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Modeling of asphaltene aggregation phenomena in live oil systems at high pressure-high temperature

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ABSTRACT

In spite of the development of different experimental methods and theoretical models devoted to understand and predict the asphaltene behavior in dead/model oils, the asphaltene aggregation phenomena in live oil systems at high pressure-high temperature conditions has been grossly missing in the available literature. In this study, a population balance model with geometric scaling approach is proposed to simulate the asphaltene aggregates growth in time for live oil systems. Appropriate collision kernels are incorporated to describe the aggregation mechanisms taking into account the effect of pressure, temperature and oil characteristics. In modeling of the asphaltene aggregation phenomena in live oil, asphaltene aggregates are considered to be pressure-temperature dependent fractal structures. Data from depressurization experiments at different temperatures, performed by high pressure microscopy, are used to validate the model. Sensitivity analyses are performed to assess the effects of model parameters on simulation results. The experimental results which are obtained through the image analysis of the depressurization process in live oils are predicted by the model with a good accuracy. The fractal analysis of the asphaltene aggregates in live oil samples at elevated pressures and temperatures discloses that the fractal dimension of the asphaltene aggregates depends strongly on pressure and temperature of the system as well as on the nature of the oil. The proposed approach can well describe the prevailing mechanisms for asphaltene aggregation in live oils at pressure and temperature conditions of oilfields.

1. Introduction and background

Flow assurance issues induced by asphaltenes are of great concern to all facets of petroleum production and processing. Asphaltenes, the most problematic and the least understood compounds of the oil and energy industry, are characterized by their solubility in aromatic solvents and their insolubility in saturated hydrocarbons [1,2]. Asphaltenes are destabilized as a result of changes in pressure, temperature, and composition during different operations [3,4]. They are responsible for a variety of problems in both upstream and downstream processes [5–7]. Wettability reversal of the reservoir rock, reduction of the well productivity and increasing the overall operating cost, plugging surface facilities and flow lines, contribution to the catalyst deactivation, and equipment fouling are some of their detrimental consequences [1,8]. Unstable asphaltene systems undergo consequent processes: beginning with “destabilization”, i.e. the transition of an asphaltene on the nanometer length scale from stable (no growth in size) to unstable (growth in size), followed by “aggregation”, i.e. the growth process of an asphaltene from the nano-meter to micro-meter and/or larger length scales, and finally “deposition” [9,10]. The precipitation will refer to the entire process of asphaltenes transitioning from stable nano-aggregates to micrometer-sized aggregates or precipitates [9]. The asphaltene stability and the mechanisms of its aggregation in crude dead oils or model oils have attracted massive experimental/modeling research attentions for several decades [1,11–24].

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These researches are mostly limited to the extracted/synthetic asphaltenes formed by addition of the saturated hydrocarbon solvents (e.g. nC_5 or nC_7) at standard lab conditions. It should be stressed that conclusions drawn from the lab asphaltene studies which are numerous and much easier to obtain may not necessarily follow for the asphaltenes formed in live oils at field conditions of pressure and temperature [25]. The majority of the previous works on thermodynamics of asphaltene stability are based on the assumption that asphaltene destabilization is an instantaneous phenomenon and the kinetic effects associated with the asphaltene aggregation have not been considered [26–30]. Neglecting the kinetic effects on asphaltenes aggregation can produce misleading results leading to ill-informed decisions under operational conditions. The many experiments reported to date indicate that asphaltene aggregation is time-dependent process that occurs very slowly [14,19,31].

Despite the extensive experimental/theoretical efforts trying to understand the aggregation behavior of asphaltenes and the morphologies of the fractal-like aggregates, the asphaltene aggregation process in live oils at high pressure-high temperature (HPHT) conditions is rarely attended in the available literature. The aggregation rate of asphaltenes is governed by the collision frequency of destabilized particles and their collision efficiency [13]. Collision (attachment) efficiency is the fraction of successful collisions between destabilized particles in the presence of energy barriers, which is equal to zero in the case of strong repulsion between particles and approaches unity when attraction forces are dominant. While the collision frequency depends on the particles concentration, size distribution of the particles, and temperature of the system, collision efficiency is a function of the strength of interaction forces between aggregating asphaltenes [32,33]. The aggregates size-growth is affected by the driving forces (precipitants such as hydrocarbon solvents for asphaltenes formed in dead oils at laboratory conditions, and pressure or temperature for asphaltenes formed in live oils) which contribute to the asphaltene destabilization. Different modeling approaches coupled with various experimental techniques have been applied to monitor the evolution of the aggregates size distribution at different conditions. The classical work of Smoluchowski is the first major attempt to model the aggregation behavior of colloidal systems, which laid the foundations of the subject [34]. Today this model is still the basis for many more advanced or specific kinetic models of aggregation. The particle size distribution and its evolution in time can be predicted using population balance modeling, which have been widely employed in studies of particulate and colloidal systems [13,15,18,35–40].

