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Predicting the interactions of H_2S-CO_2mixtures with aquifer rock, based on experiments and geochemical modeling

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Abstract

Greenhouse effect prevention, oil and gas stimulation, and other recent technologies require a recognition of the acid gas interactions with geologic formations. The experimental study coupled with hydrogeochemical modeling was focused on the impact of CO_2, H_2S and their mixtures on the formations of the Upper Silesian Coal Basin. Significant changes in structure and composition of rock samples influenced by acid gas were identified in autoclave experiments. Skeletal grains dissolution was most intense in carbonates and chlorite, and caused an increase of porosity. In the case of geological storage of H_2S, a release of significant amounts of CO_2, from dissolution of primary carbonates should be expected, as demonstrated the results of modeling.

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Keywords: acid gas sequestration, carbon dioxide, hydrogen sulfide, hydrogeochemical modeling

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1. Introduction

Interactions of acid gases with rocks is a subject of intensive research in recent decades. The interest in this subject is associated with the need to reduce global warming[1,2], with stimulation of oil and gas fields by carbon dioxide injection, and also with the use of energized fracturing fluids[3]. Despite numerous works there are still many doubts about the behavior of hydrogeochemical systems rocks impacted by injection of the acid gas. Knowledge of that issue requires deepening especially on the basis of experimental studies. In order to partially identify these phenomena laboratory experiments and modeling were performed to determine the impact of CO_2 and H_2S or their mixtures on the rocks typical for the Upper Silesian Coal Basin – USCB (Poland).

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2. Materials and methods

Core samples were selected to represent cap rock (mudstone) and aquifer (sandstone) typical of the Upper Silesian Coal Basin. Mineralogical composition of mineral assemblages was determined by XRD (Table 1). Porosity was measured by means of mercury porosimetry. Identification of mineral phases in the samples before and after the experimental tests was performed with scanning electron microscope with EDX analyzer. In the experiments the samples were placed in an autoclave filled with the brine that reproduced original composition of pore water (Table 2). The autoclave was pressurised with CO_2 and/or H_2S gases to 75-120 bar; temperature was kept at 30-50 ° C. The experiments lasted for 75 days and allowed for simulation of gas-rock-water interactions after the gas injection into system. Composition of pore waters for hydrogeochemical simulations was calculated based on equilibration of the formation water (Table 2) with mineral assemblages typical for the modeled rocks. The pore waters were characterized by TDS ranging from 135.0 to 140.0 g/l and pH between 6.0 and 7.0.

Hydrostatic formation pressures were assumed in modeling studies, carried out with the use of Geochemist's Workbench software[4]. The temperatures were estimated according to measurements and archival data. The simplified equation kinetics of dissolution/crystallization[5] was applied in the calculations. Kinetic rate constants (K – Table 1) in modeled reactions were taken from[6]. Water-rock-gas interactions were modeled in two stages according to[7].The first stage was aimed at simulating the immediate changes in the aquifer and insulating rocks impacted by the beginning of gas injection. The second stage enabled assessment of long-term effects of sequestration. The reactions quality and progress were monitored and their effects on formation porosity and mineral sequestration capacity of carbon and/or sulfur were calculated.

Table 1. Mineral composition of rock samplesand kinetic parameters applied in modeling

Sample	Cap-rock		Aquifer		
Porosity	0,034		0,251		
Minerals	Participation [% vol.]	Specific surface [cm^2/g]	Participation [% vol.]	Specific surface [cm^2/g]	log K [$mol/m^2/s$]
Quartz	47	227	31	23	-13.99
Muscovite	14	212	28	106	-11.85
Microcline	8	235	5	23	-10.06
Albite	22	229	8	23	-10.16
Chlorite IIb	9	1120	7	1120	-11.11
Calcite	-	-	9	22	-0.03
Ankerite Fe 0,55	-	-	10	21	-3.19
Phlogopite	-	-	2	1080	-12.40

Tab. 2. Pore water composition applied in experiments and modeling

Parameter	Unit	Value
pH	-	7.8
Cl^-	mol/kg	2.831
SO_4^{2-}	mol/kg	$7.8 \cdot 10^{-4}$
HCO_3^-	mol/kg	0.002
Ca^{2+}	mol/kg	0.939
Mg^{2+}	mol/kg	0.053
Na^+	mol/kg	0.842
K^+	mol/kg	0.005

3. Experimental results

The SEM analysis revealed that skeletal grains dissolution occurred in all reacted samples, and was the most advanced in the case of carbonates and chlorite. Numerous aggregates, as well as scattered, single, small crystals of pyrite were identified amongst the secondary minerals produced during the experiment (Fig. 1A, B). Cavities situated along the cleavage planes in microcline were also observed. Cavities probably developed as a result of preferential dissolution of the potassic lamellae relative to the sodic ones, as reported in[7]. On the surface of quartz grains small cavities were found, which record the initial stage of dissolution in aggressive environment. The