



This is a repository copy of *Leaching behaviour of cementitious nuclear wastefoms containing caesium and strontium*.

White Rose Research Online URL for this paper:  
<http://eprints.whiterose.ac.uk/97728/>

Version: Accepted Version

---

**Article:**

Gashier, W., Miura, T., Hashimoto, K. et al. (2 more authors) (2014) Leaching behaviour of cementitious nuclear wastefoms containing caesium and strontium. *Advances in Applied Ceramics*, 113 (8). pp. 447-452. ISSN 1743-6753

<https://doi.org/10.1179/1743676114Y.0000000146>

---

This is an Accepted Manuscript of an article published by Taylor & Francis in *Advances in Applied Ceramics* on 18/03/2014, available online:  
<http://www.tandfonline.com/10.1179/1743676114Y.0000000146>

**Reuse**

Unless indicated otherwise, fulltext items are protected by copyright with all rights reserved. The copyright exception in section 29 of the Copyright, Designs and Patents Act 1988 allows the making of a single copy solely for the purpose of non-commercial research or private study within the limits of fair dealing. The publisher or other rights-holder may allow further reproduction and re-use of this version - refer to the White Rose Research Online record for this item. Where records identify the publisher as the copyright holder, users can verify any specific terms of use on the publisher's website.

**Takedown**

If you consider content in White Rose Research Online to be in breach of UK law, please notify us by emailing [eprints@whiterose.ac.uk](mailto:eprints@whiterose.ac.uk) including the URL of the record and the reason for the withdrawal request.



[eprints@whiterose.ac.uk](mailto:eprints@whiterose.ac.uk)  
<https://eprints.whiterose.ac.uk/>

# Leaching behaviour of cementitious nuclear waste forms containing caesium and strontium

Wail Gashier\*, Russell J Hand & Hajime Kinoshita\*\*

Immobilisation Science Laboratory, Department of Materials Science & Engineering, The University of Sheffield, Sir Robert Hadfield Building, Mappin Street, Sheffield, S1 3JD, UK

\*e-mail: w.gashier@sheffield.ac.uk, \*\*e-mail: h.kinoshita@sheffield.ac.uk

## ABSTRACT

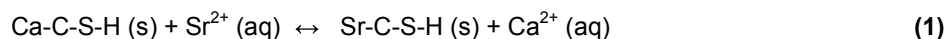
The leaching behaviour and physico-chemical properties of cementitious nuclear waste forms containing caesium and strontium waste simulants has been investigated. The cement wastefrom consisted of a 9:1 blend of blast furnace slag and ordinary Portland cement. Both non-loaded samples and samples that were waste loaded with 3wt% caesium and strontium added as nitrates have been studied. The cement hydration phases in the samples were identified and the porosity and microstructure analysed prior to leaching. The samples were leached for up to 6 months and the leached elements quantified. In the waste loaded cements portlandite was not formed and the monosulphate AF<sub>m</sub> phase appeared to be altered by the incorporation of the Sr(NO<sub>3</sub>)<sub>2</sub>. Incorporation of Cs and Sr also resulted in the increase in the leach rate of Ca<sup>2+</sup>.

**KEYWORDS:** Cement; Waste Encapsulation; Leaching; Caesium; Strontium; Monosulphate

## 1. INTRODUCTION

The radioactive isotopes caesium-137 and strontium-90 are fission products of uranium and plutonium that may be found in intermediate level waste (ILW) [1]. These isotopes are hazardous if released into the environment and should be immobilised in an appropriate matrix. In the UK cement wasteforms are currently preferred for ILW [1], with the most commonly used matrix being ordinary Portland cement (OPC) blended with blast furnace slag (BFS). These wasteforms, contained in stainless steel drums, are stored for an appropriate time to reduce the amount of isotopes such as <sup>137</sup>Cs and <sup>90</sup>Sr [1]. However, the highly mobile nature of these elements can cause significant concern if they are released into the environment. This could happen if there was early failure of the stainless steel drums during the storage period, or after their disposal in an underground repository, where eventually the wasteform will come into contact with groundwater. Therefore it is important to understand the behaviour of Cs and Sr in the wasteform, what phases are likely to be present in relation to these radionuclides, and how they behave under leaching conditions.

It has been reported that both Cs and Sr interact with calcium silicate hydrate (C-S-H) gel [2]. The Cs in OPC cements appears to be mainly sorbed onto the C-S-H gel or to remain in the pore solution [2-4] whereas Sr is thought to be retained in only the C-S-H gel [5,6,7], with the Sr<sup>2+</sup> ions substituting for the Ca<sup>2+</sup> ions:



However, Atkins and Glasser [8] state that although a small amount of Sr is immobilised in C-S-H, the majority of the substitution occurs in the calcium aluminate (sulphate) hydrates, with significant substitution of Sr into ettringite (AF<sub>t</sub> phase). Therefore further research is required to understand the way in which the Cs and Sr are retained in the wasteform because this will govern their rates of dissolution and may impact the dissolution of other elements in the cement matrix.

The paper describes an investigation into the leaching behaviour of model cementitious nuclear wasteforms containing Cs and Sr. Excess amounts of Cs and Sr were incorporated in a 9:1 blend of blast furnace slag (BFS) and OPC cement. The leaching behaviour of these elements, as well as their effects on the cement phases and microstructure, were studied.

