TOPIC 11: Monitoring (from initial state to post-closure and from sensors to data management)

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Abstract

Near-neutral pH and low redox potential (Eh) are considered to be favourable conditions for nuclear waste disposal (NWD) in clay formations, because most radionuclides have a low solubility under such conditions. NWD programs mainly focus on deep geological storage, as this is the most appropriate strategy for ensuring the long-term safety of people and environment. Cigeo is the future deep NWD facility for high-level and intermediate-level long-lived radioactive waste, to be built in France, at 500 m depth within the clayey Callovo-Oxfordian formation (COx). COx is a 130 m thick clay-rich rock, lying at a depth of 400 to 600 m, which is water-saturated with extremely low permeability, porosity and hydraulic conductivity. COx pore-water temperature, pH and pCO_2 are constant at 21-22 °C, 7.2 (±0.2) and 8.10^{-3} atm, respectively. Anoxic conditions prevail in the COx, thus pH and Eh are key parameters for monitoring the evolution of its pore water.

Our objective was to choose the appropriate material for electrodes and to design, create and optimize robust multi-parameter probes (MPPs) for reliable on-site monitoring of pH and Eh. Thus, several electrodes made of different sensitive solid materials were studied. Their Open Circuit Potential (OCP) measurements under various conditions were examined in terms of reliability and robustness. We built up innovative MPP devices for such long-term monitoring, placed at 490 m depth into a gallery of the Underground Research Laboratory of Andra at Bure. The experimental set-up assembles 2 MPPs, connected in series, which receive the seepage water extracted from the borehole EPT1201. Preserved from air contact, the water feeds up the 2 MPPs (both down fed to avoid two-phase flow) during several months, with a flowrate of 1-2 mL/h. Each MPP contains several electrodes: 2 monocrystalline antimony Sb_2O_3/Sb, for pH sensing to be compared with 2 conventional pH electrodes (CpHE); 4 AgCl reference (only Cl^- sensitive); 4 Ag_2S reference (only S^{2-} selective); 4 Pt; 2 Au, for Eh measurements. Two series of pH and Eh measurements were carried out, which both lasted several months, and permit

the comparison of OCP of all the solid electrodes versus reference electrodes. The difference between the 2 series resides in the progressive appearance or not of sulfide in the water passed through the MPPs. Sulfide production originates from sufate-reductive bacteria which use some of the organic content of CpHE. The results are summerized in figure 1. Overall, the conceived bundle of electrodes worked reliably during a timescale that is promising for monitoring the evolution of the COx pore water during a certain operational period of Cigeo.

Fig 1: A) OCP of 4 AgCl and 1 Ag_2S electrodes vs time during exposition in situ, with progressive sulfidization of water. B) pH vs time from CpHE and from Sb_2O_3 electrode using calibration curves obtained in GB conditions (absence and presence of O_2). C) Calibration curves (OCP of Sb_2O_3 electrode vs pH from CpHE obtained in GB conditions using various sulfide contents. These curves enable us to deduce the pH *in situ* in the presence of sulfide. D) pH vs time from CpHE and from Sb_2O_3 electrode using calibration curves obtained in GB conditions (absence of O_2 and presence of sulfide) and simultaneous OCP monitoring of Ag_2S electrode vs time.

Figure 1 (A, B, C, D)