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Effect of additives on liquid–liquid equilibrium properties of butane/bitumen systems with applications to solvent aided bitumen recovery processes

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ABSTRACT

Solvent-aided bitumen recovery processes are relatively new approaches to reduce the negative environmental impacts and production costs of steam assisted gravity drainage (SAGD). Thermo-physical properties of these systems such as density, viscosity, phase partitioning and saturation pressure are of great importance in design of solvent-aided processes. Butane is a promising solvent for solvent-aided bitumen recovery processes. Addition of light or heavier solvents to butane can provide an engineering solution to improve the efficiency of solvent-aided processes. In this study, equilibrium measurements of butane and bitumen mixture were conducted at temperatures of 40 and 60 °C and pressures well above vapour pressure of the solvent. Then, the effect of introducing a second solvent as an additive to the butane–bitumen mixture was investigated. Propane, toluene and dimethyl ether were added to the original mixtures of butane and bitumen in separate sets of experiments and changes in thermo-physical properties were determined. It was determined that adding butane can lower the viscosity of the bitumen by several orders of magnitude. It was also concluded that although propane can significantly increase the saturation pressure of the mixture, it results in higher amount of asphaltene precipitation. The effect of dimethyl ether however is favourable because not only increases the vapour pressure but also reduces the asphaltene precipitation similar to toluene.

1. Introduction

Steam assisted gravity drainage (SAGD) and other thermal recovery methods such as cyclic steam stimulation (CSS) have been applied to the heavy oil and bitumen reservoirs in Alberta for the past two decades. Mobilization of viscous bitumen by efficient delivery of thermal energy to the oil sands reservoirs by steam injection is an important element in thermal recovery processes. However, heat loss and inefficiency of SAGD requires consuming massive amount of energy leading to excessive greenhouse gas emissions. As the environmental issues become more stringent, implementation of new recovery processes in recovering heavy oil and bitumen are inevitable. Solvent-aided bitumen recovery methods are considered to be amongst new processes that are gaining popularity to compensate for the shortcomings of steam assisted gravity drainage (SAGD). In these methods, solvent in conjunction with heat reduce the viscosity of in-situ bitumen and increase the

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efficiency of bitumen production. Therefore, a solvent-aided process improves the efficiency of recovering bitumen and at the same time reduces the cost of operations and the adverse impacts of SAGD by using less water and less negative environmental impact.

Examples of co-injection of solvent and steam are abundant in literature (Redford and McKay, 1980; Redford, 1982; Nasr et al., 1991; Kar et al., 2017; Gupta and Gittins, 2006). However, these methods have not gained as much acceptance until recently due to inconclusive pilot projects, low oil prices and environmental motivation. Redford and McKay (1980) and Redford (1982) demonstrated experimentally that co-injection of the hydrocarbon additives and steam would considerably increase the recovery of Athabasca bitumen. Nasr et al. (1991) tested the co-injection of naphtha with steam and reported significant increase in the final recovery compared to the injection of steam only. Gupta and Gittins (2006) presented the initial results of the solvent assisted process (SAP) pilot test on EnCana's Christina Lake project. The findings were promising and an increase of 150 t/day in the heavy oil production was reported because of the co-injection of butane with steam. Another important result of their work was improvement in the quality of the produced oil because of the extraction of the light components by butane. Kar et al. (2017) performed some propane-SAGD tests and showed that the displacement recovery and the quality of oil increased compared to SAGD.

One of the important aspects of the solvent assisted processes that should be addressed is formation of the second dense phase, which is mostly known as the asphaltene rich phase. The effects of asphaltene precipitation on the ultimate recovery of ES-SAGD processes have been widely studied (Pathak et al., 2011; Li et al., 2011; Kar et al., 2015; Badamchi-Zadeh et al., 2011; Al-Murayri et al., 2016; Azinfar et al., 2017). However, the phase equilibrium of bitumen/solvent systems have not been studied in detail. Therefore, the phase behaviour of the solvents and bitumen or heavy oil systems should be examined to better understand the mechanisms associated with lighter component extractions as well as the overall performance of solvent-aided processes.

There are very limited number of studies that experimentally investigate liquid–liquid equilibrium of light *n*-alkanes and bitumen systems. Badamchi-Zadeh et al. (2009) studied propane and Athabasca bitumen mixtures and examined their phase behaviour. They reported existence of the second dense phase at propane concentration above 20 wt%. However, their study was mainly concentrated on mixtures with propane composition below 20 wt%.

