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A correlation between surface, transport and thermo-elastic properties of liquid hydrocarbon: an experimental investigation

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Abstract

The temperature dependence of surface tension and viscosity has been investigated in two multi-component liquid hydrocarbons, namely, crude oil samples with different API numbers. The surface tension is found to decrease linearly with temperature whereas viscosity exhibits Arrhenius type variation. These measured values along with the ultrasound velocity, density and the isothermal compressibility have been used to estimate a number of physical parameters such as the activation energy, attenuation factor and the shear wave velocity. Crude oil with larger API was found to have smaller activation energy. Shear velocity decreases exponentially with increasing temperature while the attenuation factor is found to increase linearly with temperature. The ratio of the surface tension to viscosity varies linearly as the square root of temperature. The product of the surface tension and the isothermal compressibility, often characterized as a fundamental or correlation length of the surface of the liquid, was found to yield a constant value for both samples.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Basin modelling is an important part of petroleum exploration which is accomplished by assuming a temperature dependent visco-elastic lithosphere that behaves elastically on a short timescale and viscously on a long timescale. Viscosity and surface tension, along with other properties, can help us understand the migration of hydrocarbon from the source rock. Viscous and capillary forces, along with gravity, control the performance of a hydrocarbon reservoir. The viscous force counteracts the pressure differentials during the production of hydrocarbon from a reservoir. The capillary forces are due to the interfacial tension between the gas, oil and water phases, and the reservoir rock. They also act in retaining oil within the reservoir pore

space during production. Both the viscous and capillary forces contribute significantly in the control of the original distribution of fluids, displacement of one fluid by another within the reservoir and simultaneous movement of fluids through the reservoir. For enhanced recovery of the crude oil the basic mechanics of oil displacement by the injected fluid is controlled by the mobilities of the individual fluids which greatly depend on viscosity and surface tension.

This paper reports the temperature dependence of surface tension, $\sigma(T)$ and viscosity, $\mu(T)$ of two samples of crude oil procured from different wells of Oman. The surface tension and its temperature dependence were investigated using the pendant-drop method, making use of an interfacial tensiometer. The kinematic viscosity of the respective samples was determined using a Cannon–Fenske viscometer. The measured values of surface tension and viscosity along with the earlier experimental results [1, 2] of the ultrasound velocity, density and isothermal compressibility, were used to determine a number of physical properties such as the activation energy, attenuation factor, shear wave velocity and the characteristic fundamental length of the samples. We have also utilized the temperature dependent data of $\sigma(T)$ and $\mu(T)$ together with the compressibility values to establish the inter-relationships among these functions.

It may be recalled that March [3], by making use of the integral equations of surface tension [4] and the viscosity [5], showed that the ratio of surface tension to viscosity of liquids is directly proportional to the square root of temperature. Egelstaff and Widom [6] showed that the product of the surface tension and isothermal compressibility of liquids is equal to a few tenths of an angstrom unit. The product is termed as the fundamental length characteristic of a liquid. Some available data [3] for liquid metals near the melting temperature have testified such connections. Unfortunately, not much work has been done in other classes of multicomponent liquids where such connections can be established over a wide range of temperatures. Our measurements of surface tension, viscosity, ultrasound velocity and density over a wide range of temperature verify and establish the inter-connection between various thermo-physical properties in multicomponent liquids, such as liquid hydrocarbons.

The temperature dependence of surface tension and viscosity and the details of the measurements are presented in sections 2 and 3, respectively. These data are further used to estimate the activation energy (section 4). The ratio of surface tension to viscosity is presented in section 5 whereas the product of isothermal compressibility and surface tension is discussed in section 6. The computed values of shear wave velocity and attenuation factor are given in section 7, which is followed by summary and conclusion in section 8.

