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### ACCEPTED MANUSCRIPT

## The effect of aromatic solvents on the onset and amount of asphaltene precipitation at reservoir conditions: experimental and modeling studies

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#### Abstract

High-pressure filtration and standard IP-143 were employed to determine the amount of precipitated asphaltene for a live oil sample and its mixtures with toluene and p-xylene. To find the upper AOP of each mixture, the obtained precipitations curves were extrapolated. PC-SAFT and solid model were used to predict the effects of several aromatic solvents on the onset and amount of asphaltene precipitation. Both models were tuned to the experimental amount of precipitated asphaltenes in the original oil and AOP of the oil mixed with toluene.

The results of this study suggest that PC-SAFT can reasonably predict the effects of aromatic solvents on the asphaltene phase behavior. The accuracy of PC-SAFT predictions was not greatly affected by the concentration and type of solvent. While solid model gave good predictions of the precipitated asphaltene for one mixture with low concentration of toluene, the prediction accuracy dropped with solvent concentration and change of solvent. Whereas PC-SAFT showed a positive correlation between the dissolving power and solubility parameter of aromatic solvents, no general correlation between these two parameters was predicted by solid model. Both models, nonetheless, predicted an almost linear relationship between the upper AOP and solvent concentration, consistent with experimental results.

*Keywords:* Asphaltene precipitation, Asphaltene onset pressure, PC-SAFT, Solid model, high-pressure filtration

#### 1. Introduction

Asphaltene is defined as a solubility class of heavy oil soluble in aromatic solvents; such as toluene and insoluble in normal paraffinic solvents; like n-heptane (IP-143, 2004). Asphaltenes may precipitate in the wellbore and near well region, leading to formation damage and wellbore plugging. Asphaltene precipitation may be triggered by a number of factors, including variations in the oil pressure, temperature and composition.

A number of models have been developed to predict the asphaltene phase behavior. These can be broadly categorized as either "colloidal model" or "solubility model". Asphaltenes in a colloidal model are assumed as colloidal particles suspended in the crude oil by resins absorbed on their surfaces. Any factor leading to resin desorption below a certain level can cause asphaltene precipitation (Subramanian et al., 2015). The solubility-based models assume that asphaltenes are dissolved in crude oil. Asphaltenes, in these models, can precipitate if their solubility drops below a certain level (Subramanian et al., 2015). In solubility models, asphaltene and solvent phases will be in thermodynamic equilibrium if the chemical potential of components in the two phases are equal. Depending on how the asphaltene chemical potential is calculated, various solubility models are proposed which are categorized into two general classes: regular solution and equations of state.

Cubic equations of state are extensively employed to predict fluid phase behavior. They are, however, unable to accurately model mixtures of molecules with large size differences such as asphaltenic oils. Additionally, they cannot provide accurate enough predictions of liquid densities required for liquid-liquid calculations. They also rely on the asphaltene critical properties which actually do not exist because asphaltenes decompose before reaching the critical state (Panuganti et al., 2012). A recent study (Panuganti et al., 2012) revealed that SRK-Peneloux equation of state tuned to the experimental AOPs cannot produce satisfactory predictions of the asphaltene phase behavior outside the conditions it was fitted.

Solid model is one of the most commonly used models to predict asphaltene precipitation. This model uses a cubic equation of state to calculates component fugacities in gas and liquid phases, whereas solid (asphaltene) fugacity is predicted using a solid model. Asphaltene precipitation in this model is considerably less affected by asphaltene critical properties and depend on more physical properties such as solid molar volume and total asphaltene content of oil.

PC-SAFT is a fairly new equation of state that can model the asphaltene phase behavior as a liquid-liquid equilibrium. The model has shown promising results for predicting the onset of asphaltene precipitation (David Ting et al., 2003; Gonzalez et al., 2007a, 2007b; Gonzalez Rodriguez, 2008; Panuganti et al., 2012; Punnapala and Vargas, 2013; Tavakkoli et al., 2015, 2014). Nonetheless, this

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