Journal of Nuclear Materials 511 (2018) 396–405

Contents lists available at ScienceDirect

Journal of Nuclear Materials

journal homepage: www.elsevier.com/locate/jnucmat

Non-linear effects of alumina concentration on Product Consistency Test response of waste glasses

John D. Vienna*, Jarrod V. Crum

Pacific Northwest National Laboratory, Richland, WA, United States

ARTICLE INFO

Article history:
Received 27 June 2018
Received in revised form
17 September 2018
Accepted 19 September 2018
Available online 20 September 2018

ABSTRACT

The effect of alumina mole fraction on the 7-day Product Consistency Test (PCT) responses, $\ln[NL_\alpha]$, of simulated nuclear waste glasses was examined. It was found that the effect is highly non-linear. At low mole fractions of Al_2O_3 ($x_{Al_2O_3} \leq 0.035$) the effect of Al_2O_3 additions is highly negative ($\frac{d \ln NL_\alpha}{dx_{Al_2O_3}} = -72$). At intermediate concentrations ($0.035 < x_{Al_2O_3} \leq 0.19$) the effect of Al_2O_3 additions is moderately negative ($\frac{d \ln NL_\alpha}{dx_{Al_2O_3}} = -11$). At high concentrations ($0.19 < x_{Al_2O_3}$) the effect of Al_2O_3 additions is highly positive ($\frac{d \ln NL_\alpha}{dx_{Al_2O_3}} = +21$). This variable impact of Al_2O_3 on $\ln[NL_\alpha]$ is speculated to be caused by the rates at which the glass corrosion process changes through various reaction regimes in static conditions at 90 °C. A model for prediction of $\ln[NL_\alpha]$ as a function of glass composition is presented. This model represents the data from 2669 glass compositions spanning a broad nuclear waste glass composition region and it is useful up to much higher Al_2O_3 concentrations than previous models (from ~10 to 25 mol% Al_2O_3). The model was validated using data subset validation methods and shown to predict validation data in the same composition region with roughly equal certainty as the model fit data. The potential causes for non-linear effects of Al_2O_3 on $\ln[NL_\alpha]$ were discussed in context of the progression of glass corrosion rates. Time resolved static corrosion data is needed to better understand these non-linear effects.

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1. Introduction

1.1. Background and motivation

Approximately 210,000 m^3 of high-level radioactive waste (HLW) is stored in underground tanks at the Hanford site near Richland WA. This waste resulted from the processing of irradiated nuclear fuels and targets for the separations of heavy elements by a number of processes [1]. The Hanford Tank Waste Treatment and Immobilization Plant (WTP) is being designed and built to manage these tank wastes. The HLW will be retrieved, separated into low-activity waste (LAW) and HLW fractions, and vitrified to form alkali-alumino-borosilicate glass waste forms for disposal. The resulting LAW glass will be disposed on the Hanford site and the HLW glass will be stored temporarily and then ultimately disposed in a federal HLW repository. The waste acceptance criteria for disposal of both HLW and LAW include a limit on the response to the Product Consistency Test (PCT) Method A – a static dissolution test performed for 7 d at 90 °C in deionized water with a targeted glass surface area to solution volume ratio of 2000 m^{-1} [2]. For HLW glass, the normalized loss (NL_α) for α = B, Na, and Li must be below those for the Defense Waste Processing Facility Environmental Assessment Glass ($NL_B = 8.35$ g m^{-2}, $NL_{Na} = 6.67$, and $NL_{Li} = 4.78$; respectively) [3–5]. For LAW glass, the NL_B, NL_{Na}, and NL_{Si} must each be below 2 g m^{-2} [6]. These PCT constraints are met using models that predict the NL_α of glass to be produced based on the glass composition. Current Hanford models predict the PCT responses of glasses planned for the early stage of WTP operation (called "baseline" Hanford glasses) [7,8]. These models correlate the natural logarithm (ln) of NL_α with linear combinations of glass components plus a limited number of second-order polynomial terms:

$$\ln[NL_\alpha] = \sum_{i=1}^{q} a_{\alpha,i} x_i + selected \left\{ \sum_{i=1}^{q} a_{\alpha,ii} x_i^2 + \sum_{i=1}^{q-1} \sum_{j=i+1}^{q} a_{\alpha,ij} x_i x_j \right\} \quad (1)$$

