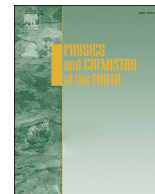




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Modelling of cementitious backfill interactions with vitrified intermediate-level waste

Graham Baston, Timothy Heath*, Fiona Hunter, Stephen Swanton

Amec Foster Wheeler, B150 Thomson Avenue, Harwell Campus, Didcot, Oxfordshire, OX11 0QB, UK

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ABSTRACT

New types of wasteform are being considered for the geological disposal of radioactive intermediate-level waste (ILW) in the UK. These include vitrified ILW products arising from the application of thermal treatment processes. For disposal of such wasteforms in a geological disposal facility, a range of concepts are under consideration, including those with a high-pH cementitious backfill (the NRVB, Nirex Reference Vault Backfill). Alternatively, a cement-based material that buffers to a less alkaline pH could be used (an LPB, Low-pH Backfill). To assess the compatibility of these potential new wasteforms with cement-based disposal concepts, it is necessary to understand their impacts on the long-term evolution of the backfill. A scoping thermodynamic modelling study was undertaken to help understand the possible effects of these wasteforms on the performance of the backfill. The model primarily considers the interactions occurring between the vitrified waste, the porewater and the backfill, within a static and (in most cases) totally closed system. The approach was simplified by assuming equilibrium between the backfill and the corroded glass available at selected times, rather than involving detailed, reactive transport modelling. The aim was to provide an understanding of whether the impacts of the vitrified wastes on backfill performance are sufficient to compromise disposal in such environments.

The calculations indicated that for NRVB, the overall alkaline buffering capacity of the backfill is not expected to be impaired by interactions with vitrified waste; rather the buffering will be to less alkaline pH values (above pH 9) but for a longer period. For the LPB, slightly lower pH values were predicted in some cases. The sorption capacities of the backfills are unlikely to be impaired by interactions with vitrified ILW. Indeed they may be increased, due to the additional C-S-H phase formation. The results of this study suggest that disposal of vitrified ILW in a cement-based disposal system with a high-pH backfill is a potentially viable disposal option.

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1. Introduction

In the UK, a framework has been set out for the long-term management of higher activity radioactive wastes through disposal in a geological disposal facility (GDF) (DECC, 2014). At present, a number of potentially suitable disposal concepts and types of geological environment are under consideration (NDA, 2010a). In the case of intermediate-level waste (ILW), potential disposal options include one in which the waste packages are placed in vaults in a higher strength rock that would be backfilled with the NRVB (Nirex Reference Vault Backfill) a high-pH

cementitious material. Alternatively, a low-pH cementitious backfill (LPB), e.g. a low-alkali cement that typically buffers to pH < 11, might be suitable. The selected backfill material will form part of an engineered barrier system (EBS) that, in turn, forms part of a multi-barrier approach to the retardation of radionuclide release and migration (NDA, 2010b).

1.1. Vitrification of ILW

Most ILW packaged to date in the UK has been encapsulated in cement-based materials, such as blast furnace slag or pulverised fuel ash composite cements (Atkins and Glasser, 1992; Ojovan and Lee, 2005). However, alternative waste treatments are currently being considered for some types of ILW. These technologies include a number of high temperature processes (sometimes described as vitrification processes) that produce vitrified wasteforms.

* Corresponding author.

E-mail addresses: graham.baston@amecfw.com (G. Baston), tim.heath@amecfw.com (T. Heath), steve.swanton@amecfw.com (S. Swanton).

Vitrification can offer potential benefits of waste volume reduction, destruction of organic material and greater physical containment than waste encapsulation in cement-based grouts. However, if such wasteforms are to be disposed of in a cementitious environment, their potential effects on the performance of a cementitious backfill need to be considered.

1.2. Backfill performance

In a cementitious ILW concept, the backfill contributes to the performance of the EBS. This study considered the effect of two illustrative vitrified ILW products on the properties of the backfills including their ability to:

- buffer the pH to alkaline conditions for a prolonged time, which inhibits the corrosion of steel waste containers and provides solubility limitation for some radionuclides such as plutonium and americium, due to the high pH, and others such as tin and uranium (VI), due to the high pH and calcium ion activity;
- provide a high sorption capacity for radionuclides released from waste containers; and
- provide a high porosity and gas permeability, to permit the migration of gases, generated by degradation of wastes and containers, out into the surrounding rock.

