The Effect of Storage in an Inert Atmosphere on the Release of Inorganic Constituents during Intermittent Wetting of a Cement-based Material

A.C. Garrabrants¹, F. Sanchez¹, C. Gervais², P. Moszkowicz², D.S. Kosson¹

¹ Vanderbilt University, Nashville, TN USA
² LAEPSI, INSA of Lyon, Villeurbanne, France

Abstract

Monolithic waste materials (e.g. Portland cement treated wastes) in many field scenarios do not remain continuously saturated, but experience intermittent wetting interspersed with periods of storage in an unsaturated environment. During storage, the matrix may loss moisture to the environment, promoting precipitation or redistribution of species. In addition, the matrix may react with the surrounding atmosphere through carbonation or oxidation. Upon subsequent leaching, changes in the chemical and physical composition incurred over the storage interval can influence the release of inorganic species. Current assessment approaches, which use continuous leaching data to project release over some assessment interval, do not allow for changes in leachability resulting from intermittent wetting and storage. Thus, this study evaluates the effect of storage intervals in an inert atmosphere on subsequent release of inorganic species from a synthetic Portland cement matrix. Tank leaching in deionized water was interspersed with storage at three relative humidity (RH) levels (nominally 0, 50 and 100% RH) in a 100% nitrogen atmosphere. Leaching data from the three intermittent wetting cases were compared to continuous leaching for the release of structural species (Ca, OH), highly soluble species (Na, K, Cl) and pH-dependent species (As, Cd, Pb).

The RH of storage environment, which acted as a boundary condition for the drying process, influenced the precipitation of species within dried pores and relaxation of pH and concentration gradients within water-filled regions. Gradient relaxation resulted from continued mass transport within saturated pores over the storage interval and was most evident when storage was conducted at 98% RH. However, when storage RH promoted drying of the matrix, the effect of gradient relaxation was balanced by precipitation. When release was normalized to total leaching time, relaxation of concentration gradients of highly soluble species resulted in greater cumulative release for the intermittently wetted cases than in the case of continuous leaching. The release of pH-dependent constituents was controlled by relaxation of the pH gradient and species solubility as a function of local pore water pH. Application of a current assessment protocol to estimate intermittent wetting release resulted in either over or underestimation of actual cumulative release, depending on the nature of the constituent of interest. These results imply that long-term constituent release from Portland cement-based waste forms should not be made by simple correction of saturated release assessments because alterations to the matrix leachability induced by the storage environment need to be considered. © 2002 Elsevier Science B.V. All rights reserved.