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# Characterization of biological waste stabilized by cement during immersion in aqueous media to develop disposal strategies for phytomediated radioactive waste

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A B S T R A C T

Dried and grated biological waste was immobilized with cement to generate specimens mimicking solidified phytoremediated radioactive waste. Stability of these solidified biological waste specimens was estimated during drastic long-term flooding, which is considered one of the typical environmental impacts affecting physical and mechanical characteristics of such specimens. For these investigations, biological waste generated during phytoremediation using the aquatic plant *Veronica anagallis-aquatica* was solidified and subsequently examined during flooding. After flooding with water of different compositions, mechanical and physical stability of the specimens was evaluated. Cementation of 3% dried biological waste resulted in a satisfying compressive strength of the resulting solidified material of more than 13 MPa. The highest value of hardness exceeded 25 MPa, and was obtained in samples cured in sea water or ground water due to mineral salts sealing the pores, whereas an insignificant decrease in durability was observed in those specimens cured in tap water. Maximum mass change caused by the water absorption during six months of curing was below 4.5% of the initial mass; this change was more pronounced for solidified waste immersed in sea water or ground water than for samples immersed in tap water. Retardation of material and status of the cement phases after flooding were investigated via FT-IR, XRD and SEM; the results point to the suitability of this cement type as powerful material for immobilization of biological radioactive waste, as manifested by acceptable durability and reasonable porosity during long immersion in different water compositions.

## 1. Introduction

Recovery of radionuclides and other heavy metals from soil or water bodies via bioremediation constitutes an environmentally friendly process, which is generally performed at ambient conditions (Dixit et al., 2015; Prakash et al., 2013). Moreover, the use of biomaterials to accumulate hazardous materials from contaminated environments has been proposed by many researchers as a relatively cost-efficient method for decontamination (Hossain et al., 2016; Abu-Khadra et al., 2016). In this context, aquatic plants are currently considered being among the best performing organism for remediation of polluted soil or water. Bioremediation by such aquatic plants starts with the uptake of “free” pollutants, which are highly diluted in soil or water, by the plant's rhizosphere. Subsequently, the pollutants are transported to target parts of the plant, were they remain in a fixed, concentrated form (Saleh, 2012; Saleh et al., 2017). Now, regarding long-term disposal of pollutant-rich plant material, efficient immobilization is required to avoid leaching of the pollutants from the plant material, i.e., to prevent re-entry of the pollutants into the ecosphere. This is typically accomplished by drying, grinding, and finally immobilizing the risky plant material, e.g., by cementation, to ensure easy handling and isolation during safe transport, disposal, and long term storage (Bayoumi et al., 2012; Saleh, 2014a; EL-Dakroury, 2014; Saleh and Eskander, 2012a). The present study provides the proof of concept for successful stabilization of dried ground biological waste of *Veronica anagallis-aquatica* by entrapment in cement paste; this waste constitutes as a model for phytoremediated radioactive waste, which will be investigated in future follow-up studies. Already decades ago, the principal mechanisms involved in stabilization of nuclear waste by cement minerals, cement, or mortar were described (Komarneni and Roy, 1981; Saleh and Eskander, 2012b; Eskander and Saleh, 2012; Eskander et al., 2012). More specifically, Portland cement was already reported 25 years ago as a viable material to immobilize inorganic pollutants like fly ash. It is believed that the main effect of such cement matrices for immobilization originates from their high internal pH-value, which allows precipitation of various nuclides as hydroxides (Atkins and Glasser, 1992). In our study,

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evaluation of mechanical and physical parameters for the hardened biological waste specimens was performed through curing in aqueous media by measuring the compressive strength and porosity. Moreover, Fourier-transform infrared spectroscopy (FT-IR), scanning electron microscopy (SEM), energy dispersive X-ray spectroscopy (EDX) and X-ray diffraction analyses (XRD) were performed. These analytical tools demonstrated the efficiency of cement as a solidifying agent for the phytoremediated waste in a durable form.

The current investigation highlights (i) the successful immobilization of macrophytes as a substitute material for radioactive waste to be immobilized in follow-up studies, (ii) the evaluation of mechanical and physical properties of cemented biological waste during flooding in water of different quality by measuring the compressive strength, porosity and spectroscopic analyses, (iii) the feasibility of using cement as binding material in hazardous waste stabilization processes.

## 2. Materials and methods

### 2.1. Preparation of solidified test specimens

Portland cement (PC), the main stabilizer used in this study, is a local cement manufactured according to the Egyptian Standard Specifications ES 4756-1/2005 and EN 197-1/2004 (Egyptian Standard Specifications, 2005). Green species of *V. anagallis-aquatica*, a semi-aquatic plant, thrive in various moist and wet habitats; e.g., they occupy Egyptian fresh waterways such as El-Manayf Canal (a branch from the Nile River) near Ismailia (Bayoumi, 2012). For the stabilization/solidification process, the plants were collected, washed with tap water, dried in an oven at 50 °C, ground to powder by a blinder, and blended with cement to create biomass-to-cement mixtures with biomass fractions of 0%, 1%, 2%, 3%, and 4% wt/wt. (Bayoumi, 2013). Table 1 shows the analysis results of Portland cement and dried plants, which were applied in this research.