## 2. EXPERIMENTAL

### 2.1 Sample Preparation

Cement samples shown in (table 1) were prepared using OPC and BFS (table 2). The OPC was sourced from Castle Cement, UK, with a fineness of 352m<sup>2</sup>/kg Blaine and the BFS was sourced from Redcar Steelworks, Teeside, UK in ground granulated form to a fineness of 286m<sup>2</sup>/kg Blaine. As waste simulants, CsNO<sub>3</sub> (Acros Organics, UK, purity 99.99%) and Sr(NO<sub>3</sub>)<sub>2</sub> (BDH Chemicals, UK, purity 99.99%) were added to the cement matrices.

The blank sample (labelled B) was prepared by firstly weighing the distilled water on a Toledo Delta Range PG 4002-5 electronic balance and pouring into the bowl of a Kenwood mixer. The designated amount of BFS was added followed by the OPC to avoid any hydration of the OPC before mixing with the BFS. The water to solid (W/S) ratio used was 0.37 as this is widely used in the UK nuclear industry [1]. The water and cement were mixed for approximately 7 minutes on mixer setting 1, until a homogenous cement paste was formed. This paste was then poured into 45 ml centrifuge tubes, which were then sealed and placed in a vortex mixer to remove any air bubbles within the paste. In the samples containing either Cs or Sr or both (labelled C, S and CS respectively), the CsNO<sub>3</sub> and/or the SrNO<sub>3</sub> was added to the water prior to the BFS and OPC and mixed using the Kenwood mixer on speed setting 1 until completely dissolved to ensure homogenous distribution of the waste simulants throughout the cement. The rest of the mixing procedure was the same as for sample B. All samples were cured for 42 days in an environmental chamber at 20°C and 95% relative humidity.

**Table 1:** Cement formulations

Sample	Cement matrix (g)			Waste simulant (g)		Total (g)
	OPC	BFS	H <sub>2</sub> O	CsNO <sub>3</sub>	Sr(NO <sub>3</sub> ) <sub>2</sub>	
<b>Blank (B)</b>	80	720	296	-	-	1096
<b>3wt% Cs and 3wt% Sr (CS)</b>	80	720	296	54.6*	90.6*	1241.2
<b>3wt% Cs (C)</b>	80	720	296	49.8*	-	1145.8
<b>3wt% Sr (S)</b>	80	720	296	-	89.9*	1206.3

\* The amount of waste simulants were calculated to provide 3 wt% of elemental Cs and/or Sr with respect to the total weight of the mix (OPC + BFS + water + nitrates).

**Table 2:** Oxide weight percentage present in OPC and BFS

Oxides	Percentage in OPC (wt%)	Percentage in BFS (wt%)
CaO	64.58	42.1
SiO <sub>2</sub>	20.96	34.5
Al <sub>2</sub> O <sub>3</sub>	5.24	13.74
Fe <sub>2</sub> O <sub>3</sub>	2.61	0.97
MgO	2.09	7.29
SO <sub>3</sub>	2.46	-
K <sub>2</sub> O	0.59	0.49
Na <sub>2</sub> O	0.28	0.22
Chloride	0.048	0.022
Insolubles	0.27	-

## 2.2 Sample Characterisation

Prior to leaching, a sample from each formulation was characterised using X-ray diffraction (XRD), thermogravimetric analysis (TGA), scanning electron microscopy (SEM) and mercury intrusion porosimetry (MIP). For XRD the samples were crushed, hydration was arrested in acetone for 7 days and dried during the preparation for analysis. A small amount of the crushed samples was ground using a pestle and mortar and sieved to <63 µm. A Siemens D500 diffractometer was used with CuK<sub>α</sub> radiation (30kV and 40mA). A scanning speed of 1°/min between 5° and 60° 2θ was used with a step size of 0.02°. For TGA some of the powder was weighed to 40 – 60 ± 0.1 mg, placed in an alumina crucible and heated at a rate of 10°C/minute from 25°C to 900°C in an N<sub>2</sub> atmosphere.

A cross-section was cut from sample CS for SEM, mounted in epoxy resin and successively ground using 120, 400, 800 and 1200 grit SiC papers, diamond polished using 6, 3, 1 and ¼ microns and finally polished using 0.05 Micron Buehler Master Prep Polishing Suspension on a Kement 300 manual polisher. The polished surface was carbon coated using an Edwards 'speedivac' carbon coating unit and examined using an FEI Inspect F SEM in backscattered imaging mode. Energy dispersive spectroscopy (EDS, EDAX Genesis) was used for phase analysis and elemental mapping.

The remaining crushed samples B and CS were used to analyse the pore structure by MIP. Each specimen was immersed in acetone at 20°C for 30 minutes under reduced pressure (20°C, 2.33×10<sup>3</sup> Pa), and D-drying (6.666×10<sup>-2</sup> Pa) for MIP was carried out for 24 hours prior to analysis. The pressure range was set from 0.14-228 MPa to estimate the pore size distribution from diameter of 0.0065 to 10µm.

