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Finite element modelling approach to investigate the degradation of intermediate level waste drums induced from interior metallic corrosion

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ABSTRACT

The encapsulation of metallic waste occurring from the fuel decladding process into cementitious formulations is a typical path within the British nuclear community. The metallic–grout mixture is stored in stainless steel drums which are maintained temporarily in ambient conditions before ultimate disposal. Some of these drums have exhibited bulges ascribed to internal metallic corrosion. The corrosion products, which occupy larger volume than the parent metals, generate stresses on the surrounding material, which inhibit the long term safety of the containment. Herein, finite element modelling was used to address the effects of the internal volume expansion on the grout and the steel integrity. A parametric study is presented to determine how several factors including the type of the grout, the location and the size of encapsulated metallic parts and the friction at the material interfaces may alter the system's behaviour.

1. Introduction

The UK has accumulated over 290,000 m^3 of Intermediate Level Nuclear Waste (ILW), with the majority derived from processing of Magnox fuel and decommissioning of power stations. ILW consists primarily of millimetre thickness metallic savings of spent fuel cladding as well as small quantities of spent uranium fuel. The strategy for treating this waste within the British nuclear community is to encapsulate it in a dispersed state within cementitious materials and store it within stainless steel drums. As a result, the grout-encased radioactive material is considered immobilised for extended periods of storage and suitable for eventual disposal. In a small proportion of drums larger pieces of metal are present, the majority of which are of cylindrical shape, derived as end crops or sections of metallic fuel rods. Recently, it has been identified that an increasing number of those packages have developed some externally measurable distortion, which in turn has posed a serious question for long term safety of the confined waste. It is predicted that this bulging is induced by an internal volume expansion caused by corrosion of the encapsulated metals [1,2]. Magnesium, aluminium, steel and uranium, which are the main components of ILW, will react early in the wasteform life with the cement pore water and then later with oxygen and water vapour in air. The corrosion products in all cases are significantly more voluminous than the parent metal. This expansion causes stress generation in the surrounding encapsulant which is primarily expressed via grout cracking and potentially even steel fracture if the exerted stress is great enough. Uranium, which is of major concern due to its radioactivity, corrodes according to the mechanism proposed in the following formulae to produce uranium dioxide (UO_2) and uranium hydride (UH_3) [3], which both have approximately half the density of the parent metal [2]. The latter is of major concern for the nuclear industry as it may behave pyrophorically in air (depending on mass reacting) [4]. It is not yet proven that this hydride can form and persist in a realistic ILW drum environment. The potential for its presence though, even if very low, increases the concern over any possible drum failure.

$$U + 2H_2O \rightarrow UO_2 + 2H_2 \quad (1)$$

$$2U + 3H_2 \rightarrow 2UH_3 \quad (2)$$

The issue that has to be tackled with any problematic drums exhibiting bulges around their circumference is how to determine the corrosion state and location of the interior metallic compounds in order to understand the distribution and mass of any remaining metal. This is due to the potential for additional volume expansion that these metal masses would provide if they corrode further, exacerbating the distortion levels of the wasteform. Knowing the

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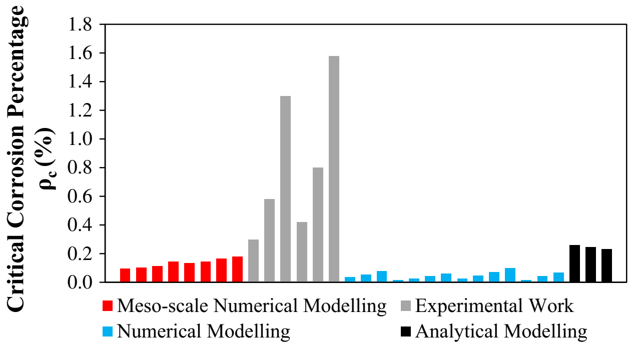

Fig. 1. Calculation of the critical corrosion percentage, i.e. corrosion percentage at surface cracking, using experimental, numerical modelling and analytical modelling. Data presented are based on relevant work found in the literature [6,12,13,15].

