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Considerations on the possibility of microbial clogging of re-injection wells of the wastewater generated in a water-dissolved natural gas field

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ABSTRACT

Brine produced from water-dissolved natural gas reservoirs should be returned to the reservoirs after the resources are recovered to prevent land subsidence. However, the ability to re-inject the brine gradually decreases and is only rectified by carrying out backwashing treatment of re-injection wells. Because the brine contains high levels of iodine also, it is also recovered from the brine using sulfuric acid and oxidizing agents. These chemicals may stimulate the growth of microorganisms that may cause the clogging. In this study, we used column experiments to investigate the possibility of the microbial clogging.

Significant clogging was observed on the columns that were treated by the brine containing both indigenous microorganisms and dissolved oxygen. In particular, iodide-oxidizing bacteria were detected from the columns and original brine dominantly; therefore, it was assumed to have an important influence on the clogging. Iodine that was produced by iodide-oxidizing bacteria corroded iron in the sand under the presence of dissolved oxygen. Eluted Iron formed ferric hydroxide colloid in the brine and it caused the clogging of the pore spaces.

We also demonstrated that deoxidized brine inhibited the iodide-oxidizing bacteria from becoming dominant and the column from the clogging through the column experiments. From these results, we can suggest removing dissolved oxygen as the most feasible countermeasures for the clogging.

1. Introduction

Brine produced from water-dissolved natural gas reservoirs should be returned to the reservoirs after the natural gas and other resources are recovered to prevent land subsidence. However, the ability to re-inject the brine gradually decreases and is only rectified by carrying out backwashing treatment of re-injection wells every 3–4 months. Colloidal materials are found in solid materials that are produced from re-injection wells by the backwashing treatment.

For convenience, the clogging of porous media may be organized into three classes: physical, chemical and microbial (Vandevivere and Baveye, 1992). Baveye et al. (1998) suggested that the biological clogging of porous media, often in conjunction with physical or chemical clogging, is encountered under a wide range of conditions. Martin (2010) also reported that the decline in re-injection ability is caused by microorganisms inhabiting re-injected brine and/or reservoirs around the re-injection wells. Volodymyr and Jian (2008) and Lim et al. (2010) demonstrated that the biological clogging is caused by impermeable biomasses and undissolved compounds that are generated by the reactions between microbial metabolites and minerals. Additionally, Baveye et al. (1998) reported an occurrence of biological clogging that is caused by gaseous products with low solubility. However, it is likely that the mechanisms of biological clogging differ depending on the particular re-injection systems and/or reservoirs. Therefore, the appropriate countermeasures for biological clogging will be different in each situation.

Our study focuses on a water-dissolved natural gas field in Chiba prefecture, Japan. Like other water-dissolved natural gas fields in Japan and oilfields in United States, the brine produced in this field contains high levels of iodine, which is a natural resource used in photography, medicine, etc., in addition to dissolved natural gas. Iodine is also recovered from the brine using a blowout method after the dissolved natural gas has been separated from the brine. Sulfuric acid and oxidizing agents are put into the brine during the iodine extraction process, resulting in an abundance of sulfate and dissolved oxygen in the re-injected brine. Sulfate and dissolved oxygen may stimulate the growth of sulfate-reducing bacteria (SRB) and

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aerobic bacteria respectively. In particular, SRB are well known to create biofilms on the surface of oil well pipes and cause corrosion and souring in oilfields (Davidova et al., 2001; Marchal et al., 2001; Eckford and Fedorak, 2002; Crolet, 2005; Xu et al., 2011). Aerobic bacteria, such as sulfur-oxidizing bacteria (SOB), are known to create biofilms under sulfur-rich conditions (Juan et al., 2007). It is likely that biofilms created by these bacteria in the sulfate-rich re-injected brine clog the slotted pipes and/or pore spaces.

In this study, we used column experiments and bacterial analyses to investigate the influence of brine microorganisms on the clogging of re-injection well pipes and/or pore spaces around the wells.

2. Materials and methods

2.1. Brine sampling

Two different types of brine were used for the experiments of this study. One was the brine collected prior to the extraction of iodine, and the other was the brine from which iodine had been extracted. In this paper, the former is designated as "raw brine" (RB) and the latter is designated as "waste brine" (WB).

Following the separation of dissolved natural gas, RB was collected under anaerobic conditions from production wellheads into sterile plastic bottles. Three RB samples: RB-a, RB-b, and RB-c which had been collected from three different wellheads were used for the experiments because microbial species varied with RB samples which had been collected from different wellheads as described below. Depth of target layers for samples RB-a, RB-b, and RB-c were 500–1100 m, 600–900 m, and 1500–2200 m respectively. In addition, WB from which iodine had been extracted was collected aerobically in sterile plastic bottles from a drainage outlet of the iodine extraction plant.