## 2.3 Leaching Test

After curing samples B and CS were cut into 15mm long and 27mm diameter cylinders. The cylinders were placed in epoxy resin leaving one surface exposed with a surface area of 573 mm<sup>2</sup>. Each cylinder was placed in a polypropylene container filled with deionised water (18.3 MΩ cm), collected from a Millipore Direct QUV 3 water deioniser at 25°C. The water was weighed using a microbalance to 238.44g; this amount was used for each sample to obtain a surface area to water volume ratio of 2.4 based on the procedure of Saito et al. [2].

The leaching tests were conducted for 7, 14, 28 days, 3 months and 6 months. The water was replaced after 2, 4, 6, 8, 15, 22, 29, 39 days and then every 10 days until 179 days, again based on the procedure of Saito

et al [2]. During each water replacement, 100ml of the leachate was saved, accumulated and mixed for each leaching period. Once each leaching period was completed 20ml of the accumulated leachate was acidified and analysed using inductively coupled plasma – mass spectroscopy (ICP-MS).

### 3. RESULTS

#### 3.1. X-RAY DIFFRACTION (XRD)

XRD patterns of the unleached samples for each formulation are shown in Fig. 1. The “0D” in the sample notation represents 0 days leaching. The trace for the blank cement sample without waste simulants (sample B) shows the formation of calcium silicate hydrate (C-S-H), portlandite (CH), hydrotalcite (Hc) and monosulphoaluminate (AFm) as expected from the literature [1,3,4,8] in addition to the gehlenite (G) contained in the original BFS.

When CsNO<sub>3</sub> was added to the matrix (sample C) no peaks for CH and Hc could be seen although the other phases present in in sample B were still observed. In addition extra peaks associated with CsNO<sub>3</sub> can be seen suggesting that some of the Cs has not been incorporated into the cement phases. Introduction of Sr(NO<sub>3</sub>)<sub>2</sub> into the system (sample S) also resulted in a different phase assemblage. Peaks for CH and AFm were absent, and peaks for calcium aluminonitrate (AF<sub>M(N)</sub>) were observed. A mixture of these changes in the phase assemblage was observed when CsNO<sub>3</sub> and Sr(NO<sub>3</sub>)<sub>2</sub> were both present (sample CS). CH and AFm were absent, and extra peaks for CsNO<sub>3</sub> and AF<sub>M(N)</sub> were observed.

#### 3.2 THERMOGRAVIMETRIC ANALYSIS (TGA)

Figure 2a shows the thermogravimetry (TG) and differential thermogravimetry (DTG) curves for samples B and CS. The majority of the weight loss in sample B was due to C-S-H and monosulphate (AFm) phases, with the rest of the weight loss, in principle attributable to hydrotalcite (Hc), portlandite (CH), although the XRD results suggest that only small amounts of CH are present or that it is poorly crystalline. A small weight loss associated with calcium carbonate (CaCO<sub>3</sub>) was also observed; carbonation probably occurred as a result of the ground TGA sample coming into contact with air. The majority of the weight loss in sample CS was due to C-S-H, the AF<sub>M(N)</sub> phase and CsNO<sub>3</sub>. Because of the overlap in the weight loss associated with these phases, it is not possible to estimate the total weight loss due to each phase. The TG and DTG profiles for sample C shown in Fig. 2b were very similar to those for sample B (Fig. 2a) except for a small weight loss at 550-600°C attributed to CsNO<sub>3</sub> decomposition [8]. The profiles also show a weight loss attributable to CH, however no peaks were observed in the XRD trace for this phase suggesting that the amount is small, or that it is poorly crystalline.

The curves for sample S were similar to those for sample CS, showing that the majority of the weight loss was due to C-S-H and the AF<sub>M(N)</sub> phases. Decomposition of Sr(NO<sub>3</sub>)<sub>2</sub>, which should appear at 645 °C [9], was not observed. This suggests that Sr was incorporated into the cement phases.

#### 3.3 SCANNING ELECTRON MICROSCOPY (SEM)

A backscattered electron image of sample CS is shown in Fig 3a along with X-ray dot maps of the area highlighted by the dotted line. As shown in Fig. 3b, the EDS spectra of this area shows that Sr and Cs are present. The dot-maps show that both Cs and Sr have localised distributions within the cement matrix. Cs seems to be concentrated in a small area whereas Sr is slightly more dispersed. This may suggest that Cs has not been incorporated into a cement phase whereas Sr has been.

#### 3.4 MERCURY INTRUSION POROSIMETRY (MIP)

Figure 4 shows the pore volume distribution for samples B and CS based on the MIP results. Sample 0DB has a larger volume of pores with a diameter of >1 µm than the sample 0DCS, which may be related to the

1 formulation of the sample. Pores of this size are regarded as extrinsic and are generally attributed to  
2 formulation and processing [10]. The significant majority of pores present within sample B have an average  
3 diameter of around 0.1  $\mu\text{m}$ ; these are known as meso-pores and arise when hydration products such as CH  
4 and AFm grow within cavities and subdivide the larger pores within the cement structure [10].

