

Thermal destabilisation of bitumen-in-water emulsions – A spinning drop tensiometry study

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ABSTRACT

Nonionic surfactant-stabilised oil-in-water emulsions offer a potentially useful vehicle for transporting heavy crude oils from oilfields to refineries or distribution terminals. Prior to refining, separation of the oil from the emulsion is necessary. Previous studies have suggested that heating the emulsion is sufficient for destabilisation and recovery of the oil. The present work examines this process on a batch laboratory scale and monitors the effect of thermal treatment on the heavy oil/water interfacial tension using spinning drop tensiometry. The present research has confirmed that heating Wolf Lake (Canada) bitumen-in-water emulsions to a temperature close to the cloud point of the surfactant results in efficient bitumen/water resolution, together with separation of a dense surfactant-rich coacervate phase that could be recycled in a commercial heavy oil transportation process. The corresponding temperature dependence of the bitumen/water interfacial tension provides further insight into the emulsion resolution process.

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1. Introduction

With the continuing re-evaluation of the world's petroleum resources, production of heavy crude oil (herein also referred to as natural bitumen) is becoming increasingly important, most notably in Canada and Venezuela. However, production and handling these materials, which have viscosities (at 25 °C) ranging typically from 1000 to 100,000 mPa s, introduces completely different challenges compared with lighter oils. Recovery methods necessarily involve reducing the viscosity of the oil in the reservoir, either by using steam to heat the production zone, as in cyclic steam stimulation or steam-assisted gravity drainage (SAGD) processes [1], or through controlled combustion of part of the bitumen deposit (e.g. *in situ* combustion) [2].

The produced and dehydrated bitumen is then diluted with available condensate to meet pipeline viscosity requirements for transportation to a refinery or export terminal. However, suitable diluents are often in limited supply and more innovative technical schemes for transporting viscous oils through pipelines have been proposed and investigated, most notably core-annular flow [3] and oil-in-water emulsions [4].

The present paper focuses on aspects of oil-in-water emulsion technology, as there has been a resurgence of activity in the development of emulsions for transportation in various heavy oil-producing regions, including Egypt [5,6], India [7], Iran [8] and Oman [9]. In

particular, it is concerned with the final stage of an emulsion transportation scheme, i.e. resolving the emulsion into its constituent bitumen and water phases. Of specific consideration is temperature-induced emulsion resolution and the corresponding effects on bitumen/water interfacial tension.

1.1. Heavy oil emulsion transportation

Fig. 1 shows a simple schematic of the emulsion transportation process [10]. Dehydrated, hot bitumen is emulsified with the required ratio of an aqueous surfactant solution in the mixing section, M, to produce a bitumen-in-water emulsion containing $\approx 70\%$ by volume bitumen, which typically has a viscosity of < 500 mPa s at a shear rate of ≈ 100 s⁻¹. The emulsion is allowed to cool and is stored until required for pipeline (or other means of) transportation. Upon reaching its destination, it is then necessary to resolve the emulsion.

The potential use of oil-in-water emulsions has been considered for the transportation of crude oils for approximately 40 years, and was initially applied to mobilising waxy crudes [11]. More recently, attention turned to the technical feasibility of producing heavy oil emulsions possessing the required stability and rheology for pipeline transportation. In fact, the feasibility has been thoroughly realised on a commercial scale.

The largest demonstrated production and export of bitumen-in-water emulsions was, until recently, that of Orimulsion. In this example, Venezuelan Cerro Negro bitumen ($\approx 9^\circ$ API) was emulsified in water, originally using an ethoxylated nonylphenol surfactant,

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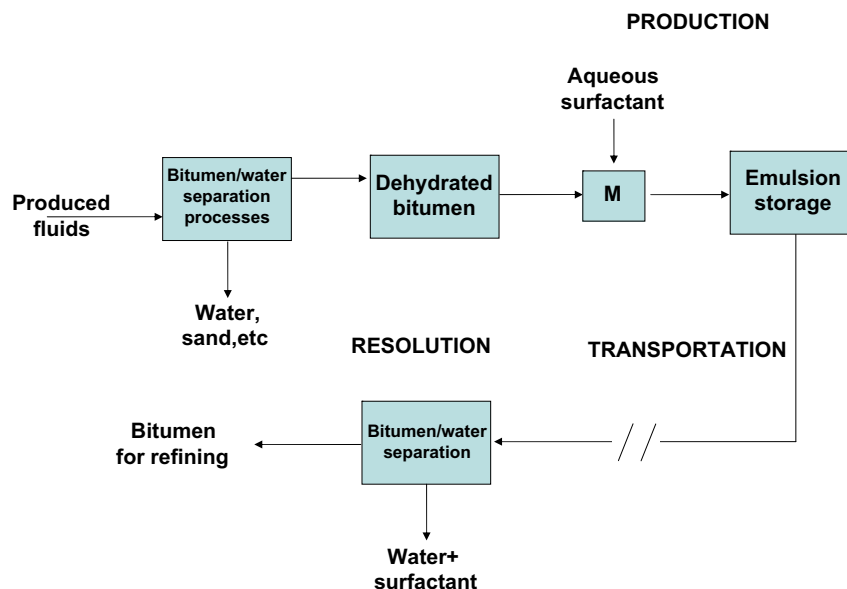


