学 位 論 文 内 容 の 要 旨

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学 位 論 文 題 名

Exploring factors influencing the formation and stability of crude oil emulsion
(原油エマルジョンの形成および安定性に影響を与える要因に関する研究)

Emulsification plays a crucial role in various areas. However, undesirable emulsions, especially water-in-oil (W/O) emulsions, formed during crude oil production, transportation, processing, and stockpiling, become significant issues in the petroleum industry. There are two ways to solve emulsification problems in petroleum: One is dependent on demulsification technologies to break the emulsions after their formation; another is to prevent emulsion formation. Preventing emulsification is the most economical and fundamental solution. Various studies have focused on evaluating the factors affecting emulsion stability and their mechanisms, which hardly explain the relationship with emulsion formation. Therefore, the main purpose of this study was to clarify the influences of different factors on both emulsion formation and stability as well as the stability mechanisms, which is beneficial for finding the ways to fundamentally avoid or reduce emulsification in petroleum.

The first part of this research work assessed the effects of pH and cations on the transformation of emulsion types and their stability based on the observed emulsion formation process and emulsion particle sizes. It was found that the final W/O emulsions were transformed from water-in-oil-in-water (W/O/W) emulsions for all brine conditions. At neutral pH, more W/O emulsions were formed through the transformation of W/O/W emulsion, but the high pH broke down most of the W/O/W emulsion into free water and oil. Emulsion particles' zeta potential or electrokinetics plays a significant role in W/O/W emulsion stability at high pH. The transformation of emulsion types in cation solutions was similar to neutral pH; the amount of W/O emulsion depends on cation species. The W/O/W emulsion stability under different pH in the following order: neutral pH < low pH < high pH, whereas final W/O emulsion stability is as follows: high pH < low pH < neutral pH; $MgCl_2 < CaCl_2 < NaCl$. The stability of the emulsion is influenced by the properties of the crude oil, such as viscos-

ity and TAN/TBN ratio, which affect the oil film thickness of the W/O/W. Based on these findings, a stability mechanism is proposed for the emulsion under different pH and cations.

The next part evaluated the impacts of temperature and crude oil properties on emulsion formation and stability of crude oils. Regardless of temperature and crude oil properties, W/O emulsion was transformed from W/O/W emulsion, and there was no big difference of W/O emulsion particle size after transformation. With the temperature increase, the particle size of W/O/W emulsion for most crude oils became smaller due to the viscosity and interfacial tension shifts, while the transformation rate was accelerated. The most stable W/O/W emulsion was formed by crude oil with lowest viscosity at high temperature. Consistent with emulsion formation process, four laws of resolved water percentage variations for various oils were obtained under temperature effect. For crude oil with a viscosity lower than 10 cp, high temperature can significantly reduce the W/O emulsion amount. Moreover, due to promoting water droplet coalescence, high temperature destabilized the W/O emulsion resulting in larger W/O emulsion particles. On the other hand, comparing the FTIR spectrum of crude oils and their W/O emulsions, the oxygen/nitrogen-containing functional groups contribute to forming crude oil emulsions. Hydrogen bonded O–H mainly dominates the W/O emulsion amount of crude oils, followed by hydrogen bonded N–H.

The final part discussed how emulsion formation and stability of various crude oils were affected by homogenization conditions including homogenizing speed and homogenization duration. Particle size, the amount, and transformation speed into W/O emulsion of W/O/W emulsion was increased at high homogenization speed where the transformed O/W emulsion was more stable. In a certain speed range, more complicated emulsion, W/O/W/O/W emulsion, can be formed for crude oil with special viscosity determined by crude oil property. At low homogenizing speed, the multiple emulsion electrokenitics of crude oils were mainly controlled by TAN, while TBN had a significant influence at 30000 rpm. Additionally, extending homogenization time can increase the amount of W/O emulsion for nine different oils, especially crude oils with high molecular weight. Furthermore, W/O emulsion stability was increased for longer homogenization time. Viscous solids considered as asphaltenes were precipitated for crude oil 6# from homogenization time extending to 60 min.