* Corresponding author.
E-mail address: john.vienna@pnnl.gov (J.D. Vienna).

https://doi.org/10.1016/j.jnucmat.2018.09.040
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where, $a_{\alpha,i}$, $a_{\alpha,ii}$, and $a_{\alpha,ij}$ are the coefficients for component i, component i squared, and component i crossed with component j, respectively; and x_i and x_j are the ith and jth component concentration in glass. The concentrations can be either mass fraction or mole fraction; in the case of the baseline models concentrations are in mass fractions, while models developed for this study use concentrations in mole fractions.

The breadth of the expected Hanford glass compositions (called "advanced" Hanford glasses) is not covered by the current baseline models. The HLW baseline glasses were designed to immobilize the first few Hanford tanks that are relatively high in either iron or zirconium and thorium and low in most other waste components. These HLW baseline glasses were also designed for relatively low waste loading as their purpose was to start up the new facility. By contrast, the advanced glasses are under development to immobilize the full range of tank wastes with very broad composition ranges and focused on high waste loadings to improve mission efficiency [9–14]. Of particular interest is the difference in concentration ranges for Al_2O_3, which was restricted to 13 mass% (~8.89 mole%) in baseline HLW glasses and projected to be up to 30 mass% (22.2 mole%) in advanced Hanford HLW glasses. Higher Al_2O_3 wastes include neutralized cladding removal waste and REDOX solvent extraction wastes that were not included in the baseline waste tanks.

It has previously been shown that Al_2O_3 has a non-linear effect on ln[NL_α] [15–18]. Fig. 1 shows the effect of changing Al_2O_3 concentration on PCT response. For each of the studies shown Al_2O_3 concentration is increased while maintaining the ratios of other glass components constant. As alumina content increases, the ln[NL_α] decreases dramatically until a concentration point near 3.5 mole% (roughly 5 mass%), after which Al_2O_3 additions only modestly decrease ln[NL_α]. It is interesting to note that the EM-07 glasses with Al_2O_3 concentrations ranging from 4 to 14 mole% show slightly "U-shaped" behavior [19]. Two hypotheses were offered to explain this non-linear effect. Feng et al. suggested that small additions of Al_2O_3 increase the total fraction of glass formers until a threshold is achieved above which significant effects are not realized [18]. Vienna et al. suggested that small additions strongly reduced non-bridging oxygen concentrations, improving short-term durability, while larger additions had a lesser effect [20]. Neither of these speculations were supported by data or further developed.

Vienna et al. developed PCT response models with a broader database of glasses that extended many of the component concentration ranges [21]. Notably, the maximum Al_2O_3 concentration was increased to 20 mass% (compared to the ≤13 mass% in the baseline WTP models). This model includes non-linear terms for Al_2O_3 concentration. This model was applied to advanced glasses, including those with relatively high Al_2O_3 concentrations. Fig. 2 shows the residual ln[NL_B] (measured minus predicted ln[NL_B]) for advanced glasses using the Vienna 2009 model [21]. The model under-predicts ln[NL_B] progressively more as the concentration of Al_2O_3 increases. This suggests that as Al_2O_3 concentration increases above roughly 19 mass% increases PCT responses.

1.2. Previous studies evaluating Al_2O_3 impacts on glass durability

The impacts of Al_2O_3 concentration on other glass durability related measurements have been reported with the results summarized here [23–29].

