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Determination of asphaltene precipitation conditions during natural depletion of oil reservoirs: A robust compositional approach

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ABSTRACT

Asphaltene precipitation causes rigorous problems in petroleum industry such as: relative permeability reduction, wettability alteration, blockage of the flow, etc. Therefore, accurate determination of onset pressures of asphaltene precipitation is necessary. These pressures can be obtained by experimental measurements on representative samples of the crude oils; however, laboratory analysis of crude oil samples is costly, time consuming and cumbersome. In this communication, three simple and accurate expressions have been proposed for prediction of lower and upper onset pressures of asphaltene precipitation as well as saturation pressures. To this end, 33 crude oil samples were collected from open literature sources. Afterward, two constrained multivariable search methods, namely generalized reduced gradient (GRG) and successive linear programming (SLP), were employed for modeling and expediting the process of achieving a good feasible solution. Then, comparative studies were conducted between the developed equations and equations of state as well as empirical correlations. The results illustrate that the developed equations are accurate, reliable and superior to all other published models. The results show that the proposed equations can predict lower onset pressure, upper onset pressure and saturation pressure with average absolute percent relative errors of 5.04%, 3.93%, and 3.81%, respectively. Besides, it is found that molecular weight of heptane-plus fraction has the greatest impact on the lower onset pressure, while methane has the most significant effect on both of the saturation and upper onset pressures.

1. Introduction

Asphaltene precipitation alters the fluid flow rate in the reservoir and causes several problems. As this phenomenon alters the wettability and causes permeability reduction and operational problems, studying the conditions at which the asphaltenes precipitate out of the solution, and determining the amount of precipitated/deposited asphaltene are of vital importance [1,2]. Experimental determination of asphaltene phase behavior is expensive, cumbersome, and time consuming. Therefore, many researchers have focused on developing predictive models for asphaltene phase behavior [3–9]. These models are mainly classified into five main groups which include, polymer solubility models, equation of state (EOS) models, colloidal techniques, thermodynamic micellization approaches, and scaling law models [10,11].

Solubility model was introduced by Hirschberg et al. [4], to predict the heat of solution. The liquid–liquid equilibrium is computed based on the Flory–Huggins polymer solution model

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[12]. Other researchers proposed similar models in which the fugacity of asphaltene was related to the liquid fugacity [6,7,9,13].

The equation of state approach was introduced by Gupta [14], in which the thermodynamic behavior of the asphaltene is predicted. Nghiem et al. [15] applied Peng-Robinson (PR) EOS to determine asphaltene precipitation considering the precipitating and non-precipitating pseudo-components. This approach needs tuning the EOS parameters using experimental results. Leontaritis and Mansoori [16] proposed the colloidal model, which then completed by Park and Mansoori [17]. In the colloidal model, asphaltene is assumed as a solid element which is bounded by resins. This model computes the chemical potential of asphaltene using two approaches, namely static and dynamic approaches. Wu et al. [18,19] applied the concept of Gibbs free energy and introduced micellization model. In this model, it is assumed that the mixture is composed of oil, resin, asphaltene compounds, and micelles as well as the deposited phase. Buenrostro-Gonzalez et al. [20] represented the molecular thermodynamic model in which both asphaltene and resin are pseudo-pure components. In this model, asphaltene precipitation calculations are obtained using liquid–liquid equilibrium.

Many experimental and theoretical studies have introduced the asphaltene precipitation outcomes. Some researchers have focused on determining the onset point of asphaltene precipitation [7,21,22]. Other studies have been conducted to measure the amount of asphaltene precipitation. The thermodynamic properties that affect the asphaltene precipitation include, pressure, temperature, resin and asphaltene content, as well as the crude oil composition. In some studies, it is assumed that asphaltene molecules with large molecules of resins form micelles and the precipitation occurs due to some polymerization reactions in crude oil. In other words, an asphaltene micelle particle has a core, formed by a large number of asphaltene and resin molecules which adhere on the surface of the core. Resins are responsible for the stability of asphaltene in the solution. If asphaltenes are not surrounded by resins, they would precipitate out of the solution [11,23,24]. This is due to low solubility of asphaltene materials in the crude oil. The other thermodynamic parameters that affect asphaltene precipitation include pressure and temperature changes. Changing in the crude oil composition during production or gas injection is another parameter that affects the amount of asphaltene precipitation. The effect of temperature on asphaltene precipitation is controversial. Some studies reveal that at low temperatures, asphaltene becomes more concentrated and susceptible to precipitation. Peramanu et al. [25] studied the effect of temperature in the range of 60–120 °C by using normal heptane as the precipitator. They showed that the stability of asphaltene is higher at lower temperatures. This means more amount of solvent is required for asphaltene precipitation. Otherwise, if the system is kept at high temperatures, the stability of the mixture is decreased; hence, less amount of solvent is needed for the precipitate formation. Hirschberg et al. [4] explained this phenomenon by solubility parameter in which rising the temperature, would increase asphaltene precipitation due to breaking the resin-asphaltene bounds.