A comprehensive experimental study on light *n*-alkanes–bitumen systems can be found in recent publications by SHARP research group at University of Calgary (Zirrahi et al., 2017a,b; Haddadnia et al., 2018a; Azinfar et al., 2018a,b). Zirrahi et al. (2017a,b) measured the solubility of light *n*-alkanes (methane, ethane, propane and butane) in MacKay River bitumen at temperatures up to 463.15 K and pressures up to 5 MPa. They also reported the density and viscosity of the bitumen rich phase and modelled their data by Peng–Robinson equation. Haddadnia et al. (2018a) measured the thermodynamic properties of Dimethyl Ether/Athabasca Bitumen including solubility, density and viscosity at temperatures of 100, 125 and 150 °C and pressures up to 6 MPa. Azinfar et al. (2018a,b) studied phase behaviour of propane/butane and bitumen fractions extracted from vacuum distillation method and proposed a generalized EoS model for their experimental data.

A detailed study of the liquid–liquid equilibrium of *n*-butane/Athabasca bitumen was carried out by Nourozieh et al. (2014). The operational pressures for liquid–liquid separation in their study were 2 MPa at 50 and 100 °C and 4 MPa at 150 °C. They reported a minimum butane concentration of 0.5 weight fraction in order to have two liquids in the system. Viscosity and density of both heavy and light phases were measured. They also reported fluctuations in the property measurements of the heavy phase. They concluded adding more butane to the system reduces the density and viscosity of the light phase. Their results showed that the overall concentration of butane and pressure have significant effect on the composition of the phases. Similar study was done by Nourozieh et al. (2012) on liquid–liquid equilibrium of Athabasca bitumen/propane systems at different temperatures and pressures as well as the extraction of light components out of those mixtures. Their results showed that at a constant temperature and initial mass fraction of the solvent, the extraction yield increased with pressure, however, the extraction yield was insensitive to the feed concentration at constant pressure. They reported that the second phase (L_2) became heavier as the pressure and solvent to bitumen ratio were increased.

Gao et al. (2017) investigated multiphase behaviour of *n*-butane/bitumen/water systems. They observed liquid–liquid separation of hydrocarbons in *n*-butane/bitumen systems with/without water at *n*-butane concentrations of 97 mol% in wide ranges of temperatures and pressures. They visually inspected the boundaries and the colour of each phase in equilibrium. Their findings indicated that with decreasing pressure the colour of L_2 became lighter suggesting a selective extraction of the bitumen component to the light phase by butane.

To design and operate a successful butane injection process many parameters such as asphaltene precipitation, vapour pressure of the system, and extraction efficiency should be studied and optimized. While higher vapour pressure is always desirable because of higher driving force in the reservoir to assist flow of the mobile bitumen towards production well, asphaltene precipitation is only favourable when it happens far from production well. Deposition of asphaltene in reservoir results in an in-situ upgrading. Lighter solvents like propane result in higher vapour pressure and more asphaltene precipitation with extraction of lighter components. Butane precipitates less asphaltene and has a lower vapour pressure compared to propane. These phenomena can be optimized using proper design of solvent composition. Addition of light or heavy solvents to the base solvent can be a tool to control the aforementioned properties. For example, in the early stages of butane injection, addition of heavy solvent prevents the asphaltene precipitation around the wellbore. After depletion of the bitumen in the area near wellbore, addition of light solvent increases the amount of asphaltene precipitation and quality of the produced oil.

In this article, a detailed liquid–liquid equilibrium study of solvent and bitumen mixtures has been conducted. Butane/bitumen system was considered as a baseline and then the effect of adding propane, toluene and dimethyl ether on phase equilibrium of base mixture is investigated. Moreover, the improvements of adding fractions of secondary solvent on PVT properties of the butane/bitumen mixtures are discussed. The rest of this paper is organized as follows: first we present the experimental apparatus utilized to conduct the experiments. Then, the results and discussions will be presented followed by summary and conclusion.

2. Experimental apparatus

The schematic diagram of the apparatus is shown in Fig. 1. It consists of an equilibration cell (5), densitometer (7), viscometer (8), and sampling cells (9), which all are installed inside a temperature controlled Blue-M oven (1) with density and viscosity data evaluation units (2,3), a receiving cell (6), feeding cells (4) and Quizix pumps (10) placed outside of the oven.

The two feeding cells (4) can store bitumen and solvent at room temperature and desired pressure and can be operated by Quzix pump to inject bitumen and solvents into the equilibration cell (5). Pressures of the feeding cells (4), equilibration cells (5) and receiving cell (6) can be controlled by Quizix pumps (10). These pumps can inject or receive water with an accuracy of (±) 0.003 cm^3 to displace the piston located inside the cells and subsequently adjust the pressure. To avoid any contamination of the fluids with water pistons are sealed with Viton O-rings.

The equilibration cell (5) can rotate half a circle around an axis by means of a rocking system attached to the cell. This system along with the rocking ball placed in the cell accelerate the process of mixing of bitumen and solvent. Therefore, equilibration can be achieved faster. The capacity of the cell is about 850 cc, which allows to have sufficient saturated phases for the thermo-physical property measurements such as den-