exact extent of corrosion could enable more accurate assessment of the possibility for containment failure. However, the potential of "seeing inside the box" has not yet been technically achieved. Consequently, predicting the extent of internal corrosion and the degree of grout and steel degradation using modelling techniques could provide an alternative route to better understanding the problem and evaluating the risk of failure. Herein, finite element modelling is employed to shed light on the implications of reactive metal corrosion, inside grouted ILW packages. A sensitivity analysis is presented to highlight the effects of different parameters on degradation of the encapsulating materials and to correlate this with the extent of metal corrosion. Since the problem described has only recently arisen, only limited confidential work has so far been performed to investigate the system's behaviour. The most relevant body of experimental work in the open literature relates to steel corrosion in concrete, which has been studied using both models and experiments [5–9]. Different systems in terms of geometry, concrete properties and modelling techniques have all been studied focusing on correlating the extent of corrosion with the cracking process. Despite the fact that the two systems have discrete differences, one originating from nuclear engineering and the other from civil engineering, a qualitative comparison can be made. One of the key factors determining the criticality of a corroding steel–concrete system is the extent of internal corrosion at the moment of the concrete's surface cracking. Several methods have been used to determine this factor including (i) analytical modelling [10–12], (ii) finite element modelling (FEM) [6,13] and (iii) experimental work [7,9,14,15]. Some of the corresponding values of corrosion percentage at concrete's surface cracking, as collected from the literature, are presented in Fig. 1. In all the cases which correspond to different systems in terms of geometry and concrete properties, surface cracking occurs at relatively low levels of steel corrosion (0.1%–1.7%). Also, there is an obvious discrepancy between the experimental and the modelling studies. This is attributed to the fact that in reality, the arising corrosion products infill all immediately accessible void and pore spaces in the surrounding concrete and do so without imposing any significant stresses. This is a phenomenon that cannot be easily simulated with modelling techniques without adding significant complexity. Considering the applicability and wide use of the modelling studies, potential issues regarding a scholastic realistic simulation of the problem as described in the previous can be neglected.

2. Finite element modelling

Extensive 2D axisymmetric finite element analyses were performed using the software package ABAQUS 6.12 to explore how the size, location and geometry of an embedded metallic body affects the degradation of a simplified ILW drum system. The geometric properties of the grout and the enveloping stainless steel were maintained constant throughout all the analyses, as their effect on the system's degradation is not part of this study. Besides, the drums used to encapsulate radioactive material in the nuclear industry are all of specific geometry (diameter, height, thickness) so there is no reason for considering different geometry. The values of the parameters that remained fixed throughout all the analyses are illustrated in Table 1. The total height of the drum, the width of the drum, the thickness of the stainless steel and the thickness of the encapsulated metallic rod are represented by H_d, W_d, T_s and T_m respectively. The corresponding figures represent a geometry which is considerably scaled down compared to the ILW drums used in the nuclear industry. The actual dimensions of an ILW drum [16] were not represented in a 1:1 scale, since the parts within the models are of significantly different geometry, tackling meshing and enhanced visualisation. For instance, the overall diameter of a typical ILW drum is 800 mm, while the corresponding thickness of the steel wall is less than 3 mm. In addition, the encapsulated material is expected to consist of metallic pieces of maximum thickness 30 mm, based on the diameter of the fuel rods where the ILW originates from. As a result of scaling down, the implemented systems were not directly comparable to the real scenario. However, it is expected that the qualitative conclusions derived from the parametric study presented in the following sections will apply for the real ILW drums, while considering how different factors control the degradation of the system. The slenderness of the models presented herein is considerably less than that of a real ILW drum and, therefore, considered to be more resistant to bulging. Small scale models could also be used as precursors for creating similar size-experimental replicates to quantitatively validate the models presented in this paper. Determining the degradation profile of the encapsulants as a result of internal metallic corrosion in a system which resembles the size of an ILW drum could be a significantly complex and time consuming process. It is expected that several years, or even decades, would be required to facilitate noticeable degradation in grout and, especially, steel in a system resembling an ILW drum corroding naturally over time. Therefore, accelerated corrosion tests are necessary to evaluate the effects of corrosion on the system's behaviour. This method has been widely applied, whilst monitoring the early stages of degradation in reinforced concrete systems [7,8]. Stitt et al. [2] produced miniaturised BFS:OPC grout samples containing uranium metal in the shape of a matchstick to monitor the early stages of corrosion behaviour using synchrotron X-ray tomography. Similar models were created and artificially corroded under hydrogen exposure on the I12 beamline, Diamond Light Source [17], to determine the corrosion behaviour from the initial stages up to total consumption of the encapsulated metal. Similar samples, where the encapsulated product would be contained in steel mini drums, could be created and subsequently corroded to determine the degradation of the encapsulants within a restricted period of time. In such a scenario, small scale systems would be necessary to comply with the experimental requirements. The

Table 1
Values of the parameters that remain constant through the numerical analyses.

Parameter	Value (mm)
H_d	100
W_d	10
T_m	1
T_s	1