

Immobilization of Borate Waste Simulate in Cement–Water Extended Polyester Composite Based on Poly(ethylene terephthalate) Waste, Part 3: Behavior of Solid Waste Form During a Scenario of Flooding Accident in the Disposal Site

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ABSTRACT: Laboratory scale testing program was conducted to investigate the feasibility of using water extended polyester cement composite (PCC) based on poly(ethylene terephthalate) waste (PET) as immobilization matrix for borate waste, originated from pressurized water reactors (PWRs), during prolonged term of disposal process and in scenario of flooding accident. Twenty-eight days cured solid blocks prepared as a premix of 3% by weight of the water extended polyester (WEP) and ordinary Portland cement (OPC) past having 40% by weight water were subjected to static immersion in three type of leachants, namely tap, sea, ground water. Weight changes, compressive strength variations, porosity, water absorption, and some other physical properties of the solid PCC waste forms were evaluated during 260 days of the static

immersion. X-ray diffraction (XRD), infrared analysis (IR), and electron scanning microscope were performed for the waste forms at the end of immersion prolonged period to follow the internal changes in the solid waste forms that may occur due to the corrosion action of the different leachants. The results obtained from the different measurements confirm the performance of the PCC solidified waste form, based on the mentioned formulation, as an acceptable matrix in the various disposal sites and for extended disposal time even at the worst flooding environment. © 2007 Wiley Periodicals, Inc. *J Appl Polym Sci* 104: 4113–4120, 2007

Key words: disposal; different leachants; PET waste, polyester cement composite; mechanical durability

INTRODUCTION

According to the International Atomic Energy Agency (IAEA), the disposal of radioactive waste is defined as the emplacement of hazardous wastes in an approved specific facility that is intended to isolate the waste from human and environment aiming at their protection.¹

Disposal process represents the last step in the waste management schemes. Near field, deep geological, ice sheet, sea, subsea, . . . are some proposed options for the disposal of radioactive and hazardous wastes.^{2–7} These options are applicable to various wastes, which would be conditioned and/or packed before disposal.

In addition to the many tasks that arise from the safety handling, transport, and storage, the prime role of a candidate solid waste form matrix is the

barrier function to the immobilized radionuclide migration from the radioactive monolith to the environment during the prolonged disposal time.

In the repository, during a postclosure period of hundreds of years, the final waste forms will be subjected to variable aggressive elements in the environment. The breakthrough of water inside the disposal site is one of these aggressive elements, which will expose the final waste forms to chemical ingredients of water that influence its external and internal structure. The dissolution phenomenon is considered to be one of the most important factors in the disposal process and that affect in the physical and chemical properties of cementitious structure. The dissolution-induced alteration is considered to consist of the leachant of cement constituents into the pore water and then transportation of these leached constituents to the environment.⁸

This study is part of a comprehensive program, aims at evaluating the applicability of polymer cement composite (PCC) waste form, composed of the ordinary Portland cement (OPC) and water extended polyester (WEP) based on the recycled poly(ethylene

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TABLE I
Chemical Composition of Ordinary Portland Cement (OPC) (wt %)

Chemical compositions (wt %)	Calculated compound composition (%)
SiO ₂ (19.84)	C ₃ S (53.11)
Al ₂ O ₃ (4.74)	C ₂ S (16.89)
Fe ₂ O ₃ (4.0)	C ₃ A (5.81)
CaO (61.01)	C ₄ AF (12.16)
MgO (2.5)	
K ₂ O (0.6)	
SO ₃ (2.4)	
Insoluble residue (0.95)	

terephthalate) (PET), coming from consumed soft drink bottles, as an encapsulating matrix for borate waste solution that originates from the pressurized water reactors (PWRs).

In this part of work the behavior of the PCC waste form during an accident of flooding disposal sites with any of the three different types of water was followed. Infrared, X-ray diffraction analyses, and electron scanning microscope were used to trace the changes that may occur during the immersion course. Mechanical integrity, weight change, porosity, bulk density, water absorption, specific gravity, . . . of the candidate PCC were some of the selected parameters for laboratory testing under the immersion conditions in any of the three different types of water during a period of 260 days.