Fig. 1. Simplified emulsion transportation scheme as applicable to heavy oil/bitumen, highlighting the three main stages, production, transportation and resolution. M represents a mixing stage.

although this was subsequently changed on environmental and economic grounds [12]. Since 1990, some 27 million tonnes of Orimulsion has been produced and exported around the world [13]. This product was developed for use directly as a fuel in power stations, with no requirement to recover the bitumen for refining, unlike in transportation/refining schemes.

1.2. Heavy oil-in water emulsion resolution

To date, there has been relatively little work published on the resolution stage of the heavy oil emulsion transportation process. In 2004, Saniere et al. considered that the (emulsion) resolution stage presented a problem, since “such a process is not available” [14]. However, a Canadian trial conducted in 1982 outlined how a Wolf Lake bitumen emulsion, also stabilised with an ethoxylated nonylphenol surfactant, was effectively destabilised upon heating to 120–140 °C [1]. In another study [5], Zaki required a commercial demulsifier to resolve Geisum crude oil emulsions at 50 °C stabilised with an anionic surfactant. Grosmont crude oil emulsions stabilised by NaOH activation of latent crude oil surfactants were breakable upon re-acidification, but no further details were given [15].

Salager and co-workers [16,17] outlined the general requirements for an emulsion transportation process, including requirements for a thermal resolution stage. The proposed approach stresses the importance of formulation variables, in particular the surfactant hydrocarbon group and (for ethoxylated surfactants) the number of ethylene oxide units per molecule (EON); the alkane carbon number (ACN) of the oil phase; and the aqueous phase salinity. The “hydrophile–lipophile deviation” reflecting the solubility tendencies of the surfactant is expressed by the generalised equation [16]:

$$\text{HLD} = \alpha - (\text{EON}) - k(\text{ACN}) + bS + C_T(T - 25) \quad (1)$$

where α quantifies the surfactant lipophilic group, k , b and C_T are system-dependent constants, S is the salinity and T the temperature (in °C). When $\text{HLD} = 0$, the surfactant possesses equal affinity for each phase, leading to an “optimum condition” of minimum interfacial tension. This is achieved by varying (“scanning”) any of the formulation variables shown in Eq. (1) which, as the authors indicate, can be used to control emulsion properties such as

stability and droplet size. This treatment is based on the assumption that the surfactant will reside in either or both of the major phases within the range of conditions used during scanning the variables.

Clearly, the use of heat alone offers the most straightforward option for the resolution stage of any emulsion transportation process. Several emulsion transportation schemes have reported the use of ethoxylated nonionic surfactants as stabilisers, owing to their tolerance to the variable salinity of formation waters likely to be employed as emulsification media. The temperature-dependent phase behaviour of surfactants of this type is well known, such that low temperatures favour aqueous solubility, and consequent stabilisation of oil-in-water (o/w) emulsions. At a certain temperature, known as the cloud point (T_{cp}) of the surfactant, a surfactant-rich (coacervate) phase separates as a haze from aqueous solutions. For ethoxylated nonionic surfactants, T_{cp} increases with the EON and is also influenced (both positively and negatively) by the presence of hydrocarbons and other solubilises, as demonstrated in the classic work of Shinoda and Arai [18]. Oil-in-water emulsions stabilised with the same type of surfactant are destabilised at the phase inversion temperature (PIT), which is also influenced by the structure of the surfactant and the nature of the oil and can differ from the corresponding T_{cp} by up to 20 °C for *n*-alkanes [18]. T_{cp} and PIT are both dependent on the ionic composition of the aqueous phase as they are influenced by water structure-making and breaking tendencies and colloidal electric double layer interactions [19]. NaCl, used in the present study, would be expected to lower both T_{cp} and PIT [19].

Whether inversion (to a water-in-oil emulsion) or clean separation of the two phases occurs during the resolution process is not fully understood in the literature. Conceivably it involves both kinetic and thermodynamic factors related to the nature of the oil phase and the ability of the surfactant to stabilise the inverse water-in-oil (w/o) emulsions at high temperature. In this latter regard, demulsification of the w/o emulsions by phase-separated coacervate droplets may occur, as has been invoked for their action as antifoam agents [20].

1.3. Bitumen/water interfacial tension studies

By analogy with results from studies involving pure oils and surfactants [21,22], oil/water interfacial tension is a key indicator

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