5 The total porosity was 0.064 ml/g for the ODB sample and 0.096 ml/g for the ODCS. In the ODCS sample a  
6 significant volume of pores have diameters of around 0.01  $\mu\text{m}$ . These micro-pores are considered to be an  
7 intrinsic feature attributed to the C-S-H gel [10], which coincides with the increased amount of C-S-H  
8 observed in the TGA results. The larger amount of C-S-H could also explain why there is a negligible volume  
9 of pores with diameter  $>1 \mu\text{m}$  in the sample CS as the C-S-H gel may have filled this space [11].

### 10 11 12 **3.5 INDUCTIVELY COUPLED PLASMA MASS SPECTROMETRY (ICP-MS)**

13 The amounts of Ca, Ca and Sr in leached into water for samples B and CS were analysed. The normalised  
14 leaching rates,  $\text{NR}_i$ , were calculated using

$$15 \quad \text{NR}_i = \frac{\bar{M}_i}{m_i A_s t} \quad (4)$$

16  
17  
18  
19  
20  
21  
22  
23  
24 where  $\bar{M}_i$  is the average amount of element  $i$  in leachate (mg);  $m_i$  is the elemental mass fraction of element  $i$   
25 in the cement;  $A_s$  is the surface area of cement exposed to solution ( $\text{cm}^2$ ) and  $t$  is the leaching interval (days).  
26 Fig. 5a shows that Ca leached from sample CS at a significantly higher rate than from sample B. Both rates  
27 decreased with time and became nearly constant at around 3 months. Fig. 5b shows that the leaching rate of  
28 Cs was almost three times that of Sr. It is also clear that these elements leach out at a significantly higher  
29 rate than Ca from the same sample.  
30  
31  
32  
33  
34

## 35 **4. DISCUSSION**

### 36 37 **4.1 Incorporation of Cs and Sr**

38 The results show that the introduction of  $\text{CsNO}_3$  and/or  $\text{Sr}(\text{NO}_3)_2$  have different impacts on the cement phases  
39 formed in the system. XRD and SEM indicates that the majority of the  $\text{CsNO}_3$  was not associated with any  
40 other phases. However introduction of  $\text{Sr}(\text{NO}_3)_2$  led to the absence of the portlandite phase  $\text{Ca}(\text{OH})_2$  and the  
41 formation of  $\text{AF}_{\text{M(N)}}$  instead of AFm. It is considered that  $\text{Sr}(\text{NO}_3)_2$  was incorporated in AFm phase to form  
42  $\text{AF}_{\text{M(N)}}$ . According to Park et al. [12], the nitrate anions can substitute for the sulphate anions in AFm to form  
43  $\text{AF}_{\text{M(N)}}$ . Balonis [13] also suggests that the calcium aluminohydrate phases can incorporate various anions  
44 into an anion site and can be described by the following general formulae:  $3\text{CaO} \cdot \text{Al}_2\text{O}_3 \cdot \text{CaX} \cdot n\text{H}_2\text{O}$  and  
45  $3\text{CaO} \cdot \text{Al}_2\text{O}_3 \cdot \text{CaZ}_2 \cdot n\text{H}_2\text{O}$  where  $X = \text{SO}_4^{2-}$  and  $Z = \text{NO}_3^-$ . It seems likely that Sr can substitute for the Ca in  
46  $\text{CaZ}_2$  within such a phase and this is currently being investigated.  
47  
48  
49  
50

51 The weight loss due to the  $\text{AF}_{\text{M(N)}}$  phase in CS (Fig 2a) is larger than the weight loss due to the AF<sub>m</sub> phase in  
52 B. This could be explained by the formation of a more significant amount of  $\text{AF}_{\text{M(N)}}$  in sample CS than AF<sub>m</sub> in  
53 sample B. This could also explain the absence of portlandite in the CS samples; in order to produce more  
54  $\text{AF}_{\text{M(N)}}$  more Ca would be required and therefore less is available to form the portlandite phase.  
55  
56

### 57 **4.2 Leaching Behaviour of Ca, Cs and Sr from Sample CS**

58 The more porous nature of the sample resulting from the incorporation of Cs and Sr may be one reason for  
59 the significantly higher leaching rates compared with sample B, as more surface area is in contact with the  
60  
61  
62  
63  
64  
65

leachate solution. The high leaching rate of Cs could also be due to its presence in the sample as clustered  $\text{CsNO}_3$ . The leaching of Sr might increase the leaching rate of Ca from sample CS as compared to sample B; leaching of Sr incorporated in  $\text{AF}_{\text{M(N)}}$  could result in an increase in Ca dissolution from the same phase.

It is important to note that CH was not formed in the waste loaded sample CS due to the introduction of the waste simulants, which could also influence the leaching behaviour of the material. It has been reported that when OPC is leached in water, initially CH diffuses into the leachate to replenish the  $\text{Ca}^{2+}$  ions removed from the pore solution, and only when the CH has been leached does leaching of the C-S-H begin [7]. This is due to the chemical equilibrium between the two main hydrate phases in the cement and their component ions in the pore water:



Because of the lack of CH in sample CS, leaching of CSH most likely occurred from the start of leaching.