Methodology

Preparation of the PCC waste form

The unsaturated polyester (UP), based on the glycolysis of PET, waste is prepared by mixing the shredded soft drink bottles with propylene glycol and diethylene glycol, and then the products of this glycolysis process are reacted with maleic anhydride and adipic acid. Styrene monomer, in turn, is added to the resulting UP at a ratio 40 : 60 wt:wt % to give styrenated polyester (SP). On the other hand, WEP is obtained by dispersing 20% of distilled water containing 0.2 mmol/g diethanolamine into 80% of the obtained SP. Finally, the PCC is obtained by premixing 3% by weight of WEP with cement paste, which

was prepared through the hydration of OPC (Table I), with water at *w/c* ratio equal to 0.40. The obtained PCC slurry is poured into polyethylene molds and agitated vigorously by hand to keep the WEP suspended in the cement paste. The hydrated PCC was allowed to set and hard for 28 days at room temperature ($30 \pm 5^\circ\text{C}$) in humid atmosphere. More details for the preparation of the UP, WEP, and PCC are fully described in previous works.^{9,10} At the end of curing period, a well standing monolithic cylindrical blocks of PCC waste form are obtained.

Immersion process

The worst-case flooding scenario for the repository environment was simulated in the laboratory by immersing statically the 28 days cured cylindrical solid specimens of the PCC in any of three different types of leachant, namely, tap water, ground water, and sea water. The analyses of some ions of interest in the three types of water are represented in Table II successively. After a predetermined immersion period, solid blocks and leaching water were separated by decantation. The solid blocks were air dried and their changes in weight, compressive strength, porosity, water absorption, and bulk density were evaluated.

Compressive strength measurements were carried out for three to five blocks with height 6.0 ± 0.2 cm and diameter 3.10 ± 0.05 cm using Ma-Test Measuring Machine E-159 SP, Italy. The porosity was measured for the immersed blocks according to boiling water saturation (BWS) technique following ASTM C 20-74.

The measurements of pH for the leachant solutions were performed using Orion Research Model 701 A Digital Ionalyzer, USA, at the end of each immersion periods. Also, the soluble calcium contents in the leachant solutions were determined using Varian SpectrAA 220 Atomic Absorption Spectrometer, Varian, and Australia.

To follow the changes that may takes place internally for the cured PCC blocks at the end of the immersion test, X-ray diffraction (XRD) and infrared analysis (IR) were followed. For XRD, Diamo Corporation (USA) equipment equipped with Co K_α radiation and Fe filter, $\lambda = 1.79026\text{\AA}$, 2θ range from 10 to 80°, step size = 0.1°, step time = 1 s was used.

TABLE II
Concentrations of Some Ions of Importance in Different Types of Water Used for Immersion (A) Tap Water (Giza district); (B) Ground Water (Abu Zaabel Well No. 202); (C) Sea Water (Mediterranean Sea, Alexandria)

pH	Soluble cations (ppm)				Soluble anions (ppm)		
	K ⁺	Na ⁺	Mg ⁺⁺	Ca ⁺⁺	Cl ⁻	SO ₄ ⁻⁻	HCO ₃ ⁻
6.74	0.09	1.09	1.21	1.40	0.77	0.77	1.8
7.2	23	149	13	74	137	317	272
8.02	8.4	652.2	96.9	28.1	496.9	60.8	183

On the other hand, the IR analysis was carried out using FTIR, model 670, Nicolet, MA.

For the study of particle morphology and mass microstructure the Jeol JXA-840A Electron Probe Microanalyzer Scanning electron microscope (Japan) was employed with stage magnification of 2500 \times . The actual pretreatment and preparation consisted only of the gold plating of the samples in S150A Sputter Coater Edwards (England).

