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Assessment of strontium immobilization in cement–bentonite matrices

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HIGHLIGHTS

- Strontium immobilizations in cement–bentonite waste matrices were investigated.
- The presence $SrCl_2$ and bentonite did not lead to formation of new hydration phases.
- The mechanical performance of the studied matrices was acceptable.
- Sr^{2+} binding is due to substitution on Ettringite and Montmorillonite lattice.
- Strontium leaching resulted from first order reaction and diffusion mechanisms.

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ABSTRACT

The feasibility of immobilizing strontium in cement–bentonite matrices was investigated by studying the effect of mineralogical phase development on mechanical and containment performances. Within this context, the chemical composition and physical properties of bentonite were determined. Different cement–bentonite waste matrices were prepared and analyzed using XRD technique to trace the changes in phases during the curing period. The mechanical performance of these matrices was evaluated by measuring the compressive strength throughout their curing period and the containment performance was determined by conducting long-term static leaching test then the experimental results were checked against some regulatory limits. The results indicated that the presence of strontium and bentonite did not lead to formation of new hydrated phases. The mechanical performance of the matrices is acceptable and the enhanced compressive strength was attributed to the progression in the formation of cement hydrated phases and the pozzolanic reaction between bentonite and lime in cement–bentonite matrices. The speciation data and phase structures analysis indicated that Sr^{2+} containment in cement might be due to Ca substitution in Ettringite structure and cations exchange on Montmorillonite lattice. The mathematical analysis of the long-term leaching results indicated that strontium leaching resulted from a combination of first order reaction and diffusion mechanisms.

1. Introduction

Cement based materials have been used in radioactive waste management to produce stable waste forms and build engineering barriers in disposal facilities. The immobilization of low and intermediate level radioactive wastes in cementitious matrices is the most commonly used technique to produce inexpensive waste matrix that complies with regulatory requirements [1]. Conventional cementitious waste matrices consist of waste, cement, additives, and water [1,2]. The additives are used to enhance the mechanical performance of the waste matrix and/or reduce the leachability of problematic radionuclides e.g. ^{137}Cs, ^{60}Co, and ^{90}Sr [3–6]. Blast furnace slag, fly ash, kaolin, zeolites, and bentonite have been tested for their potential use as additives [3,5,7–19].

Bentonite was proposed as buffer and/or shaft seal in geological disposal for nuclear wastes due to its low permeability, chemical and physical stability, and compatibility with different host environment [20]. The utilization of bentonite as liner and waste matrix additive was supported by the availability of smectite ion exchange and amphoteric surface complexation sites for the adsorption of radionuclides [21]. Limited studies in the literature investigated the immobilization of radiocontaminants in cement–bentonite matrices [3,15,17,22]. These studies assessed the leaching and mechanical performances of the produced matrices and estimated the diffusion coefficients.

This work is directed to assess strontium immobilization in cement–bentonite matrices by investigating the hydrated mineralogical phases and their effect on the immobilization process.

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Strontium was selected for this study due to its biochemical characteristics and solubility in the alkaline pH environment of cement matrix [23,24]. To trace the mineralogical phases development, different cement–bentonite matrices were prepared and analyzed using XRD technique. The mechanical and containment performances of the matrices were evaluated by checking the experimental results against some regulatory limits. Strontium speciation in the presence of hydrated phases and phase structure data were studied to investigate strontium containment on hydrated matrices. Finally, Sr^{2+} leaching mechanisms from cement–bentonite matrices were investigated.

2. Experimental

2.1. Materials and preparation of immobilization matrices

The studied matrices were prepared using commercial Ordinary Portland Cement (OPC) and natural bentonite, their chemical compositions are listed in Table 1 [25]. Important bentonite physical characteristics such as Specific Surface Area (SSA), Particle Size Distribution (PSD), and losses during calcinations were determined. SSA was measured using BET method, at which nitrogen adsorption isotherm was quantified by a fully automated surface area analyzer (Nova instrument, Quantachrome Corporation, USA). PSD was analyzed using sieves ranged from 30 to 250 μm.