The leaching rate of Ca significantly decreases between 7 and 28 days. Usually as leaching progresses, the the solubility limit of Ca is approached slowing down the leaching rate. However in these experiments the water was regularly replaced and thus the concentration of Ca in solution varied up and down. Thus the decreasing Ca leach rate for sample B could be because CH from the top layer has fully leached out. CH leaching from deeper within the cement bulk sample travels through longer pathways, thus decreasing the leaching rate.

A similar decrease in the leaching rate of Sr was also observed. This is in agreement with Crawford et al. [14] who stated that when  $\text{Sr}^{2+}$  present in the cement surface is completely leached out into solution, ions from deeper within the cement sample diffuse into the solution in order to maintain the equilibrium. This means the ions take longer to reach the solution and thus slowing the rate of leaching. Shrivastava et al. [15] described this as the surface wash-off process. A similar phenomenon is likely to be the reason for the decrease in Cs leach rate by around 70% after the first 7 days.

The leaching rates of Cs and Sr are compared with those of Ca in Fig 6. For congruent leaching, all the data should fall on a straight line with a gradient of 1. Fig.6 does not show this behaviour and therefore incongruent leaching is occurring, with Cs leaching more readily than Sr, which leaches more readily than Ca. This is compatible with Cs not being bonded into any of the cement phases. It may also indicate that Sr bearing  $\text{AF}_{\text{M(N)}}$  is more readily leached than other phases, however further research is needed to determine this.

## 5. CONCLUSION

$\text{CsNO}_3$  and  $\text{Sr(NO}_3)_2$  behave differently when introduced into an OPC:BFS matrix.  $\text{CsNO}_3$  is thought not to be immobilised, and reforms during the hydration reactions within the cement, thus not impacting the crystalline phases formed. As a result the diffusion rates of Cs are significantly higher than any of the other elements within the cement.

$\text{Sr(NO}_3)_2$  is not reformed during the hydration reaction and alters the phases formed within the cement. Portlandite is absent and a nitrated  $\text{AF}_m$  phase is formed. The leaching rates of Sr are significantly lower than Cs suggesting better immobilisation within the cement.

## 6. ACKNOWLEDGEMENTS

We would like to thank Dr Hashimoto of Hokkaido University, Japan for the MIP measurements.

Wail Gashier would like to thank EPSRC for sponsorship studentship enabling him to undertake this work.

## REFERENCES

- [1] M.I. Ojovan, W.E. Lee, *An Introduction to Nuclear Waste Immobilisation*, Elsevier, Oxford, 2005.
- [2] J.V. Hanna, L.P. Aldridge, E.R. Vance, Cs speciation in cements, *Mat. Res. Soc. Symp. Proc.* 663 (2001) 89–101.
- [3] K. Noshita, T. Nishi, T. Yoshida, H. Fujihara, N. Saito, S. Tanaka, Categorization of cement hydrates by radionuclide sorption mechanism, *Mat. Res. Soc. Symp. Proc.* 663 (2001) 115–121
- [4] S. Aggarwal, M.J. Angus, J. Ketchen, Sorption of Radionuclides onto Specific Mineral Phases Present in Repository Cements, Report AEA Technology NSS/R312, 2000, pp. 1–64
- [5] J. Tits, E. Wieland, C.J. Müller, C. Landesman, M.H. Bradbury, Strontium binding by calcium silicate hydrates, *Journal of Colloid and Interface Science* 300 (2006) 78–87
- [6] Wieland, Domink Kunz, Rainer Dahn, Strontium Uptake by Cementitious Materials, *Environ. Sci. Technol.* 2008, 42, 403–409
- [7] H. Saito, A. Deguchi, Leaching Tests on Difference Mortars Using Accelerated Electrochemical Method, *Cement and Concrete Research*, 30, 2000, pp. 1815 – 1825E.
- [8] M. Atkins, F.P. Glasser, Application of Portland Cement-Based Materials to Radioactive Waste Immobilisation, *Waste Management*, Vol. 12, pp. 105-131, 1992
- [9] M. Maneva, N. Petroff, Kinetics of dehydration and thermal decomposition of  $\text{Sr}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$  and its deuterated analogue with a view to obtaining SrO, *Akadémiai Kiadó*, 2005.
- [10] R.D. Spence, *Chemistry and Microstructure of Solidified Waste Forms*, Lewis Publishers, USA, 1993.
- [11] N.B. Milestone, J.P. Gorce, 'Determining how water is held in composite cement binders', *Journal of the Australian Ceramic Society* Volume 48[2], 2012, 244 – 248
- [12] Joo-Yang Park, Hye-Jung Byun, Won-Ho Choi, Wan-Hyup Kang, Cement paste column for simultaneous removal of fluoride, phosphate, and nitrate in acidic wastewater, *Chemosphere* 70 (2008) 1429–1437
- [13] M. Balonis, 'The Influence of Inorganic Chemical Accelerators and Corrosion Inhibitors on the Mineralogy of Hydrated Portland Cement Systems', University of Aberdeen, Aberdeen, June 2010.
- [14] Crawford R W, Glasser F P, Rahman A A, Angus M J and McCulloch C E 1971 Radioactive waste management and development of waste 6195
- [15] O.P. Shrivastava, R. Shrivastava, *Bull. Mater. Sci.*, Vol. 23, No. 6, December 2000, pp. 515–520. Indian Academy of Sciences