- Kim et al. [17] reported the impact of Al_2O_3 concentration on the Materials Characterization Center Test 1 (MCC-1); a test performed at 90 °C and 10 m^{-1}, in deionized water for 28 d. The results showed a "U-shaped" response for glasses with 6 or 9 mass% B_2O_3 and a monotonically negative response for soda-lime silicate glasses or sodium-alumino-borosilicate glasses with 12 mass% B_2O_3. They attribute the negative effect to removal of non-bridging oxygen (NBO) concentrations caused by the alkali ion charge compensating added Al_2O_3 in the four-coordinated state ($^{[4]}Al$). The increase in MCC-1 responses at Al_2O_3 concentrations above 12 mass% in glass was not understood, nor was the reason that it occurred only for the glasses

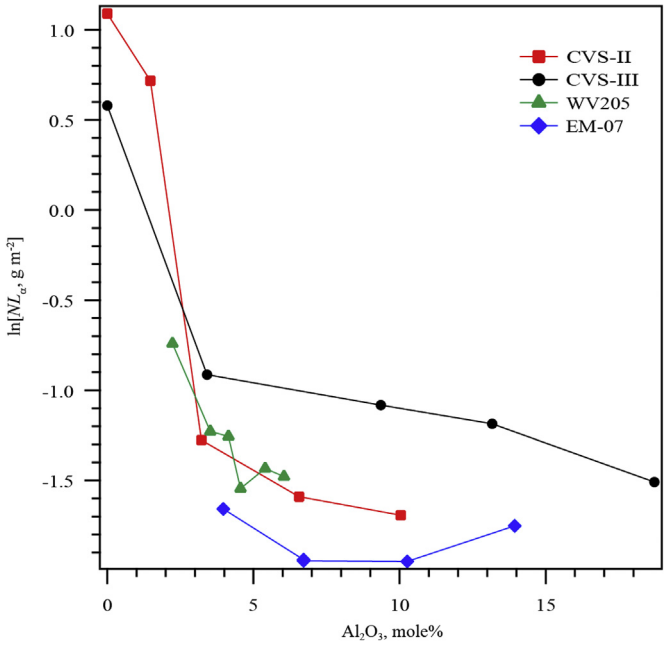

Fig. 1. Effect of Al_2O_3 mole% change on ln[NL_α, g/m^2] based on data from CVS-II study [α = B] [15], CVS-III study [α = Na] [16], WV205 study [α = B] [18], and EM-07 study [α = B] [19].

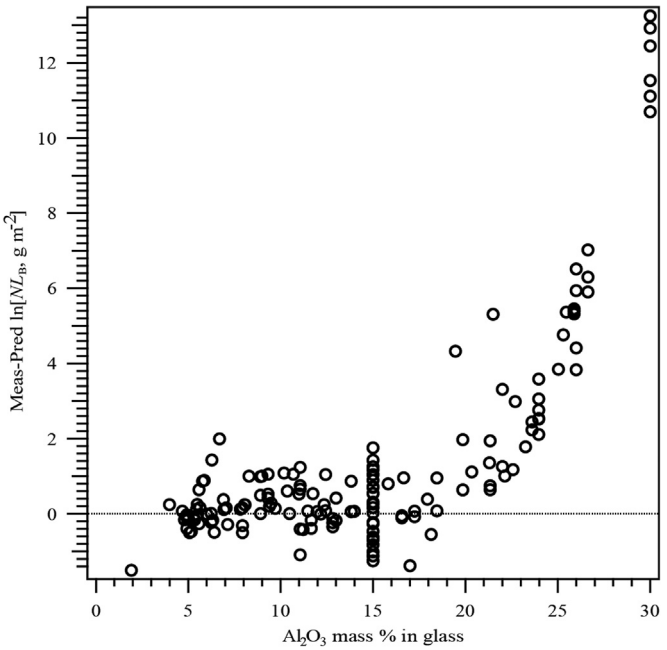

Fig. 2. Impact of Al_2O_3 concentration on PCT prediction residuals (after Vienna et al. [22]).

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