The ability to buffer to an alkaline pH has been interpreted here as the maintenance of the pH, above pH 9, for the period covered by radiological risk assessments (Swan and Jackson, 2007). This study aimed to provide an initial modelling evaluation of the compatibility of vitrified ILW with the NRVB using a relatively simple methodology. The approach taken was to calculate the chemical releases from a wasteform at fixed time periods, based on a dissolution rate relevant to the conditions in a GDF. Thermodynamic modelling was then used to predict the minerals formed when the components released from the wasteform equilibrate with the NRVB and the effects of the predicted mineral changes on the performance of the NRVB were then considered. For comparison a similar approach was applied to an illustrative composition for a low-pH cement, recognising that no low-pH backfill has been formulated as part of the UK geological disposal programme. The thermodynamic modelling approach assumes full chemical reaction between the backfill and the vitrified waste, for a given extent of waste dissolution. Whilst this approach may be less realistic than a detailed kinetic model, it is simpler and it maximises the potential effects of the interactions. It is therefore considered a conservative approach with respect to the impacts on backfill performance.

Table 1
Composition of clinoptilolite and Geomelt® vitrified waste stimulants.

Clinoptilolite ^a		Geomelt ^{®b}			
glass component	mass fraction	glass component	mass fraction	metal component	mass fraction
Al ₂ O ₃	9.28%	SiO ₂	43.5%	Si	8%
B ₂ O ₃	0.09%			Cr	5%
BaO	0.18%			Ni	2%
CaO	1.62%	La ₂ O ₃	7.3%		
Fe ₂ O ₃	0.81%	CeO ₂	14.0%		
K ₂ O	1.17%	Na ₂ O	6.6%		
MgO	5.57%	MgO	4.3%		
Na ₂ O	14.87%	Al ₂ O ₃	7.8%	Al	5%
SO ₃	0.07%	CaO	5.7%	Fe	77%
SiO ₂	65.99%	Fe ₂ O ₃	10.8%		
SrO	0.25%				
TiO ₂	0.11%				

^a As specified in Utton et al. (2012).

^b Mean bulk compositions of two phases produced from vitrification of a simulant high metal waste stream using the Geomelt® ICV process-see Schofield et al. (2016).

2. Materials modelled

2.1. Illustrative vitrified ILW wasteforms

For this study, the compositions of products from two full-scale vitrification trials of simulant ILW were selected to represent possible UK vitrified ILW: clinoptilolite sand glass (Deegan et al., 2007), a sodium alumina-silicate-type glass; and products from a vitrification trial of a high-metals waste stream using the Geomelt® in-container vitrification (ICV) process (Schofield et al., 2016). These waste products are shortened to 'clinoptilolite glass' and 'Geomelt® product' respectively. It is noted that the product compositions used were from demonstration trials on inactive simulant wastes and had not been optimised in terms of their wasteform performance.

2.1.1. Clinoptilolite glass

The manufacture of clinoptilolite glass waste simulant (Deegan et al., 2007) and its characterisation have been described by Utton et al. (2012). For this study, the product's composition and dissolution behaviour are the significant properties, together with the physical parameters of the associated wasteform. The composition of the clinoptilolite glass used in the calculations is shown in Table 1. The second largest oxide mass (after SiO₂) is sodium oxide, which arises predominantly from the addition of sodium hydroxide to the waste mix to aid glass formation. Therefore, dissolution of clinoptilolite glass is expected to produce significant amounts of NaOH particularly at long times. The glass dissolution rate applied in this work was determined from recent experimental studies of clinoptilolite glass in near-saturated, high-pH, calcium hydroxide solution at 50 °C (Schofield et al., 2016) and would lead to complete dissolution and reaction after 34,000 years. The dissolution rate was based on the rate of release of potassium into solution at early times, and a congruent glass dissolution rate for all other elements was assumed. At longer times the dissolution rate will typically fall (Gin et al., 2003). The rate applied in this work is therefore a maximum rate and the actual timescales for glass dissolution may be considerably longer. The dissolution rates and physical properties of clinoptilolite glass wasteform are shown in Table 2.

2.1.2. Geomelt® product

Trials have been conducted on the Geomelt® ICV process for a simulant high-metal waste stream, vitrified with a sodium silicate glass grit and a hematite additive. The outputs from these trials (Schofield et al., 2016) provided data for a typical vitrified high-metal ILW product, which were applied in this study. The process

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