### Figure captions

**Figure 1:** XRD trace for samples of each formulation before leaching (0D = 0 Days of leaching)

**Figure 2:** (a) TGA & DTG curves for CS and B before leaching (b) TG & DTG curves for C and S before leaching



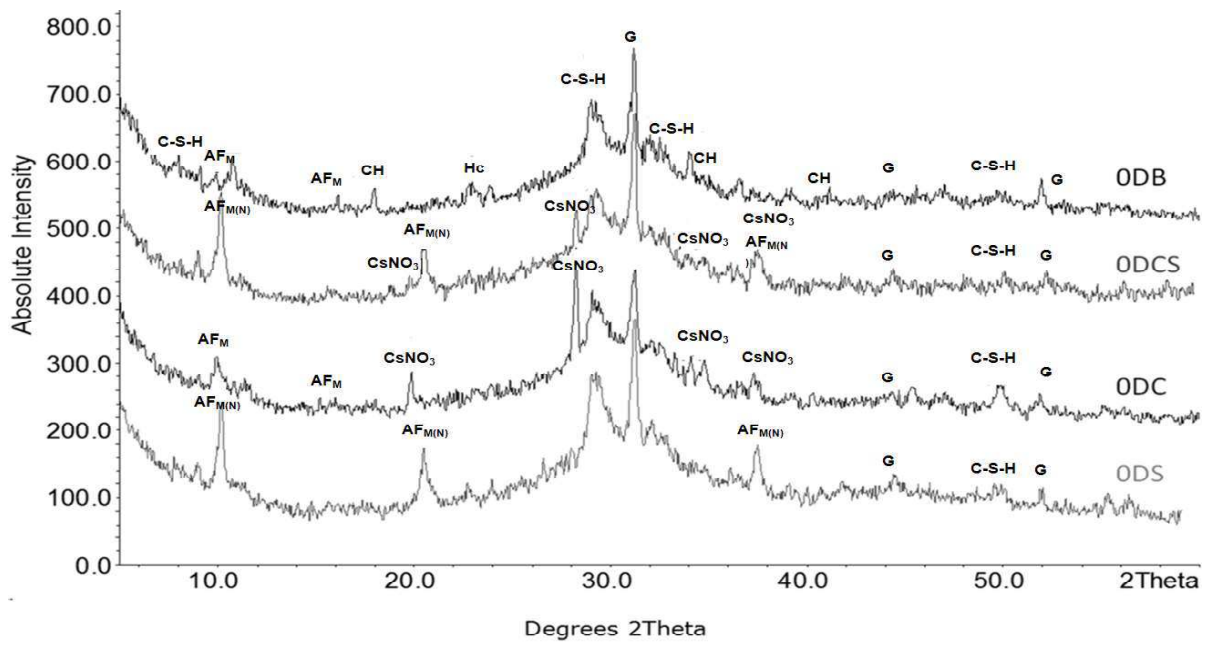
**Figure 3:** SEM results on CS sample before leaching (a) back scattered electron image with X-ray dot maps of the area highlighted by the dotted line and (b) EDS trace from a point within highlighted region

**Figure 4:** MIP results for B and CS before leaching

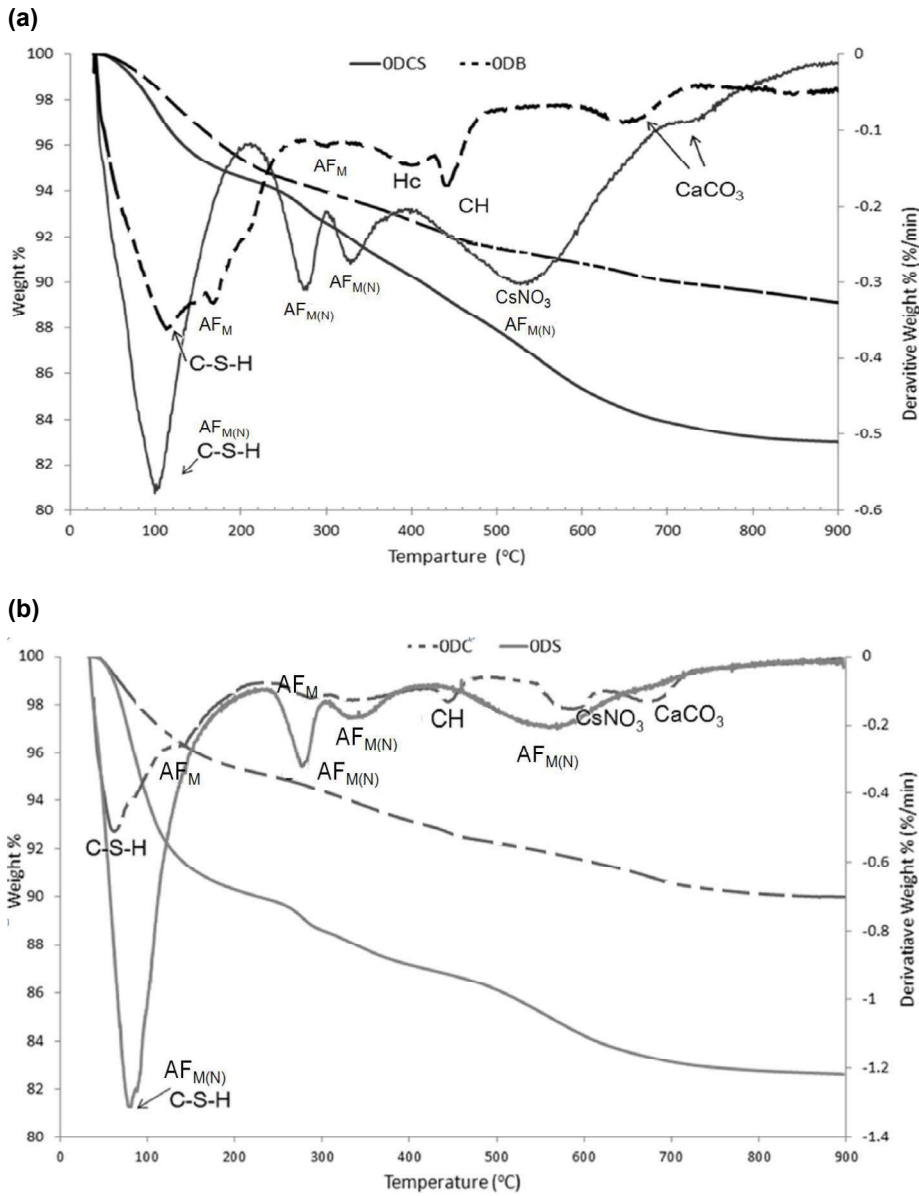
**Figure 5:** Normalised leaching rates over 6 months (a) Ca from sample B and CS (b) Cs and Sr from sample CS

