

## The Effect of Kaolinite Particles on Enhancing Oil Recovery from Sandstone Reservoirs

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<https://doi.org/10.15017/1866311>

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出版情報 : 九州大学, 2017, 博士 (工学), 課程博士  
バージョン :  
権利関係 :

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論 文 名 : The Effect of Kaolinite Particles on Enhancing Oil Recovery from Sandstone Reservoirs

(砂岩貯留層からの原油増進回収に及ぼすカオリナイト粒子の影響)

区 分 : 甲

### 論 文 内 容 の 要 旨

Oman has developed extensive intermediate and heavy oil reserves, and traditionally uses enhanced oil recovery by steam injection (steam-EOR). In Omani sandstone reservoirs, where oil is produced by steam-EOR and water flooding, large amount of oil sludge consists from clay mineral particles are included in the produced oil. The most common clays produced from sandstone reservoirs are kaolinite, illite, mica and montmorillonite. It has not been cleared what functions the clay minerals play in the steam-EOR, because low salinity water (LSW) is expected to be formed with mixing condensed water in formation brine water after steam injected into the reservoir. If clay minerals are sensitive to low salinity formation water in the reservoir, cations in the water and the oil polar components in the reservoir will affect the reservoir properties especially the wettability which is one of the major factors affecting oil production rate and recovery ratio. It also has an effect on oil and formation water flow behaviors such as oil-water emulsion formed in the reservoir during the steam injection EOR process.

In this study, two series of experiments have been carried out to clear the effects of the clay minerals on oil production from the sandstone reservoirs. First experiment was for investigating effects of kaolinite on viscosity and wettability of water-in-oil emulsion formed by steam injection, because low salinity water (LSW) has a sensitive affect to the rock matrix wettability with the presence of kaolinite during steam-EOR. Second experiment was for investigating effects of kaolinite particles on oil production by water-flooding tests using three conditions of oil and kaolinite in the Berea sandstone cores.

This dissertation consists of 5 chapters.

Chapter 1 prefaces the fundamentals of EOR and their applications in Oman oil fields. Then, the focus was on popular EOR methods which is the water flooding for intermediate oil and the steam injection/hot water for heavy/intermediate oil in Omani sandstone reservoirs. The production of sludge along with produced oil is one of the major issues faced in the oil fields. From those situations, the objectives of present research were targeted on the effects of clay minerals on viscosity of water-in-oil emulsion and their wettability after contacting with the fresh water and the mechanism of effecting oil production rate from the reservoirs.

Chapter 2 explained characteristics of the crude oils and sludge sampled from the skimming tank at two Omani oil fields. The clays consist of 66% kaolinite and 14% smectite are produced with 12% bulk volume in the subjected field. The clay minerals were separated from the oil sludge using a centrifugal separator and the Soxhlet extraction method. Types of the clay minerals and the multiple elements within the sludge sample were identified by X-ray diffraction (XRD) and Semi-Quantitative X-ray (SQX). The results showed that the kaolinite is the main content of clay minerals in these fields. Therefore, in this research the fresh kaolinite particles (powder and slurry) were used by mixing in the oil samples.

Chapter 3 presents the effect of kaolinite particles on viscosity and surface tension of water-in-oil (W/O) emulsions formed from an Omani heavy crude oil (API gravity of 21.5°) by steam injection. The W/O emulsion was formed and investigated intentionally to simulate the actual field phenomena, that during steam injection an emulsion will be formed and produced consisting of condensed water, oil, and sludge. Rheological properties of generated emulsions were investigated over the temperature range of 30 to 80 °C, by a viscometer and photomicrographs. The presence of kaolinite particles change the viscosity of W/O emulsions with about 20% lower than that without kaolinite, and induced a thixotropic behavior of W/O emulsions. The viscosity of W/O emulsion formed with 10% (w/w) kaolinite decreased with the increase in the water/oil ratio (WOR), however a reverse trend was found in the case of emulsion formed without kaolinite particles. The effects of kaolinite particles on wettability were investigated after contacting with steam condensed water. The wettability alteration was measured by contact angle of steam condensed water on the surface of oil/kaolinite mixture at various temperatures of 50, 70, and 90 °C. The wettability of the oil including kaolinite particles for distilled water (condensed water) have been compared with the case for high salinity brine about 3.14% of NaCl concentration. The result shows that kaolinite particles could alter the wettability of a formation from oil-wet to water-wet after contacting with distilled water by reducing the contact angle about 40% lower than that of oil without kaolinite particles. Those behaviors show a possibility of enhancing oil production by decreasing the viscosity and changing the wettability to water-wet with kaolinite particles.

Chapter 4 describes the core flooding tests to investigate the effect of kaolinite particles on oil flow behavior in the reservoir conditions. In the flooding tests, Berea sandstone cores were used saturated with intermediate oil (API gravity of 30°) sampled from the Omani sandstone reservoir that producing oil sludge. To replicate the reservoir conditions, kaolinite-fine particles slurry of 0.4µm in average size was used to be injected to the sandstone cores for water-flooding tests that consist from two flooding stages using formation brine water and condensed water as LSW. Three cores having almost the same properties of permeability and porosity, 50 mdarcy and 0.28 % respectively was set to three different conditions of oil and kaolinite-fine particles for each water-flooding test. The first core (C1) was saturated with oil only, the second one (C2) was filled up with kaolinite-fine particles slurry then saturated with the oil, and the third one (C3) was saturated with the mixture of kaolinite-particles slurry and the oil. The clay was mixed in the oil by 10% (w/w) that is same ratio of the C2 core. The results of the water-flooding tests showed that 30 % increase of oil recovery was obtained in the cases including kaolinite fine particles compared to that of oil only. In addition, the wettability of the cores contained kaolinite fine particles showed stronger water-wet than the core without kaolinite particles. Zeta potential measurement was also conducted to measure the surface charge of kaolinite particles in brine and water. The kaolinite fine particles were negatively charged as -15 mV in the brine, while it was -50 mV in the condensed water as LSW used in the flooding test. The difference in kaolinite charges can explain the reason of increasing oil recovery ratio in the water-flooding test, was induced by kaolinite fine particles in the cores. Injecting the LSW allow to increase the negativity charge of the clay to break the cations ( $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$ ) binding effect and detach from the surface carrying the organic oil polar component keeping the surface water-wet thus, increase the oil production. The ions were traced in the effluents in the water flooding test using the condensed water, and it was found that the concentration of  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  in the connate water reduced from their initial concentration of 722 and 788 ppm to 34 and 26 ppm respectively with pH increasing from 6.8 to below 9.0. It is concluded that presence of kaolinite fine particles induced the wettability alteration together with decreasing of salinity, increasing of pH and exchange of ions  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  ions.

Chapter 5 is a summary and conclusion of major findings with recommendations for future work.