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Electrochemical impedance spectroscopy: a non-destructive method to study the corrosion of aluminum-magnesium alloys in a magnesium phosphate cement-based matrix

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Introduction

The reprocessing of spent fuel designed for natural uranium – graphite – gas 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 aluminum stabilization, this metal being oxidized by the pore solution, with production of dihydrogen. Its passivation due to the formation of a protective layer of alumina only occurs for pH values within the range 3-9. Magnesium phosphate cement (MPC)-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 . This work aims at investigating the corrosion of Al-Mg alloys in MPC mortars using electrochemical impedance spectroscopy to monitor the corrosion kinetic.

Study

The corrosion behavior of several aluminum-magnesium alloys (Mg content varying from 0 to 5%wt) in a MPC matrix is investigated using electrochemical impedance spectroscopy (EIS) at open circuit potential (OCP). A specific cell is designed, comprising 4 electrodes (1 in Al-Mg alloy and 3 in Pt) embedded in the cement matrix, and making it possible to get information both on the corrosion of the metal and on the evolution of the matrix over time. The impedance of the system is measured at OCP by applying a small sinusoidal potential disturbance (amplitude 10mV, frequency range $0.1 - 10^7 \mathrm{Hz}$). An electrochemical mechanism in 4 stages is proposed for the corrosion process, taking into account water diffusion, Al^{3+} diffusion and passivation by precipitation of Al_2O_3. An equivalent electrical circuit is deduced from Fick's law and Butler-Volmer's equations, and is then used to model the experimental impedance values. It allows to determine the kinetic parameters linked to the corrosion mechanism. The corrosion rate is finally calculated using these parameters and compared to the corrosion rate inferred from dihydrogen production, measured by gas chromatography. The different alloys are compared between each other depending on their Mg content.

EIS is also used to study the corrosion under irradiation of the same system. Cells with electrodes and filled with MPC mortar are gamma-irradiated from the fresh stage to a dose of radiation of 1MGy, at a dose rate of ~1kGy/hour. Impedance spectroscopy measurements are performed during the irradiation; the results are then analyzed to determine the evolution of the corrosion parameters, but also of the matrix properties.

Conclusion

The results confirm that:

- EIS at OCP is a non-destructive technique well adapted to monitor the corrosion of metals in a cementitious material,
- the Mg content in the Al alloy has limited influence on its corrosion rate in the MPC matrix,
- irradiation seems to have a limited effect on Al alloy corrosion.