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Separation and desulfurization of bitumen from oil sand using oxidation treatment combined with ultrasound

超音波併用酸化処理によるオイルサンドからのビチューメンの分離と脱硫

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1. Introduction

Major deposits of oil sands have been found in Alberta, Canada, and Orinoco, Venezuela.[1] Oil sands are a mixture of bitumen (heavy oil), sand (siliceous material), and water. Bitumen can be used as a fuel and its content in oil sand is 10–15 wt %. Therefore, it must be separated from sand particles before use. Normally, the oil sand, which is mined by open-pit mining method, requirs treatments with hot water (<100 °C) and steam (>100 °C) in rotating drums, because the viscosity of bitumen is very high (>50,000 cP). Therefore, the energy consumption of high temperature treatment is a problem. To overcome this problem, researchers have been studying on recovery of bitumen from oil sand at lower temperature. Previous study, we also succeeded bitumen recovery from oil sand at low temperaure, 85 °C, using hydrogen peroxide solution (H_2O_2) and sodium hydroxide (NaOH) with ultrasound irradiation.[2, 3]

Alberta bitumen includes high concentration of sulfur of 4.6±0.5 wt %. Most of the sulfur is organic sulfur, 62% is aromatic and 38% is aliphatic sulfur.[4, 5] Hydrodesulfurization (HDS) is used commercially to decrease the sulfur content of bitumen. However, the HDS process requires a high temperature (>300 °C) and the high-temperature process consumes large amounts of energy. We have been focused on oxidative desulfurization (ODS), which can be conducted under mild conditions, including low temperature and pressure. ODS method can remove sulfur from bitumen into the solution by oxidation of sulfur using H_2O_2 follwed by removal of it using an alkaline solution.

Therefore, the problem of above both processes, bitumen recovery and desulfurization, is consumption of significant amounts of energy. In the previous study, we demonstrated the simultaneous recovery and desulfurization of bitumen from oil sand using oxidative desulfurization with ultrasonic irradiation and tetrahydrofuran (THF) at 20 °C.[6, 7] We successfully recovered 88% of the bitumen from oil sand and removed 42% of the sulfur from the bitumen. In this study, we invesitigated the appripriate condition to the simultaneous recovery and desulfurization of bitumen from oil sand by changing the concentration of H_2O_2, NaOH and THF with ultrasonic irradiation at 45 °C. In this munuscript, we introduce the effect of the concentration of THF on recovery amount of bitumen and desulfurization ratio of bitumen.

2. Experimental

Alberta bitumen which includes 5.2 wt % of sulfur was used. Bitumen content in oil sand is 12.3 wt %. The experimental apparatus is shown in **Fig. 1.** Sonication was performed using an ultrasonic generator (Kaijo TA-4021) and a 28 kHz transducer. The output of the sonication device was adjusted to 200 W. Alberta bitumen (3 g) was poured into a solution containing H_2O_2 (3 and 5 wt %, 15 ml) and THF (15 ml). The solution was then irradiated for 10 min at 45 °C. Subsequently, saturated NaOH (15 ml) was added into the solution, and the solution was again irradiated for 10–180 min. The treated solution was placed in a water bath at 70 °C until THF was completely evaporated. The treated bitumen was then collected, washed, and dried, and the sulfur content in the treated bitumen was analyzed by combustion ion chromatography. The weight of recovered bitumen was measured using an electronic balance.

3. Results and Discussion

Fig. 2 shows the results of the recovery rate of bitumen from oil sand using ultrasound irradiation for 10 min at 45 °C at various additional amount of THF (0, 5, and 15 ml). Comparison with the recovery ratio of bitumen treated by ultrasound, stirring condition (750 rpm) also conducted. The recovery ratio of ultrasound was higher than that of stirring at each additional amount of THF. Especially, recovery ratio of bitumen at 15 ml of THF showed 100%. The difference of recovery ratio between ultrasound treatment and stirring

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treatment must be come from the difference of surface stripping effect of bitumen from oil sand particles. And the difference of recovery ratio by the additional amount of THF must be come from the difference of bitumen viscosity. When 1 g of bitumen dissolved into 15 ml of THF, the viscosity was changed from 24,870 cP of original bitumen to 218 cP at 45 °C. This viscosity is almost same with that of original bitumen without addition of THF at 100 °C.