RESULTS AND DISCUSSION

Water is at the heart of most of the physical and chemical causes underlying the deteriorations of concrete structures.¹¹ Cement-based solid products exposed to water undergo carbonation process that refers to the reaction of the carbon dioxide, in the surrounding, with water to form carbonic acid, which then combined with calcium ions leached from the hydrated cementitious materials to form insoluble calcium carbonate salts (CaCO_3). Carbonation phenomenon proceeds from the surface of the cured cement products inwards but does so extremely slowly.¹²

A white thin layer (≈ 0.1 mm thickness) start to appear very clearly on the surface of solid PCC blocks immersed statically in sea water after 30 days. On the other hand, by the visual examination of PCC blocks immersed in ground water, this layer is less obviously seen, while for those immersed in tap water this layer is hardly detected even after 260 days of immersion. This may be due to the high salt contents of both sea and ground water that affect highly the leaching of lime [$\text{Ca}(\text{OH})_2$] from the surface of the PCC and its reaction with the carbon dioxide, of surrounding, forming the thin layer of calcite (CaCO_3). To confirm this reaching, this thin layer was scratched from the surface of PCC blocks that immersed in sea water for 260 days, and subjected to XRD and the obtained diffractogram is represented in Figure 1. Peaks at $\delta = 1.63, 1.93, 2.63, 5.42 \text{ \AA}$. . . are refer to the lime [$\text{Ca}(\text{OH})_2$] leached from the PCC blocks, while that at $\delta = 4.91 \text{ \AA}$ may attributed to the formed calcite (CaCO_3). The proposed reactions for the formation of this layer can be illustrated as follows. The acidity of naturally occurring water is generally due to the dissolved carbon dioxide (CO_2), which is found in significant concentration in mineral, sea, and ground water as previously stated



A cation exchange reaction between carbonic acid and calcium hydroxide [$\text{Ca}(\text{OH})_2$] leached from the hydrated PCC processed as follows

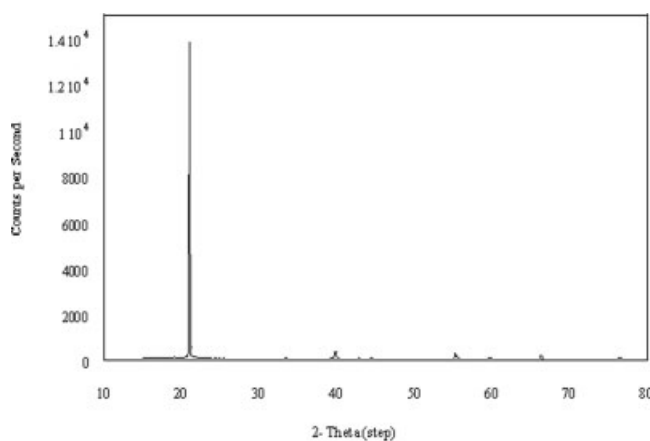
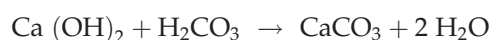


Figure 1 X-ray diffraction of the white thin layer scratched from the surface of polymer cement composite (PCC) blocks immersed in sea water after 260 days (X-axis): 2θ (step); (Y-axis): counts per second.



This leads to the precipitation of the insoluble calcium carbonate salts (CaCO_3) and step further ingress of water into the interior of PCC products through reduction in permeability. This kind of protection would be available under static conditions. Nearly, similar behavior for the carbonation of concrete components was discussed in the literature.¹³

Figure 2 represents the X-ray diffractogram for nonimmersed plain cement in addition to (PCC) solid blocks cured for 28 days and after 260 days of static immersion at room temperature ($30 \pm 5^\circ\text{C}$) in the three types of water. It is clear that no eventual differences for the peaks positions for immersed and nonimmersed blocks were detected. The only difference is the high propagation of peak at the 2θ value 21.00° for PCC blocks compared to both nonimmersed and for plain cement. This peak may attribute to the growth of Portlandite crystals (CH) formed during the hydration of C_3S . A polymer coat is formed around the CH crystals, inhibits their growth and results in the formation of large numbers of small CH crystals rather than a few large crystals.¹⁴ Also this highly propagated peak may practically shared by the peak attributed to the calcite formed during the carbonation process.

On the basis of the XRD data, it could be concluded that no serious internal changes take place for hard PCC immersed in the three types of water for 260 days.

