Dissolution of Organic Matter in Siltstone by Supercritical CO2

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The underground storage of CO_2 is an effective technology for reduction of CO_2 emissions into the atmosphere. To store CO_2 for a long time, the integrity of cap rock is one of the most important factors. Shale or mudstone formations are expected as the cap rock. These rocks, however, contain organic matter, and supercritical CO_2 has the extractive capability of organic substances. Underground condition at 1000 m depth is typically 50C and 10 MPa, whereas the injected CO_2 becomes supercritical fluid. This state of CO_2 could affect the sealing performance of cap rock. Recently, various studies on supercritical CO_2 extraction of organic matter from oil shales have been reported (McKay and Chong, 1983, Jaffe et al., 1997, Koel and Ljovin, 2000). These researches have been conducted mainly for pursuing high recovery rates of oil etc., and so the research targeting the imperfection of cap rock in CO_2 underground disposal were not reported. In this study, the effect of supercritical CO_2 on sealing performance of cap rocks was examined as a first step by the porosity measurement after supercritical CO_2 treatment on siltstone samples.

To measure the content of organic matter in the sample, the pulverized siltstone sample was set in a batch type autoclave and was treated with supercritical CO_2 at the pressure of 10 MPa and the temperature of 50, 100 and 150C. The treatment periods were 10, 20 and 30 days. After the treatment, the amount of organic matter in powder samples were measured by an organic CN analyzer and compared to the contents in the non-treated sample. The contents of carbon and nitrogen do not seem to be changed remarkably by the treatment. Slight change of C and N contents might be ascribed to the light hydrocarbon extraction. It should be noted that even if CO_2 would extract all organics, only a few thousandth of the original weight would be lost. However, in case that the structure of pore drastically changed, leakage through cap rock could be resulted.

The siltstone sample was cut in the size of about 1 cm^3. The block samples were set in the high-pressure vessel, and samples were treated with supercritical CO_2 at the same condition as powder experiment for 10, 20 and 30 days. The porosity and the pore radius distribution were measured by mercury porosimetry to estimate the permeability affected by supercritical CO_2. The average porosity after the treatment at 50C increased about 1%. All of the samples have had bimodal pore radius distribution. The pore radius distribution of the sample after treatment experiment has slightly shifted to larger over the whole pore size. If this shift were caused by supercritical extraction, and this phenomena is generalized, storage CO_2 would affect on sealing performance of cap rock to some extent. From the measured pore radius distributions we can estimate some physical properties such as permeability (Lin et al., 1999). The calculated permeability seems to increase slightly in the case of treatment at 50C within 10 days. Extraction process might be very fast.

The effect of supercritical CO_2 on sealing performance of cap rock does not seem to be remarkable. However, permeability does not only depend on porosity. If organic matter that bonds grains like a paste was dissolved, the permeability of the rock would increase. Measurement of permeability is necessary for estimating the sealing performance. Also the mechanical properties should be measured in order to examine comprehensively.