The immobilization matrices were prepared by mixing cement with 5, 10, and 15 wt% of bentonite. The reference matrix was prepared using plain OPC. Preliminary investigations were carried out to determine the optimum water to cement ratio that was found 0.45. The matrices were prepared by placing the required amount of solids in a smooth non-absorbent surface, and a crater was formed in the center. The required amount of $SrCl_2{\cdot}6H_2O$ solution (0.1 g/l) was poured into the crater by the aid of a trowel. The mixing operation was then completed by continuous vigorous mixing. At the end of mixing, the paste was directly poured into cubic moulds (5 cm side length). The paste was placed in the moulds in two approximately equal layers; each layer was compacted and passed along the surface of the moulds until homogenous specimen was obtained. After the top layer was compacted, the moulds was then vibrated to remove any air bubbles and to have a better compaction of the paste. Immediately after moulding, moulds were kept in air for 24 h. The moulds were demoulded and were cured in a humid atmosphere.

2.2. Mechanical and containment tests

To understand the nature of the immobilization process and assess the mechanical and containment performances of the studied matrices, mineralogical phase identification, compressive stress testing, and static long-term leaching tests were carried out.

Table 1
Chemical composition of OPC and bentonite [25].

Chemical composition	OPC	Bentonite
CaO	63	2–6
SiO_2	20	49–55
Al_2O_3	6.0	20–24
Fe_2O_3	2.1	2.5–6
MgO	1.5	0.5–2
Na_2O	0.5	1.1–2.4
K_2O	–	1.2–1.4
Others	6.9	–
LOI at 850 for 2 h	–	9–10

2.2.1. Mineralogical phase identification

TGA, BSE–SEM, and XRD are available techniques for identifying cement hydration phases. Recent developments in commercial XRD software have improved the performance of this technique [26]. In this study, XRD analysis was selected to identify the mineralogical phase changes. The cured cementitious matrices were grounded after different time spans (4, 7, 21, and 28 days) and the powders were submitted to X-ray diffractometer (PANalytical – Empyrean diffractometer).

2.2.2. Compressive strength testing

The development of the mechanical performance was studied by evaluating the compressive strength at different time spans (4, 7, 21, 28 days) using microprocessor based compression testing machine.

2.2.3. Static long-term leaching test

IAEA long-term static leach test was performed using 28 day cured matrices; the matrices were immersed in distilled water, so that the ratio of leachant to waste matrices volumes equals 10 [27]. Leachant samples were withdrawn and analyzed using atomic absorption spectrophotometer (Buck scientific, VGP210) daily for a week, weekly for a month, and monthly for three months. The leachant pH and temperature were in the range 11.5–12.2 and 28.5–30 °C.

3. Results and discussion

Water demand, setting, hydration reaction and compressive strength of the produced waste matrix are affected by the physical characteristics of cement, waste and additives. The results of bentonite physical characterization revealed that, the PSD is in the range 40–100 μm. It is worthy to note that cement PSD falls below 100 μm that will yield in good mixing [28]. The bentonite grading curve, illustrated in Fig. 1, is showing steep slope over a narrow range of particle diameters. This might indicate that bentonite is uniformly graded. The coefficient of uniformity ($C_u = D_{60}/D_{10}$) was found to equal 1.6, which confirms the uniformity of the particle size distribution of bentonite [29–31]. Published results indicated that PSD of fine aggregate has great influence on the properties of produced cementitious material, where narrow PSD will accelerate the hydration [32–34]. Bentonite SSA was found to be 92.2 m^2/g and the loss during calcinations at 850 °C for 2 h is 9–10 wt%. The relatively high SSA value indicates that there are more available sites for contaminant retention [35]. Based on the above findings,

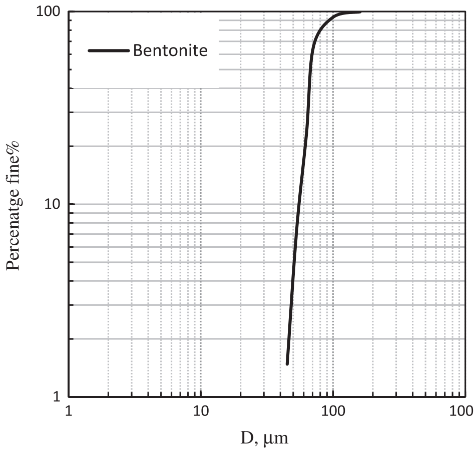

Fig. 1. Bentonite particle size distribution.