The IR spectra after 260 days immersion in the three different leachants for the PCC candidate as waste form for solidification of borate waste that originated from PWRs are shown in Figure 3. It is clear from the figure that the only difference between the non-immersed solid specimens and statistically

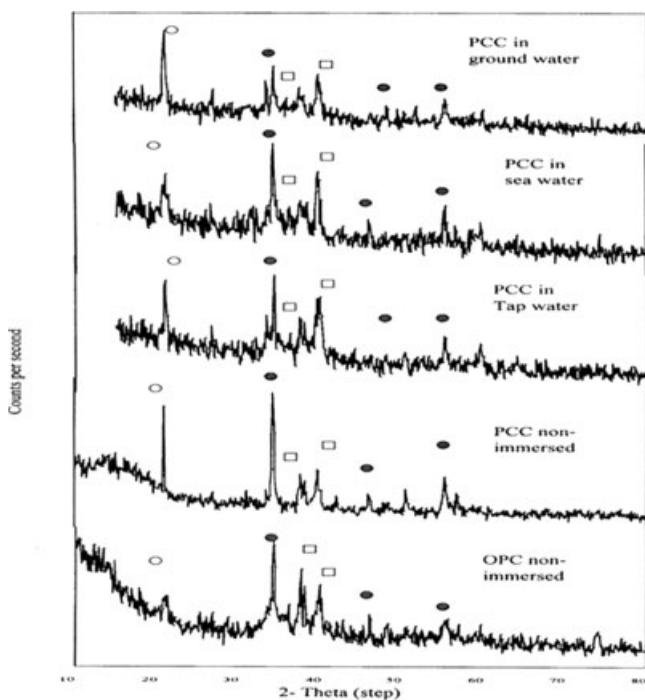


Figure 2 X-Ray diffractogram of ordinary Portland cement (OPC), polymer cement composite (PCC) before and after immersion in different leachants for 260 days [○: CH, ●: C–S–H, □: C₃S and C–C] (X-axis): 2θ (step); (Y-axis): counts per second.

immersed ones is due to the propagation of the band around 1640 cm^{-1} . This band may refer to the stretching frequencies of the carbonyl group ($-\text{C}=\text{O}$) formed due to the carbonation process. This result supports that previously obtained from XRD analysis for PCC, and consequently affirms the ability of the candidate PCC waste form to withstand the deterioration even after the immersion for 260 days in any of the three types of leachant.

Very little information in the literature is helpful in discussing the way of degradation of the PCC solid waste forms subjected to the principle characteristics of the environment. For this reason, a research program on the durability of the candidate PCC was initiated by our Radioisotope Department, Atomic Energy Authority in Egypt, as a part of the waste forms characterization. Immersion experiments were carried out, statistically, addressing the effect of leachants type and/or the time of immersion on the following parameters:

- Weight change as an index of the waste form durability.
- Variations in compressive strength of the candidate matrix as a function of immersion conditions.
- Porosity, water absorption, bulk density, specific gravity of the PCC at the end of each immersion period.

- Leaching of calcium from the PCC waste forms in the different leachants during the immersion process.

The performance of PCC solid blocks exposed to tap; sea and ground water were evaluated by measuring the weight changes, at different immersion periods, due to the water element attack. Figure 4 describes the weight behavior of PCC and OPC 28 days cured specimens and in the three different leachants as a function of square root of immersion periods. It is clear from the figure that a linear relationship between the weight changes and the square root of time is exhibited. Hence, it can be a clue for that a diffusion mechanism is controlling the permeability of the different immersing water ingredients. Also, it is worth mentioning that the candidate PCC matrix is more stable to the three leachants than the OPC blocks immersed in the same conditions (Fig. 4). As a result, it could be concluded that the PCC matrix candidate for immobilization of borate wastes can be highly withstand the various environmental disposal conditions even under very aggressive conditions, i.e. the flooding of the disposal site for more than 8 months.