The most important parameter affecting asphaltene precipitation is pressure. Hirschberg et al. [4] expressed the effect of pressure on asphaltene precipitation using a thermodynamic model. While the reservoir pressure is higher than saturation (bubble point) pressure, the asphaltene solubility increases with pressure. The minimum solubility and maximum asphaltene precipitation occur at bubble point pressure. Some studies claimed that asphaltene precipitation is a reversible process at high temperatures [4]. The reversibility of asphaltene precipitation at low temperatures is still a challenging topic. As the dissolution of the precipitated asphaltene is a slow process, it takes a long time for the reversibility [26]. However, at high temperatures, this process is proved to be reversible with respect to pressure or composition [27–29].

The stability of asphaltene at reservoir conditions is studied by finding the upper and lower onset pressures of asphaltene precipitation. Besides, determining the saturation pressure is very crucial as the maximum amount of asphaltene precipitation is occurred at this point. Asphaltene precipitation phase envelope of an Iranian crude oil sample is illustrated in Fig. 1.

EOS's are able to predict the aforementioned pressures; however, due to complexity of asphaltenes phase behavior, EOS's fail to accurately predict these pressures. Moreover, EOS's have many adjustable parameters which should be tuned from experimental data points. Fahim [10] developed three distinct models for correlating upper and lower onset pressures as well as saturation pressure. He used regression analysis to develop his models. However, these models fail to satisfactorily predict the aforementioned pressures in many crude oil samples. Besides, some intelligent models have been developed for prediction of the amount of asphaltene precipitation and onset pressures [1,30–32]. It should be pointed out intelligent models such as least square support vector machine (LSSVM), artificial neural network (ANN), ANN optimized by particle swarm optimization (PSO), have been used to model different parameters and properties in oil and chemical industries [33–36]. However, these models are black box and cannot provide clear relationship between the models' output and the inputs. Recently, novel mathematical techniques have been used in oil and gas industries to accurately model some important parameters. LINGO (Linear Interactive and General Optimizer) software has been successfully applied in mathematics, science, and industry. Very recently, we used this software and applied constrained multivariable search methods for correlating crude oil viscosity [37] and PVT properties of crude oil systems [38]. The obtained models have very simple mathematical formats and can predict the modeled properties with a high degree of accuracy.

This study aims to develop three simple and accurate models for predicting the upper and lower onset pressures as well as saturation pressure using constrained multivariable search methods. To this end, a data set (33 crude oil samples) covering a large range of reservoir conditions (i.e., temperature and pressure) and fluid properties are collected from open literature sources. Afterward, appreciate input parameters for the models are selected. Next, Generalized Reduced Gradient (GRG) and Successive Linear Programming (SLP) as two constrained multivariable search methods are incorporated to develop three accurate models for the prediction of interested pressures. Moreover, to compare the accuracy and validity of the developed models and pre-existing models (SRK EOS, PR EOS, and Fahim's models), statistical and graphical error analyses are utilized, simultaneously. Besides, to deepen our understanding about the influence degree of each parameter on the

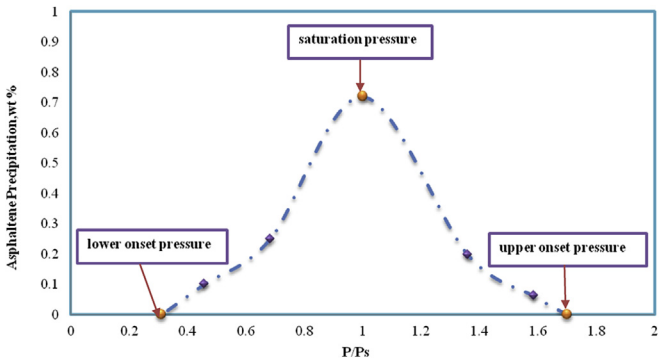

Fig. 1. The amount of asphaltene precipitation as a function of pressure ratio (pressure divided by saturation pressure) for an Iranian crude oil sample [1].