

## Surface tension behavior of aqueous solutions of a propoxylated surfactant and interfacial tension behavior against a crude oil

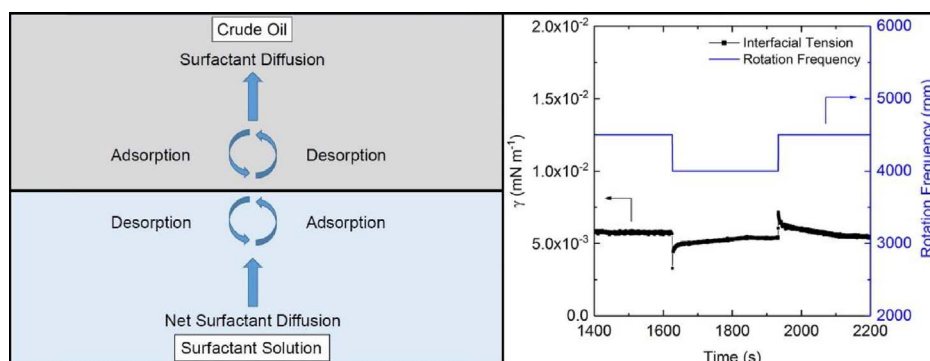


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### GRAPHICAL ABSTRACT



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### ABSTRACT

Equilibrium interfacial tensions (EIFTs) are important in determining the oil recovery efficiency in surfactant-based enhanced oil recovery (EOR) processes, in which ultralow IFT values (less than  $10^{-2}$  mN m<sup>-1</sup>) are often needed. The dynamic IFTs (DIFTs) for un-pre-equilibrated drops in contact with aqueous solutions and the adsorption mechanisms at the oil-water interface are important for the initial stages of EOR and for screening various surfactants for field applications. The IFT behavior of a commercial anionic surfactant, termed S13D, a single-chain propoxylated sodium sulfate salt has been studied. The synthetic brine used (9700 ppm total dissolved solids) was similar to that in an actual reservoir of oil, from which purified crude oil samples were prepared.

The dynamic and equilibrium surface tensions (DSTs and ESTs) and the IFTs were measured at 24 °C, with the emerging bubble/drop method, or the spinning bubble/drop method. The DSTs for surfactant concentrations from 0.1 to 10,000 ppm by weight have a simpler adsorption mechanism than the DIFTs, involving only diffusion from the aqueous phase and adsorption/desorption. Surface and interfacial tension relaxation tests after surface area perturbation were performed for establishing the validity of the ESTs and EIFTs. The critical micelle concentration (cmc) was 12 ppm in water and 1 ppm in brine. The lowest observed EIFT was 1 mN m<sup>-1</sup> with water and ultralow,  $2 \times 10^{-3}$  mN m<sup>-1</sup>, with brine. The equilibration timescales were shorter with brine than with water, for both DSTs and DIFTs, and slightly longer for DIFTs, because evidently the adsorbed surfactant desorbed and diffused away from the interface, for partitioning in the oil phase. The results suggest that the IFTs were associated with the typical adsorbed soluble monolayers at the oil-water interface, and that solubilization effects did not affect the measured DIFTs and EIFTs.

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**Nomenclature***Acronyms*

A	Surface area of a bubble or drop of volume V, mm <sup>2</sup>
$\bar{A}$	Average area per molecule, nm <sup>2</sup>
cmc	Critical micelle concentration, M or ppm
C <sub>surf</sub>	Surfactant concentration, M or ppm
DIFT	Dynamic interfacial tension, mN m <sup>-1</sup>
DST	Dynamic surface tension, mN m <sup>-1</sup>
EBM	Emerging bubble method
EDM	Emerging drop method
EIFT	Equilibrium interfacial tension, mN m <sup>-1</sup>
EOR	Enhanced oil recovery
EST	Equilibrium surface tension, mN m <sup>-1</sup>
f(L/R)	Correction factor, dimensionless
IFT	Interfacial tension, mN m <sup>-1</sup>
L	Length of the spinning bubble or drop, m
LY	Laplace-Young equation
n	Refractive index
R	Radius of the spinning bubble or drop at the center, m
S	Surface area of the drop or bubble of interest, m <sup>2</sup>

S13D	PETROSTEP <sup>®</sup> S-13D HA, commercial surfactant provided by Stepan Company
SBM	Spinning bubble method
SDM	Spinning drop method
SIFT	Steady-state interfacial tension, mN m <sup>-1</sup>
SST	Steady-state surface tension, mN m <sup>-1</sup>
ST	Surface tension, mN m <sup>-1</sup>
TX100	Triton <sup>™</sup> X-100, commercial nonionic surfactant
wt%A	Weight percent of active components of a surfactant

*Greek Symbols*

$\gamma$	Surface or interfacial tension, mN m <sup>-1</sup>
$\gamma_0$	Surface or interfacial tension of the pure solvent against air or oil, mN m <sup>-1</sup>
$\Gamma$	Surface density of adsorbed surfactant, mol cm <sup>-2</sup>
$\Delta\rho$	Density difference between two phases, kg m <sup>-3</sup>
$\nu$	Rotation frequency of a sample tube, s <sup>-1</sup> or rpm (rotations per minute)
$\rho$	Density of a phase, kg m <sup>-3</sup>
$\omega$	Circular frequency of a sample tube, $\omega = 2\pi\nu$ , s <sup>-1</sup>