It is a relevant approach for accessing porosity when interested in certain concrete durability aspects.¹⁵ Porosity and other relating parameters for PCC blocks immersed in the three different types of water and for different periods was determined according to ASTM C20-74 based on BWS technique

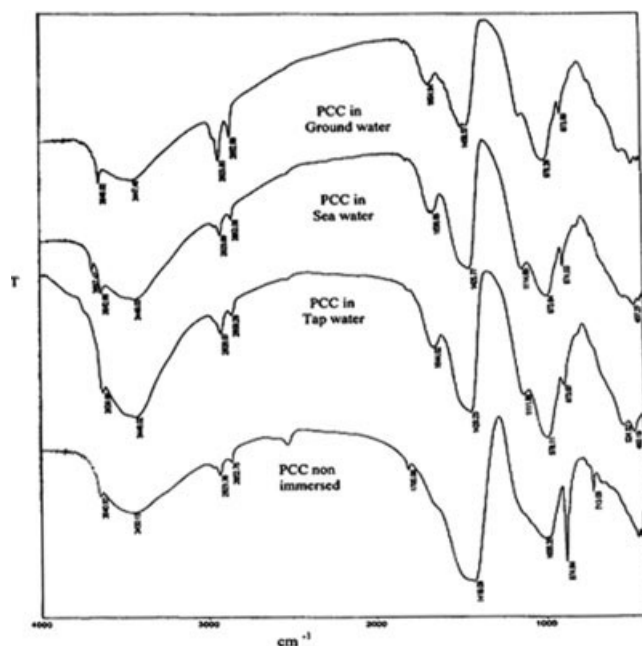


Figure 3 IR spectra of polymer cement composites (PCC) before and after immersion in different leachants for 260 days (X-axis): Wave numbers (cm^{-1}) (Y-axis): %transmittance.

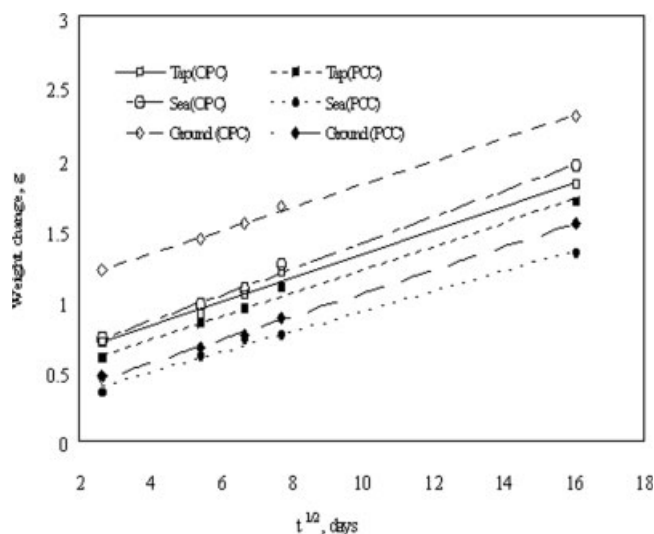


Figure 4 The change in weight of ordinary Portland cement (OPC) and polymer cement composite (PCC) blocks immersed in three leachants as function of square root of immersion time in days (X-axis): $t_{1/2}$, days (Y-axis): Weight change (g). □: tap (OPC); ○: sea (OPC); ◇: ground (OPC); ■: tap (PCC); ●: sea (PCC); ◆: ground (PCC).

as previously stated and the data obtained are represented in Table III for tap, sea, and ground water, respectively. Generally, it was observed that both porosity and water absorption percentages increase for the PCC waste form immersed in tap water to reach 39.55 and 24.44%, respectively, after 260 days of immersion at $30 \pm 5^\circ\text{C}$ compared to 32.77 and

20.28% for control non-immersed specimens (Table III). On the other hand, very slight increase in the two parameters was detected for the PCC specimens immersed in ground water at the same conditions of immersion up to 30 day. However, it should be noted that the values of porosity and water absorption percentages decline again to reach 31.93 and 18.12%, respectively, after 260 days compared to 32.77 and 20.28% for non-immersed blocks. For the PCC blocks immersed in sea water, slightly different trend was observed where a detectable increase in porosity and water absorption percentages occur within the first 7 days, then a high decrease takes place to reach a minimum value after 260 days (i.e. 26.17 and 15.36%, respectively) (Table III). Generally speaking, the increase in the porosity values may refer to the leaching of the cementitious materials due to the diffusion of water into the pores of PCC specimens and removing lime and calcium sulfate components.¹⁶ While the reduction in porosity values for the PCC blocks immersed in both ground and sea water afterwards can be attributed to the carbonization process where the calcite protective layers claim to be formed and fill the pores of immersed blocks as previously described.¹⁷ The pronounced reduction in the porosity and water absorption percentages for the PCC immersed in sea water may be due to the high salinity of the sea leachant solution compared to tap and ground water (Table II).