**Figure 6:** Leaching rates of Cs and Sr with respect to the Ca leaching rates within the CS sample

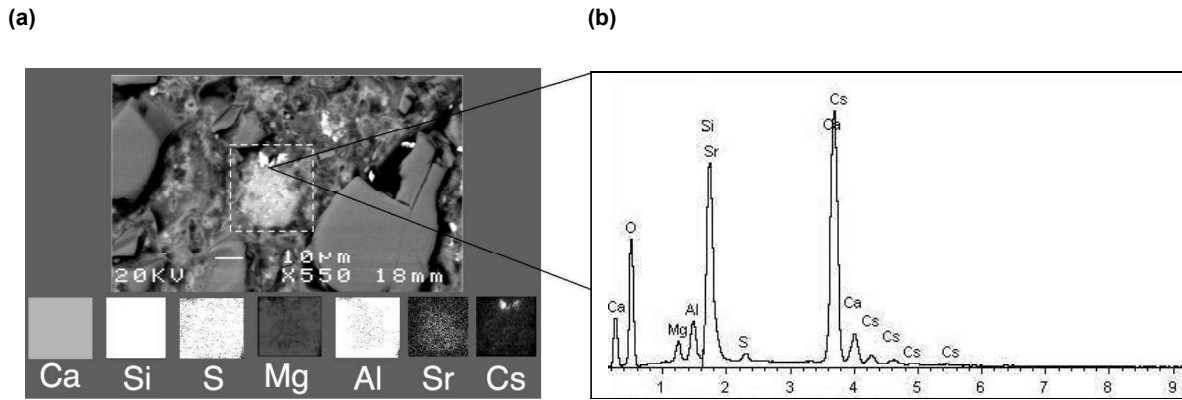
1  
2  
3  
4  
5  
6  
7  
8  
9  
10  
11  
12  
13  
14  
15  
16  
17  
18  
19  
20  
21  
22  
23  
24  
25  
26  
27  
28  
29  
30  
31  
32  
33  
34  
35  
36  
37  
38  
39  
40  
41  
42  
43  
44  
45  
46  
47  
48  
49  
50  
51  
52  
53  
54  
55  
56  
57  
58  
59  
60  
61  
62  
63  
64  
65



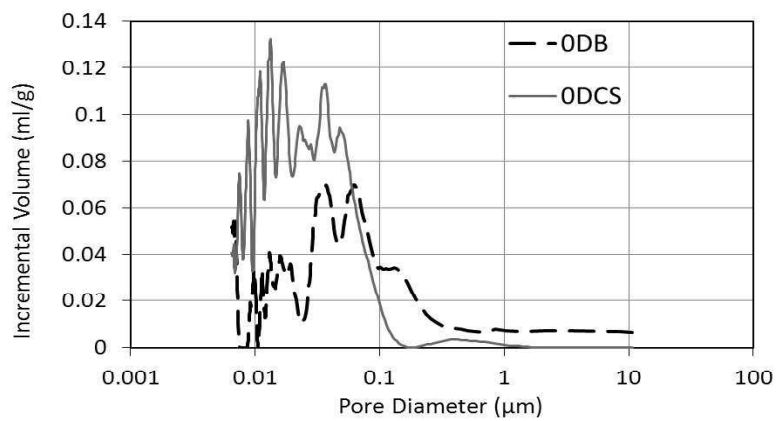
**Figure 1:** XRD trace for samples of each formulation before leaching (0D = 0 Days of leaching)