In this work, a geometric population balance model is developed to simulate the growth and size distribution of asphaltene aggregates in live oil systems at HPHT conditions. In modeling the aggregation process of asphaltenes in live oil, asphaltene aggregates are considered as pressure-temperature dependent fractal structures. Also, the effects of pressure and temperature on model parameters are fully considered. The presented model is validated with data from the depressurization experiments at different temperatures performed by high pressure microscopy (HPM) in two Iranian live oils with different stability at elevated pressures and temperatures. Also, sensitivity analyses are done to evaluate the effect of model parameters on simulation results.

2. Experimental method

2.1. Fluid samples

Two different live oils (referred to as sample-A and sample-B) from Iranian oil fields were used in this work. Single-phase sampling technology (SST) was used to collect representative oil samples for asphaltene study [41]. Table 1 shows the general properties and SARA (saturate, aromatics, resins, and asphaltenes) analysis of the oil samples used in this study.

Table 1
General specifications of the oil samples.

Specifications	Unit	Sample-A	Sample-B
H_2S	(mol%)[a]	0.37	0.00
N_2	(mol%)[a]	0.07	0.02
CO_2	(mol%)[a]	1.44	0.20
C_1	(mol%)[a]	50.68	37.11
C_2	(mol%)[a]	9.26	7.51
C_3	(mol%)[a]	6.11	5.90
iC_4	(mol%)[a]	1.24	1.15
nC_4	(mol%)[a]	2.84	3.79
iC_5	(mol%)[a]	1.04	1.57
nC_5	(mol%)[a]	1.17	1.64
C_6	(mol%)[a]	2.82	4.81
C_7	(mol%)[a]	3.09	4.25
C_8	(mol%)[a]	2.85	3.58
C_9	(mol%)[a]	3.02	3.27
C_{10}	(mol%)[a]	2.01	2.81
C_{11}	(mol%)[a]	1.44	2.60
C_{12}^+	(mol%)[a]	10.54	19.81
Molecular weight of reservoir fluid	(g mol^{-1})	79.9	122.1
Reservoir temperature (T_r)	K[b]	408	366
Reservoir pressure (P_r)	MPa[c]	64.2	41.0
Saturation pressure (P_b)	MPa	25.7	17.7
Gravity of dead oil	°API	33.8	25.0
Solution gas oil ratio (GOR)	(vol vol^{-1})[d]	259.1	97.8
Oil density at reservoir temperature and pressure (ρ_{or})	(kg m^{-3})[e]	654.8	750.3
Saturates	(mass%)[f]	64.90	54.50
Aromatics	(mass%)[f]	29.10	30.30
Resins	(mass%)[f]	5.10	12.30
Asphaltenes	(mass%)[f]	0.90	2.90

[a] The uncertainty is $\pm 5 \times 10^{-3}$.
[b] The uncertainty is ±0.1 K.
[c] The uncertainty is $\pm 5 \times 10^{-2}$ MPa.
[d] The uncertainty is $\pm 6 \times 10^{-2}$.
[e] The uncertainty is ±7 kg m^{-3}.
[f] The uncertainty is $\pm 5 \times 10^{-4}$.

2.2. Instrumentation and procedures

A flow assurance system equipped with a HPHT visual PVT cell, a HPM, and a HPHT filtration is utilized to investigate the asphaltenes aggregation behavior in live oils at HPHT conditions. The schematic diagram of the experimental setup is shown in Fig. 1. The HPM enables direct visual observation of the solid particles (e.g. asphaltene) at elevated pressures and temperatures. It also permits to monitor the growth of asphaltenes as a function of pressure, temperature, and effect of various chemical treatments. Using the HPM system, a series of isothermal depressurization (pressure depletion) experiments are conducted to monitor the evolution of the asphaltene aggregates. During the depressurization process, the pressure is isothermally and discontinuously reduced at discrete steps. At each step the pressure is kept constant over time to reach stable conditions of aggregates growth, defined as equilibrated time ($t_{equilib}$). At $t_{equilib}$ no more changes are observed in the aggregation process over time at each constant pressure-step and fixed temperature. The cell contents are observed visually and recorded by the HPM video camera during the course of the experiment. Measurements and image analysis/processing are performed using *ImageJ*® package, version 1.44. The visual PVT cell is used to study and quantify the phase behavior and properties of reservoir fluid,