A direct relationship can facilitate the mechanical integrity of the concrete composite from the knowledge of its pore system characteristics.¹⁸ The results

TABLE III
Physical Properties of Polymer Cement Composite (PCC) Subjected to Immersion Test at Different Periods and in Different Leachants

Immersion time (days)	Leachants	Exterior volume V (cm ³)	Volume of impervious portion (cm ³)	Volume of open pores (cm ³)	Apparent porosity P (%)	Water absorption A (%)	Apparent specific gravity T	Bulk density B (kg/cm ³)
Zero	–	26.85	18.05	8.8	32.77	20.28	2.4	1.62
7	T	27.32	18.12	9.20	33.67	20.99	2.42	1.60
	S	26.32	16.68	9.68	36.78	22.54	2.58	1.63
	G	28.17	20.2	7.97	34.71	20.84	2.18	1.56
30	T	24.99	16.47	8.52	34.09	19.96	2.59	1.71
	S	25.55	16.87	8.68	33.50	20.15	2.55	1.69
	G	23.7	15.54	8.16	34.43	19.24	2.73	1.79
45	T	25.16	16.40	8.99	35.41	20.58	2.66	1.72
	S	25.53	17.66	7.87	30.83	17.62	2.53	1.75
	G	25.55	17.04	8.72	33.85	19.81	2.58	1.71
60	T	25.19	16.02	9.17	36.4	21.5	2.66	1.69
	S	26.58	18.26	8.32	31.30	19.31	2.36	1.62
	G	26.64	17.92	8.72	32.73	19.82	2.45	1.65
260	T	25.26	15.59	10.2	39.55	24.44	2.68	1.62
	S	26.63	19.66	6.97	26.17	15.36	2.31	1.7
	G	24.96	16.99	7.97	31.93	18.12	2.59	1.76

T, Tap water; S, sea water; G, ground water.

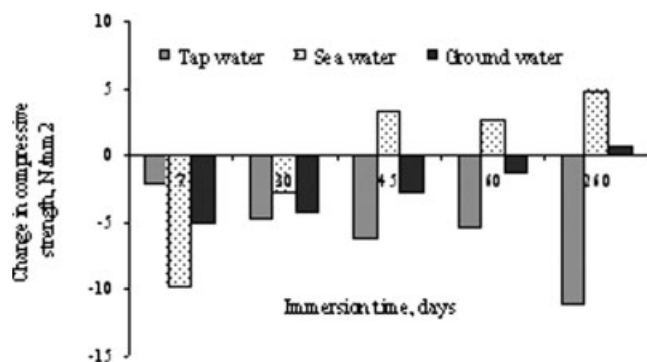


Figure 5 The variation in compressive strength integrity of polymer cement composites (PCC) due to the immersion period in the different leachants. (X-axis): Immersion time, days (Y-axis): Change in compressive strength (MPa).

obtained for the porosity and pore parameters measurements were confirmed by the compressive strength measurements for the PCC cured waste forms immersed in the three different leachants for different immersion periods and their strength were measured at the end of each period up to 260 days. Figure 5 represents the change in compressive strength of PCC blocks immersed in the different types of water for different periods compared to nonimmersed ones. It is clear that lose in compressive strength was observed for the all PCC immersed in the three leachant after 7 and 30 days. This result confirms the result obtained from porosity measurements. Similar trend was reached for similar concrete products immersed in ground water.¹⁹ The lose in compressive strength for blocks immersed in tap water is continued while gain in compressive strength was obtained for blocks immersed in sea and ground water after 45 and 260 days, respectively (Fig. 5).

