Resumen de Tesis Doctoral

UNIVERSITAT POLITÈCNICA DE CATALUNYA
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Título de la tesis	Understanding the mobility of caesium, nickel and selenium released from waste disposal: Chemical retention mechanisms of degraded cement
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(Mínimo 1 y màximo 4, podéis verlos en http://doctorat.upc.edu/gestion-academica/carpeta-impresos/tesis-matricula-y-deposito/codigos-unesco)

Resumen de la tesis de 4000 caracteres máximo (si se superan los 4000 se cortará automáticamente)

Cementitious materials are used to condition or stabilise waste and to build infrastructure in disposal sites. Moreover, they are envisaged to form part of engineered barrier systems as container, backfill or liner materials in radioactive waste disposal concepts. In the event of contact with water, contaminants dissolve and their mobility is influenced by the employed cementitious materials. Therefore, sound understanding of the interactions between contaminants and degrading cementitious materials in flowing water is essential for safety assessment.

The aim of this study was to identify the processes affecting retention of caesium, nickel and selenium on Hardened Cement Paste (HCP) during its degradation, from sane to severely degraded states. The focus was put on the underlying mechanisms and possible remobilisation of previously retained contaminants due to the changing composition of the HCP. Caesium, as Cs(I), nickel, as Ni(II) and selenium, as Se(VI), were chosen because they are considered as safety relevant radionuclides for nuclear waste disposal, represent different chemical characteristics and their stable isotopes can be used in experiments. To address shortcomings of previous studies in this field a combined approach was developed. First, a previously used thin-layer flow-through reactor was adapted and improved for the needs of studying contaminant retention and release during degradation of the multiphase material HCP. Second, retention and degradation were studied in equilibrated batch systems as well.

Regarding degradation of HCP the following results were obtained:

(1) The thin-layer flow-through setup was established for degradation of HCP at far-from-equilibrium conditions and a number of relevant experimental data were obtained. (2) A kinetic degradation model satisfactorily reproduced experimental results on HCP degradation. For this, a set of dissolution rate constants of cement phases was optimised which can also be used for other modelling studies. (3) The same model also satisfactorily reproduces results from experiments with different aqueous CO2 concentrations and with different solution types, i.e. synthesised granitic groundwater (GG water) and deionised (DI) water. (4) When quantitatively comparing different solution types, degradation of HCP equilibrated with GG water is stronger than after equilibration with DI water, due to higher aqueous CO2 concentration. Further, the effects of carbonate buffering and carbonation on HCP at far-from-equilibrium conditions were identified and quantified. (5) Four characteristic stages of HCP degradation in flow-through conditions were classified, taking into account carbonate buffering effects. The different stages can be discerned on-line by measurement of pH, Ca and Si concentrations in outflow solutions.

Regarding retention of Cs, Ni and Se(VI) in HCP conditioned systems the following results were obtained:

(1) Caesium and selenate distribution coefficients were determined in equilibrated systems at different degradation states of HCP. (2) Caesium and selenate retention was quantified at flow conditions during continuous degradation of HCP in DI and GG water and likely retention mechanisms were narrowed down.(3) In the case of nickel, the solubility limiting phases formed in presence of HCP were identified at different degradation states. (4) The formation of a so far non-described nickel-silicate-hydrate was observed in the more degraded system at pH around 11.6. (5) The influence of different aqueous CO2 concentrations on Cs, Ni and Se(VI) retention was demonstrated to be minor.

This study showed that the persistency of contaminant retention by adsorption in degrading cementitious systems is not only a question of distribution coefficients at different degradation states, but also a question of how fast these degradation states are reached.

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