Chemical components of brine samples are shown in Table 1. Iodide concentration in RB samples was approximately tenfold higher than that in WB sample. In contrast, sulfate was not detected in RB samples but concentration was relatively high in WB samples. Similarly, concentration of dissolved oxygen was much greater in WB sample than in RB samples.

2.2. Preliminary column experiments for investigating influential factors on the clogging

2.2.1. Experimental setup

Columns were made by densely packing sand sampled from an outcrop in the gas field into corrosion-resistant stainless vessels. New columns were made in the same way and used for each experiment. The effective inner diameter and length of each column was 30 mm and 100 mm, respectively. The sand was composed of (%): SiO_2, 58.8; Al_2O_3, 21.9; Fe_2O_3, 11.0; CaO, 4.57; K_2O, 1.98; TiO_2, 1.66; SrO, 0.06; ZrO_2, 0.05. In addition to silica and alumina, iron was one of the major components of the sand. "Fe_2O_3" represents the total iron content expressed as Fe_2O_3. Actually, the ferrorichterite ($Na(CaNa)Fe_5^{2+}Si_8O_{22}(OH)_2$) was detected as a main iron mineral from the sand by X-ray diffraction (XRD) analysis.

Fig. 1 shows the particle size distribution of the sand. The particle size range of the sand was from 0.28 μm to 1290 μm. The median particle size of the sand was 61 μm.

A constant head approach was used to estimate the clogging of the column, as shown in Fig. 2. In this method, the flow outlet of the column was placed in the atmosphere and the outflow rate of the brine was measured under constant head conditions. The constant head was maintained with a Mariotte bottle arrangement. Decrease in brine surface in the Mariotte bottle was measured every 4–8 h, and change in flow rate was evaluated. A 0.2 μm membrane filter was installed at the air intake of each Mariotte bottle to prevent microbial contamination of the brine.

2.2.2. Influential factors considered in this study

We investigated the influence of dissolved oxygen, sulfate and iodide, whose contents were quite different between RB and WB as shown in Table 1, on the clogging.

The column experiments were carried out using untreated brine samples (Run 1) and the brine supplemented with 100 mg l^{-1} of sulfate (Run 2). Filter-sterilized brine was also used for the experiments to estimate the influence of microbes on the clogging (Run 3). Table 2 outlines the presence or absence of dissolved oxygen, sulfate, iodide and microbes in the brine used for the column experiments.

The influence of dissolved oxygen was estimated by comparing the experimental results obtained under aerobic and anaerobic conditions. Anaerobic column experiments were carried out in an anaerobic chamber with oxygen concentrations <0.05%. The influence of sulfate and iodide was estimated by comparing results of the experiments using RB and WB. The influence of sulfate was also investigated in experiments using RB samples supplemented with sodium sulfate (final concentration 100 mg l^{-1}). Microbial species which influenced the clogging could be also investigated through the experiments because microbial species varied with brine samples as described below.

2.2.3. Experimental procedure

Columns were vacuum saturated with the brine prior to experimentation. Subsequently, the valve between the bottle and

Table 1
Chemical components of the raw brine and the waste brine used in this study.

	RB-a	RB-b	RB-c	WB
pH	7.8	7.8	7.8	6.7
I^-	129	123	105	10
Cl^-	18,000	19,000	20,900	16,900
SO_4^{2-}	<0.5	<0.5	<0.5	120
NO_2^-	<0.5	<0.5	<0.5	2.9
NO_3^-	<0.5	<0.5	<0.5	1.0
HCO_3^-	1460	1070	1100	880
NH_4^+	181	181	172	250
Ca	110	210	320	190
Fe	<0.5	<0.5	<0.5	<0.5
Na	9900	10,400	11,000	9900
K	310	340	420	300
Mg	340	460	230	300
Mn	<0.5	<0.5	<0.5	<0.5
Dissolved oxygen	0.28	<0.5	<0.5	6.28

Unit: mg/l.

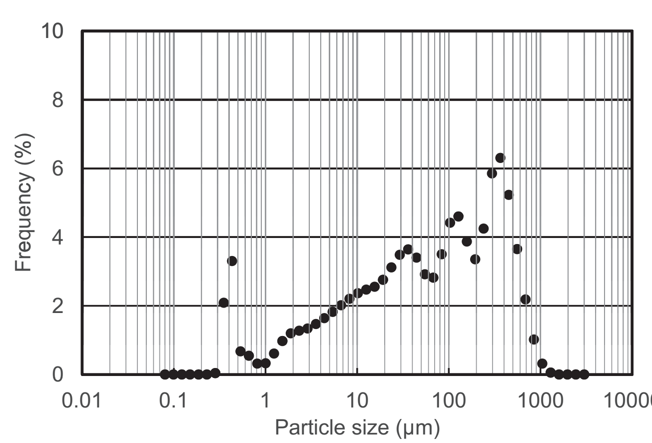

Fig. 1. Size distribution profile of the sand grains used in this study.