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Contribution of electrochemical impedance spectroscopy to the study of aluminium-magnesium alloys corrosion in magnesium potassium phosphate cement pore solution

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Introduction

Reprocessing of spent fuel designed for natural uranium – graphite – gas nuclear reactors has produced some radioactive wastes containing aluminium-magnesium alloys, which need to be stabilized and solidified before their final disposal. Portland cement (PC) is extensively used for the conditioning of low- or intermediate-level radioactive wastes; however, its high alkalinity is a serious obstacle to aluminium stabilization, this metal being oxidized by the pore solution, with production of dihydrogen [1]. Magnesium potassium phosphate cement (MKPC)-based materials, with a pore solution pH between 8 and 9 provided that the Mg/P molar ratio of the cement is kept close to 1, may show better compatibility with this kind of waste [2]. This work thus aims at investigating the electrochemical behavior of Al-Mg alloys in synthetic solutions representative of MKPC pore solutions using electrochemical impedance spectroscopy. In addition, a panel of analytical techniques (ICP-AES, SEM-EDS, XRD, TGA, XPS, gas chromatography) is used to characterize the corrosion products as a function of the pore solution composition and grade of alloy.

Study

The corrosion behavior of several aluminum-magnesium alloys (Mg content varying from 0 to 5%wt) is investigated in synthetic solutions mimicking the pore solution of:

- a conventional Portland cement paste,
- a MKPC paste after 28 days of curing under endogenous conditions,
- a MKPC paste having reached thermodynamic equilibrium with respect to K-struvite ($\text{MgKPO}_4 \cdot 6\text{H}_2\text{O}$), the main cement hydrate formed in this material.

Polished metal pieces are immersed in the different solutions and corrosion is followed by quantifying dihydrogen released in the gas phase, Al and Mg species dissolved in solution and metal weight loss. Electrochemical impedance spectroscopy (EIS) is also performed at open circuit potential (OCP) using a specific cell comprising 4 electrodes (1 in Al-Mg alloy and 3 in Pt) immersed in solution. An electrochemical mechanism in 4 stages is postulated for the corrosion process, taking into account charge transfer, water and Al^{3+} diffusion as well as passivation by precipitation of Al_2O_3 . An equivalent electrical circuit is deduced from Fick's law and Butler-Volmer's equations, and the electrical parameters are adjusted by fitting experimental impedance spectra. The kinetic parameters linked to the corrosion mechanism are then calculated from the electrical parameters and used to assess the corrosion rate. This latter is finally compared to the corrosion rate inferred from dihydrogen release. Characterization of corrosion products or precipitates is also performed.

Conclusion

The results confirm that:

- the Al-Mg alloys are fully corroded within a few days in the solution mimicking the pore solution of a Portland cement paste, regardless of their Mg content,
- corrosion is much more limited in MKPC pore solution; nevertheless, the corrosion rate can vary significantly depending on the solution composition,
- in contrast, the Mg content in the Al alloy has limited influence on the corrosion rate in MKPC pore solution, whatever its composition,
- the corrosion rate values inferred from gas chromatography analysis, solution analysis and EIS at OCP are consistent, thus supporting the postulated corrosion mechanism.

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