

Carbonation of low- and high-pH, OPC and Alite based cementitious systems

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Cementitious systems are commonly characterized by high pH (12-13). Upon exposure to the atmospheric CO₂ or carbonate-rich porewater, carbonation takes place, resulting in the formation of new phases and changes in the micro-structure. In the recent years, low-pH matrices are developed to mitigate the effect of perturbations caused by the pH gradient between the cement and the environment. Additionally, cementitious matrices are used for solidification and stabilization of low- and intermediate-radioactive waste streams. The exposure of such matrices to carbonation degradation may affect their retention capability towards radionuclides (e.g. ⁹⁰Sr²⁺).

In this work, the effect of laboratory enhanced carbonation on high- and low-pH Portland cement matrices is studied compared with parallel alite-based model systems. The matrices were characterized for their phase composition (XRD, TGA), micro-structure (SEM, optical microscopy) and Sr retention capability before and after carbonation. The leaching behavior of the matrices was studied using EPA 1313 and 1315 methods. Moreover, the effect of leaching on cementitious carbonated monoliths was studied and characterized.

The carbonation of low pH systems is characterized by the formation of calcite and vaterite, while the carbonation of the high-pH systems yielded only calcite. In the low-pH systems, produced with high silica content, evidence for the formation of crystalline silica phases due to carbonation was found. In the high-pH systems, enhanced carbonation caused the formation of an impermeable crust at the rim of the samples, that reduced further carbonation and provided an effective leaching barrier, thus, increasing Sr retention. In the low-pH systems, the carbonation front migrated readily into the material, and had only minor effect on the Sr retention of the matrix.

To conclude, with respect to Sr retention, the advantages acquired using low-pH matrices for radioactive waste stream solidification, were found to be mitigated by carbonation.