The initial drop, as previously explained, may be due to the leaching of the cement constituents, which result in formation of microvoids, while the gain in compressive strength can refer to the deposition of the insoluble calcite (CaCO_3) salt inside the PCC pores leading to its filling. These results are in a satisfactory agreement with similar published work and are confirmed by previously XRD and porosity measurements.²⁰ However, it is worth mentioning that in the case of PCC blocks immersed in tap water even with the lose in compressive strength integrity, the PCC waste forms still have compressive strength values largely greater than that recommended for transportation, handling, and disposal of final radioactive waste solid waste forms.²¹

Porosity and mechanical integrity measurements affirm that the PCC waste form made from OPC and water extended polyester based on recycled PET candidated for borate waste incorporation can with-

TABLE IV
The pH of Leachant Solutions at the End of Different Immersion Periods

Immersion period (days)	Tap water	Sea water	Ground water
Zero	6.72	8.02	7.2
7	11.1	10.7	10.9
30	11.1	10.9	10.9
45	11.1	11.0	11.0
60	11.1	11.0	11.0
260	11.1	11.0	11.0

stand the different disposal environments even in the flooding the disposal site for long periods.

Theoretically, any environment with pH less than 12.5 may become aggressive to the cementitious products due to the reduction in the alkalinity of the pore fluids, which would eventually led to destabilization of the hydration products.²² However, our candidate PCC waste form shows acceptable properties, visually and experimentally, in the three immersion solutions even at pH values less than 12.5 (Table IV).

The change in chemical constituents of the cementitious waste forms due to leaching has been intensively studied because it is assumed to affect the transportation of radioactive nuclides and the surrounding environmental chemistry in the repository.⁸ The leaching of soluble calcium after 260 days from 28 days cured PCC blocks immersed in the three leachant solutions is shown in Figure 6. It is

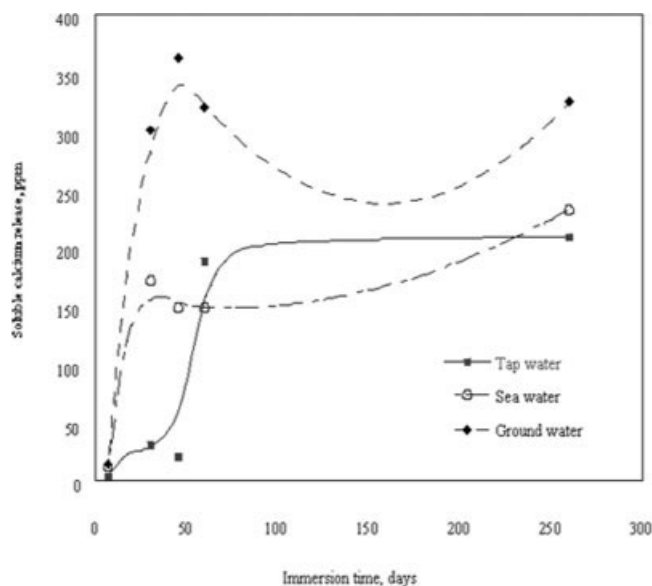


Figure 6 The release of calcium from polymer cement composite blocks immersed in the leachant solutions versus the immersion periods (X-axis): Immersion time, days (Y-axis): soluble calcium release (ppm) ■: Tap water; ○: sea water; ◆: ground water.

