cement offer anion and cation sites for uptake of radioactive species, e.g. Cl in AFm
c) *characteristic phase formation* – e.g. Ca-U-OH or $CaSn(OH)_6$ for U and Sn encapsulation respectively
d) *Oxy/hydroxyl precipitation* – creation of usually metastable precipitates, e.g. $Cr(OH)_3$
e) Combination of above mentioned mechanisms.

The particular processes of retention are controlled by hydrated