### 2.2. Flooding the test specimens

After the blending process, the biomass/cement samples were hydrated by adding water at a proportion of 35% (wt./wt.) water/cement Resulting cemented biological waste paste was mixed thoroughly and poured in polyethylene (PE) cylindrical molds, compacted manually, closed tightly and allowed to set and harden at a temperature of 25 ± 5 °C under humid conditions for 28 days according to the test protocol specified by ASTM C31 2000. Afterwards, the PE cylinder covers were removed, solid cylindrical blocks were obtained and immersed in different aqueous media for various periods according to the International Standard Organization (ISO) 6961/82 Standard Method for "Long-Term Testing of Solidified Radioactive Waste Forms". Flooding conditions typically prevailing in the repository environment were simulated in the laboratory by statically immersing the solidified biological waste in tap, ground or sea water (Saleh and Eskander, 2009; Eskander et al., 2013). Analysis results for significant ions present in the aqueous media are shown in Table 2.

### 2.3. Mechanical stability testing of test specimens

Average values of consistency and porosity with and without immersion for extended periods were evaluated. Therefore, after predetermined immersion periods, selected solid blocks were removed from the immersion vessel, weighted and subjected to evaluation of durability, porosity and other experimental investigations to assess their stability. Compressive strength measurements were recorded based on the hydraulic press using a Ma-Test Measuring Machine E-159 SP, Italy. Porosity and water absorption parameters for hard blocks were determined according to the boiling water technique (ASTM C20 2015). As a mass change indicator during extended periods of immersion, the water content was determined for each aggregate fraction.

**Table 1**
Compositions of Portland cement and dried biomass.

| Oxides of Portland cement, % (wt./wt.) | | | | | | | |
|---|---|---|---|---|---|---|---|
| $SiO_2$ | $Al_2O_3$ | $Fe_2O_3$ | CaO | MgO | $K_2O$ | $SO_3$ | Insoluble residues |
| 19.84 | 4.74 | 4.0 | 61.01 | 2.5 | 0.6 | 2.4 | 0.95 |
| Elemental composition of dried biomass, % (wt./wt.) | | | | | | | |
| H | N | C | | | | | |
| 2.86 | 2.98 | 20.44 | | | | | |

**Table 2**
The concentrations of some common ions in tap, ground and sea water.

| Media | pH | Soluble cations (ppm) | | | | Soluble anions (ppm) | | |
|---|---|---|---|---|---|---|---|---|
| | | $K^+$ | $Na^+$ | $Mg^{2+}$ | $Ca^{2+}$ | $Cl^-$ | $SO_4^{2-}$ | $HCO_3^-$ |
| Tap water | 6.90 | 0.08 | 1.07 | 1.2 | 1.4 | 0.77 | 0.7 | 1.8 |
| Ground water | 7.20 | 23 | 149 | 13 | 74 | 137 | 317 | 272 |
| Sea water | 7.93 | 8.4 | 652.6 | 96.9 | 28.06 | 496.9 | 60.8 | 183 |

### 2.4. SEM-EXD, FT-IR, and XRD characterization of test specimens

Fractured pieces of the solid biological waste were investigated under scanning electron microscopy (with magnifications between 300× up to 5000× using SEM – JEOL JSM-5600LV with accelerating voltage of 10 kV) equipped with an EDX unit (SEM/EDX). In addition, ground samples were investigated based on IR and XRD techniques using an IR spectrometer (NICOLET-iS10Thermo Scientific) and a X-ray diffractometer (PANalytical's X'Pert PRO, The Netherlands), respectively.

## 3. Results and discussion

### 3.1. Mechanical testing

Specimens originating from encapsulation of 3% biological waste in cement revealed more than 13 MPa of compressive strength (Fig. 1). However, this value is more than the minimum criterion of compressive strength for solidified waste, i.e., 3.45 MPa, as suggested by US Nuclear Regulatory Commission (NRC) Standard. This value by far fulfills the transportation and disposal requirements such as mechanical, physical and chemical stabilities during various caustic weathering events (Bayoumi, 2013); consequently, this 3% mixture was applied for the further investigations in this study. Literature reports that compressive strength values higher than 50 MPa can be obtained for cement-based solidified test specimens after addition of defined quantities of kaolin clay; this clay addition also decreased the long-term leaching rate of radionuclides (Saleh, 2014b; Osmanlioglu, 2002). Here, it has to be emphasized that, different to our research, those studies did not involve encapsulation of any biological materials; further, application of kaolin clay might significantly contribute to the overall process costs.

Results of determined total porosity and stability both immersed and not immersed specimens are shown in Fig. 1. Regarding compressive strength, the integrity is nearly the same or higher than the initial value recorded for the untreated control samples. By subsequently evaluating three series of hardened biological waste after various periods of leaching in different types of water, it could be stated that:

i. After 180 days of leaching, no macroscopically visible disintegration or cracking were observed in all samples.
ii. Results of integrity and porosity of cemented biological waste specimens leached in water of various compositions indicated their beneficial mechanical properties.
iii. Negligible drop in integrity was observed in samples cured in tap water. On the contrary, considerable increase in mechanical