**1. Introduction**

Low ( $< 1 \text{ mN m}^{-1}$ ) and ultralow ( $< 10^{-2} \text{ mN m}^{-1}$ ) interfacial tensions (IFTs) between an oil phase and an aqueous phase containing surfactants, polymers, and salts are quite important in emulsion stability and particularly in two-phase flows occurring in certain enhanced oil recovery (EOR) processes [1,2]. Because of practical economic considerations in chemical EOR processes involving the use of surfactants, it is important to achieve low IFTs at surfactant concentrations; that is, at concentrations which are lower than 10 wt% or even lower than 1 wt%. Moreover, the surfactants or surfactant blends under consideration need to be tailored to the salt concentrations (“salinity”), temperature, oil type, and other reservoir-specific conditions [3–7]. Currently, selecting a surfactant in practice is mostly empirical, and is often primarily based on a combination of apparent phase behavior tests and IFT data or estimates of the IFT values [6–14]. No firm guidelines on the relationships between the surfactant molecular structures and the IFT values have been reported in the open literature, except for some systems where pure hydrocarbons were used as model oils [15]. Thus, a critical need exists for developing methods for screening surfactants properly by using reliable equilibrium IFT (EIFT) values, and also, dynamic (time-dependent) IFTs (DIFTs).

The EIFT is generally considered to be the most important parameter that affects the flow of aqueous and oil phases in underground oil reservoirs, based on its role in the capillary number [7,9,11–14]. Nonetheless, some reported IFTs may not have been established to uniquely define the EIFT values. Then, correlating the EIFT values to other properties, such as to the molecular structure, the surfactant solubility in water or oil, or the solubilization of oil in water and water in oil, cannot be done rigorously. Moreover, IFT values are often reported after a giving period of time for a specific experiment, without due consideration of the actual timescale of equilibration. Therefore, determining which of the many dynamic IFT values is the EIFT value may be somewhat arbitrary, because the IFT depends on time. To ensure that a steady-state IFT value (SIFT) is indeed the EIFT value, one needs to know the timescale of equilibration. Similar arguments can be made for the surface tension (ST), dynamic surface tension (DST), steady-state surface tension (SST), and equilibrium surface tension (EST).

The equilibration timescales ( $t_{95}$ ,  $t_1$ , or  $t_2$ ) are defined in this article as the time required for the DIFT or DST value to change by 95% of the total change from the initial value to the steady-state value. Such

timescales can range from 1s to more than  $10^4$  s, and depend on the surfactant type, surfactant concentration, salinity, temperature and the type of oil [16,17]. Even though DIFTs have been observed and reported, their time dependence and relationship to the EIFT have received little attention in the EOR literature [18–24]. The aim here is to develop systematic protocols and molecular insights for addressing these issues in order to provide a methodology that will allow for appropriate screening of surfactants for a specific EOR or reservoirs. The results should lead to new insights on the fundamental physics associated with the interfacial tension in oil-water systems, and possibly afford researchers the ability to elucidate the mechanism or mechanisms involved in reaching ultralow IFTs.

For EOR applications, low and ultralow IFTs between a surfactant solution and a crude oil are generally measured only with the spinning drop method (SDM). This is because the current emerging-drop-based or pendant-drop-based instruments are incapable of measuring IFTs lower than about  $1 \text{ mN m}^{-1}$ . In order to establish whether a measured SST or SIFT is equal to the equilibrium value, one needs to test that the SST or the SIFT is stable and independent of area perturbations, as shown schematically in Fig. 1. For the emerging bubble method (EBM) and the emerging drop method (EDM), the interfacial area can be changed by abruptly varying the bubble or drop volume. For the spinning bubble method (SBM) or the spinning drop method (SDM), the area is changed by using the same bubble or drop volume and by varying the rotation frequency ( $\nu$ ) where the circular frequency ( $\omega$ ) is given by:  $\omega = 2\pi\nu$ . In this article, we present procedures for obtaining reliable SIFT and EIFT values from DIFT data for oil/aqueous solution systems with the EDM and, where possible, with the SDM.

We present ST and IFT results for a commercial propoxylated anionic surfactant, which has the capability of producing, against the crude oil studied here, low IFT values with water solutions and ultralow IFT values with the brine solutions used. We also report a limited ST study of a standard nonionic surfactant for reference and calibration purposes. Moreover, the solubility of the surfactant and the critical micelle concentrations (cmc) of the surfactant in various aqueous solutions were determined as no phase behavior information has been reported in the literature for this surfactant in water or a brine. The concentration dependence of the DST and of the EST for up to 10,000 ppm (1.0 wt% active, 1.2 wt% total) surfactant concentrations were determined. The  $t_{95}$  timescales and some  $t_1$  or  $t_2$  timescales of re-equilibration after area perturbation (see Fig. 1) were also determined.

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