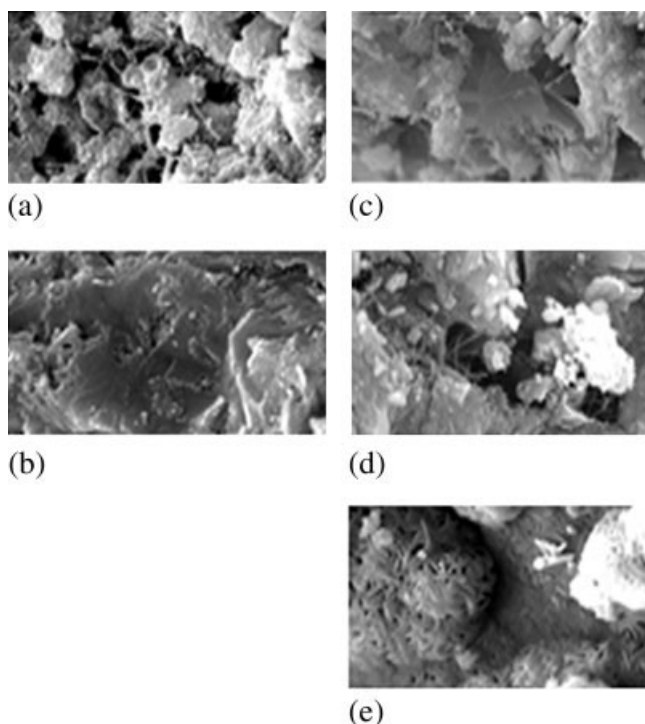


Figure 7 Scanning electron micrographs of ordinary Portland cement (OPC), polymer cement composites (PCC) before and after immersion in different leachants for 260 days. (a) OPC nonimmersed; (b) PCC nonimmersed; (c) PCC in tap water; (d) PCC in ground water; (e) PCC in sea water.

clear from the figure that the highest released of soluble calcium was detected for PCC blocks immersed in sea water and reached ≈ 360 ppm after 45 days. This value represents nearly 0.37% of the Ca added as CaO in OPC. It is worth mentioning that the release of soluble calcium declines after 45 days to reach only 320 ppm at 60 days showed a tendency to coverage into a constant value for the next 260 days. The decrease in the Ca release may be due to the carbonation process that takes place for the PCC blocks during the immersion in the sea water. However, it could be predicate that, supposing the permanent flooding condition the disposal site, more than 200 years are needed for the complete depilation of calcium from the PCC blocks. However, it should noted that, although the PCC blocks immersed in ground and tap water exhibit the same trend, the released Ca in both cases are less than that released from PCC blocks immersed in sea water (Fig. 6). These results lead to the suggestion that the candidated PCC composite made of ordinary Portland cement at w/c ratio 40% and containing 3% WEP based on PET waste could withstand the worst conditions of flooding of the disposal site.

Figure 7(a,b) shows the scanning electron micrographs ($2500\times$ magnification) of portion of a labora-

tory scale non-immersed plain cement, ($w/c = 0.40$) and cement-polymer composites ($w/c = 0.40$ and $c/p = 3\%$) samples. Figure 7(c-e) represents scanning electron micrograms of cement-polymer composite specimens, having the same composition to that of non-immersed in three leachants (tap, ground, and sea water) for 260 days.

C—S—H, which represent the main hydration products of the OPC, have foil- and plate-like crystals [Fig. 7(a)]. On the other hand, Portlandite (CH), i.e. crystalline calcium hydroxide, which represents the second most abundant product in the hydrated cement paste, appears in the form of relatively large crystalline aggregate and suppose to be intimately mixed with the C—S—H.¹⁶ Their entanglement makes it hard to define their exact shape (Fig. 7(a)).²³ It is clear from Figure 7(b) that the surface of the non-immersed PCC based on PET waste is characterized by a continuous large polymer, mutually intergrown with considerable number of compact points and areas connecting the cement paste and forming the binding phase of the products.²⁴ Therefore, the hydration products of cement formed in the polymer layers cannot be distinguished out right by their appearance [Fig. 7(b)]. The fracture of the cement-polymer composite samples after immersion in tap water is characterized by porous surface [Fig. 7(c)], and its feature after that is comparable to the analogous sample before immersion [Fig. 7(b)]. On the other hand, the PCC specimens immersed for the same period in ground and sea water described by peculiar qualities to each other [Fig. 7(d,e)] but differ from these immersed in tap water [Fig. 7(c)], in the predominant presentation of calcite crystals. However, it is worth mentioning that the calcium carbonate crystals in the micrograph for samples immersed in sea water [Fig. 7(e)] are abundant, widespread, and lump. SEM imaging gives detailed information about the structure of the specimens of PCC before and after immersion in the three leachants, and consequently confirms the physicomachanical results obtained for the behavior of the solid waste forms in the disposal environment.

CONCLUSIONS

Based on infrared analysis, X-ray diffraction, electron scanning microscope, weight and compressive strength evaluations, porosity and other pore parameter measurements, it could be stated that: the proposed PCC made from OPC and water extended polyester obtained from recycled PET can efficiently withstand the aggressive flooding condition in various disposal sites. Also it means that the PCC waste form that candidated to immobilize borate waste can be disposed of in any environment safely.

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