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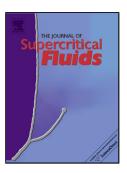
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ACCEPTED MANUSCRIPT

Deuterium tracing study of unsaturated aliphatics hydrogenation by supercritical water during upgrading of petroleum heavy oil. Part I: Non-catalytic cracking

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Abstract. It is believed that the unique solubility and diffusivity of organics in supercritical water (SC-H₂O) have a positive influence on cracking and upgrading of heavy oil; however, the important issue would be the chemical reactivity of SC-H₂O. By applying isotope labeling technique, the present study has verified the chemical role of SC-H₂O as the environmentally benign hydrogen-donor solvent in cracking the petroleum vacuum residue. The high pressure batch experiments were applied at several temperatures (350- 450°C) and pressures (170-50 MPa) keeping density constant at ca. 0.4 g/ml, respectively. After 1h contact time between heavy water (D₂O) and vacuum residue (VR) in a ratio of 80/3 g/g, the hexane soluble product (maltene) was analyzed by attenuated total reflectance (ATR) spectroscopy. Substantial carbon-deuterium bonding (16% relative to overall hydrogenation by water and hydrocarbon) was detected in saturated compounds (alkanes) produced from the cracking process with water, higher than 425°C as the temperature beginning of considerable miscibility between water and oil. Accordingly, together with the modified physical properties (solvation and dispersion effects), hydrogen so formed from SC-H₂O would be accessible to cover radical species formed during the cracking of asphaltenes and other oil species, and consequently reduced coke formation compared with the dry pyrolysis. This kind of in-situ hydrogen donation is in competition with hydrogen elaborated from condensation of polycyclic aromatic in the oily phase in which formation of coke is deduced. The higher portion of hydrogen by water relative to the hydrocarbon condensation causes reduction in radical addition reaction rate and suppression of coke, therefore allows the higher conversion of residues to desirable liquid products.

Keywords: Isotope labeling technique, supercritical water, deuterium oxide, heavy oil cracking, hydrogen donor solvent.

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