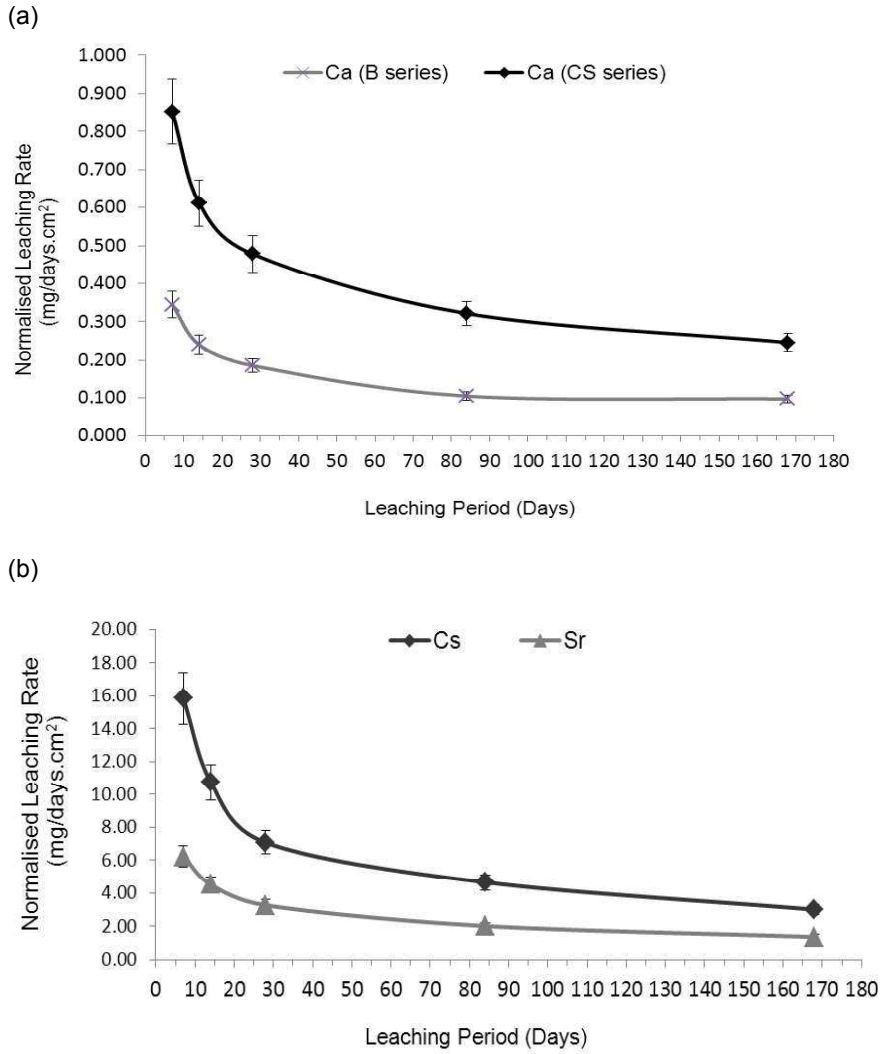
**Figure 2:** (a) TGA & DTG curves for CS and B before leaching (b) TG & DTG curves for C and S before leaching



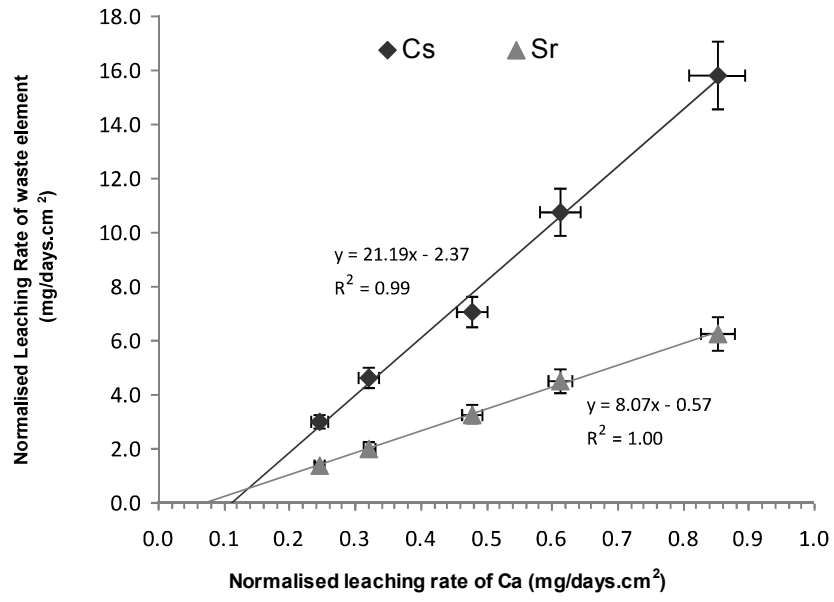
**Figure 3:** SEM results on CS sample before leaching a) back scattered electron image with X-ray dot maps of the area highlighted by the dotted line and b) EDS trace from a point within highlighted region



**Figure 4:** MIP results for B and CS before leaching



**Figure 5:** Normalised leaching rates over 6 months (a) Ca from sample B and CS (b) Cs and Sr from sample CS



**Figure 6:** Leaching rates of Cs and Sr with respect to the Ca leaching rates within the CS sample