Next, we demonstrated the simultaneous recovery and desulfurization of bitumen from oil sand using 15 ml of THF and ultrasonic irradiation at 45 °C. H_2O_2 was added into the solution to adjust the concentration of the solution at 0, 3, and 5 wt %. After irradiation of oil sand for 10 min in H_2O_2 and THF, the oil sand formed an emulsion in the solution. Subsequently, saturated NaOH (15 ml) was added into the solution, and the solution was again irradiated for 10–60 min. **Fig. 3** shows the desulfurization ratio for the bitumen treated under each H_2O_2 concentration at 45 °C. The desulfurization ratio increased when the irradiation time increased till 60min. Under the condition of 3 wt % of H_2O_2 and 60 min of irradiation, bitumen recovery ratio and desulfurization ratio were reached to 93% and 86%, respectively. Abramov reported that 95% of bitumen could be recovered by the treatment using alkalis (NaOH, Na_2SiO_3) and ultrasound at 67 °C. Comparison with this reported ratio of bitumen recovery, our result is good. Furthermore, desulfurization ratio was also showed high percentage of 86% at low temperature of 45 °C.

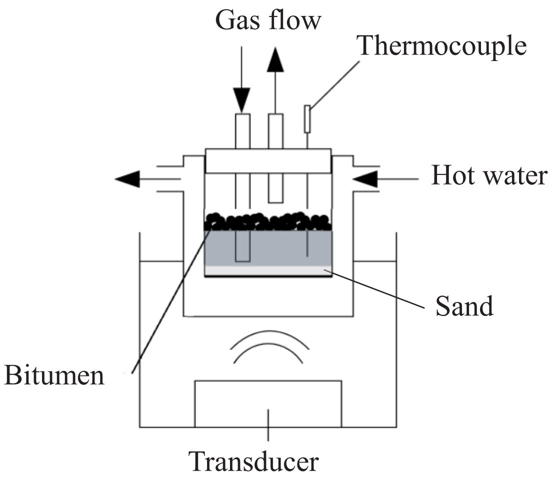

Fig. 1 Schematic design of experimental apparatus of recovery and desulfurization of bitumen at 45 °C with ultrasound irradiation.

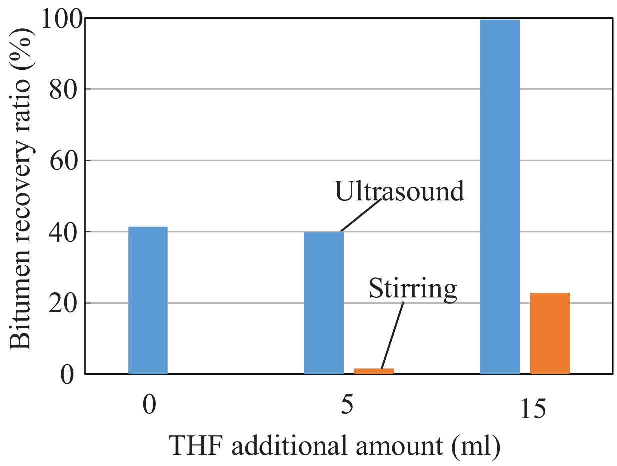

Fig. 2 Recovery ratio of bitumen from oil sand using ultrasound or stirring for 15 min at the condition of solutions (30 ml) containing each amount of THF (0, 5, and 15ml) at 45 °C.

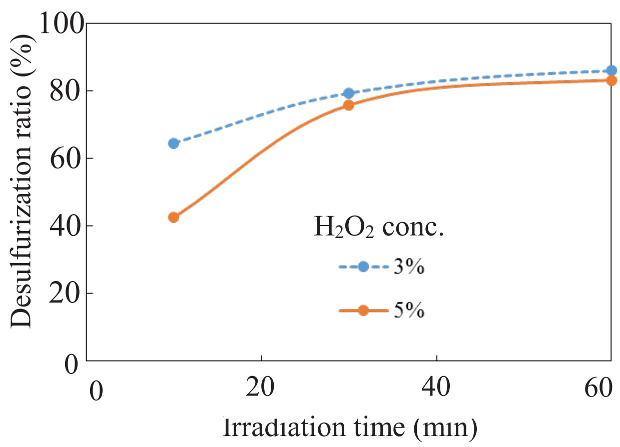

Fig. 3 Effects of ultrasound irradiation time and H_2O_2 concentration on desulfurization ratio of recovered bitumen from oil sand.

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