Abstract

We investigate the interaction between Ca rich cementitious materials and bicarbonate rich pore water with the primary focus on pore size effect on the dissolution of portlandite and calcium silicate hydrates (C-S-H), and the precipitation of calcite, in a coupled reactive transport modelling framework. The concept of pore-size dependent solubility is extended to reactive transport modelling within cementitious materials. For the two pore sizes 1e-6 and 1e-8 m considered here the effect of pore size on the solubility of portlandite, C-S-H and calcite is different, with portlandite and calcite less affected and C-S-H more affected due to a higher water/ C-S-H interfacial tension. This difference leads to a different evolution of porosity at the interface of a cementitious material and bicarbonate-enriched pore water between systems with large or small pores. In systems with mixed large and small pores, if diffusion through large pores is much faster than through small pores, precipitation will only occur in large pores because the pore solution diffused into small pores has already become under-saturated with respect to calcite due to precipitation in large pores first. These results have important implications for carbonation of cement, recovery of hydrocarbon and geothermal energy and geological storage of carbon dioxide.