ARGAZ: A NEW DEVICE FOR EXPERIMENTAL STUDY OF THE COUPLING BETWEEN HYDROGEN PRODUCTION AND HYDROGEN TRANSFER THROUGH SATURATED CALLOVIAN-OXFORDIAN ARGILLITE

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A specific experimental device has been designed to produce hydrogen at the metal-argillite interface by electrochemistry. The target is for one hand to reproduce the production of hydrogen occurring when a metal is corroded by the water contained in the porosity of the mudstone. On the other hand, the transfer of the hydrogen through the mudstone can be studied.

The specific features of the experiment are the following:

- Hydrogen is generated inside a cell by electrochemistry, at the interface between the argillite and a metallic surface; no gas injection is required;
- Electrochemistry gives the possibility to control the hydrogen production rate;
- Hydrogen generation implies water consumption: the water comes from the porosity of the bulk argillite, near the interface;
- That one-dimensional experiment has been built around a cylindrical sample of bulk and undamaged argillite coming from the Callovian-Oxfordian formation.

Inside the device a cylindrical sample of argillite is placed above a nickel plate. Around the argillite, a ring of compacted bentonite ensures a mechanical confinement. When saturated, the bentonite will apply a swelling pressure close to the total pressure encountered by the sample in the geological formation. The hydrogen is generated at the interface nickel-argilite. The nickel plate is one of the two electrodes required for electrochemistry. At the top face of bentonite, iron electrode is used to close the electrical circuit. The hydrogen produced at the bottom face of the mudstone is expected to go across the argillite towards the top face. A porous plate connected with a sampling bottle allows the capture of hydrogen. The argillite sample has a diameter of 50 mm, and a height of 50 mm. It is obtained by over-coring a core sample, and by a careful machining leading to a perfect geometry and surface quality.

The production rate of hydrogen can be calculated from the current intensity by the Faraday law. A global measurement will be achieved by analysing at regular intervals the content of the sampling bottle. A miniature pressure sensor placed at the nickel/argillite interface is expected to provide the overpressure due to hydrogen production.

The device is equipped with miniature pressure sensors. Three of these sensors are placed inside the argillite at different levels from the nickel-argillite interface, namely at 12.5 mm, 25 mm and 37.5 mm. These sensors aim to measure the pressure of the fluid (liquid plus gas pressures) in the porosity of the argillite. Thanks to their little size, these sensors do not disturb a lot the interstitial pressure in the mudstone. As the media is initially saturated, they will indicate the possible over pressure due to hydrogen at the sensing point.

The device is also equipped with nickel wires. Three pairs of wires are placed inside the argillite at different levels from the nickel-argillite interface. These sensors are located at the same levels as the pressure sensors. These electrodes will be used first to monitor the potential redox front due to hydrogen transport in aqueous phase and secondly to monitor the gas phase transport if it takes place. This monitoring is based on high frequency impedance measurements allowing determining the seeming ionic resistance of the part of argillite located in between the nickel wire. It has been shown that bubbles transport modified highly this seeming ionic resistance.

The first phase of the study will consider a moderate production rate of hydrogen. An equivalent corrosion rate, about 1 to 3 μm/year will be considered. During this phase hydrogen will probably dissolve in the water contained in the porosity of the mudstone.

After results analysis, other production ratings will be tested. Particularly, the possible de-saturation of the argillite near the interface, due to the water consumption and gas generation in competition with gas transfer and dissolution, will be studied. The local overpressure near the interface will be monitored.