2. Experimental determination of surface tension, $\sigma(T)$

Two different crude oil samples were procured from the oil fields of Saih Rawl and Ghafeer located in the Sultanate of Oman. Hereafter we refer to the two samples as Sample I (Saih Rawl) and Sample II (Ghafeer). The major ingredients of crude oil are aliphatic, alicyclic and aromatic hydrocarbons covering carbon numbers which may vary from C_1 to C_{60} . The oil also contains sulfur, oxygen and nitrogen compounds in small percentage (<1%). The chemical composition of crude oil may vary from different producing regions and hence we have used an accepted index number called API (American Petroleum Institute oil gravity number) to classify our samples,

$$\text{API} = (141.5/\rho) - 131.5 \quad (1)$$

where ρ is the density (g cm^{-3}) measured at 15.6°C and at atmospheric pressure. Our measured density leads to API numbers for these samples as

$$\text{Sample I: API} = 31.52$$

$$\text{Sample II: API} = 34.39.$$

Table 1. Measured viscosity and surface tension, and the calculated shear wave velocity and attenuation parameter.

Temp (K)	Sample I (API = 31.52)				Sample II (API = 34.39)			
	Surface tension, $\sigma \times 10^{-3}$ (N m ⁻¹)	Viscosity $\mu \times 10^{-3}$ (Pa s)	Shear velocity v_s (m s ⁻¹)	Attenuation parameter $k \times 10^4$ (m ⁻¹)	Surface tension $\sigma \times 10^{-3}$ (N m ⁻¹)	Viscosity $\mu \times 10^{-3}$ (Pa s)	Shear velocity v_s (m s ⁻¹)	Attenuation parameter $k \times 10^4$ (m ⁻¹)
293.15	22.9	37.6	7.39	849.6	22.4	15.4	4.8	4165.2
298.15	22.5	28.9	6.49	968.4	22.0	12.6	4.3	4600.7
303.15	22.1	23.0	5.81	1082.1	21.7	10.5	4.0	5031.2
308.15	21.6	18.9	5.27	1192.4	21.3	9.0	3.7	5414.1
313.15	21.2	15.7	4.82	1303.8	21.0	7.8	3.4	5800.0
318.15	20.8	13.5	4.48	1404.0	20.6	6.9	3.2	6146.9
323.15	20.4	11.3	4.10	1531.4	20.2	6.1	3.0	6529.1
328.15	19.9	9.8	3.83	1639.1	19.9	5.4	2.9	6920.5
333.15	19.5	8.6	3.58	1754.3	19.5	4.9	2.7	7296.5
338.15	19.1	7.5	3.37	1867.5	19.2	4.4	2.6	7693.4
343.15	18.7	6.6	3.16	1989.4	18.8	3.9	2.5	8074.2

The surface tension of the oil samples at various temperatures was measured using the pendant-drop technique. The measurements were made using an OCAH 200 tensiometer from Dataphysics equipped with a temperature controlled specimen chamber. The temperature of the air inside the specimen chamber is controlled to within ± 0.1 °C using a thermocouple probe placed at the centre of the chamber. The drop of the specimen was formed at the tip of the needle of an electronically controlled dispensing syringe. The contour of the drop was captured by a high-speed camera. The diameter of the needle, used as a calibration reference by the contour analysing software, was known with an accuracy of $\pm 10^{-5}$ m. Each measurement was repeated 100 times in rapid succession and the final value of the surface tension at each temperature was obtained from statistical analysis of the data.

The analysis of the contour of the drop was made using the Young–Laplace equation. This equation expresses the balance between the surface tension and the gravitational force on the drop. The software therefore requires the accurate temperature dependence of the density of the specimen and the gravitational acceleration at the location of the experiment to extract the surface tension from the contour of the drop numerically. The temperature dependent density $\rho(T)$ of the crude oil samples was measured using an Anton Paar density meter (DMA 5000) with an accuracy of $\pm 10^{-3}$ kg m⁻³,

$$\rho(T) = \rho_r + \Lambda_\rho(T - T_r) \quad (2)$$

where ρ_r is the density at room temperature T_r and at atmospheric pressure, the values of which are 864.825 kg m⁻³ for Sample I (API = 31.52) and 849.843 kg m⁻³ for Sample II (API = 34.39). $\Lambda_\rho = -(\mathrm{d}\rho/\mathrm{d}T)$ is the temperature gradient, which for Sample I and Sample II respectively stand as -0.667 kg m⁻³ K⁻¹ and -0.685 kg m⁻³ K⁻¹.

The measured surface tension, σ (N m⁻¹), of the two samples is given in table 1. The plots of σ versus T (K) in figure 1 exhibit a linear dependence of σ on T for both the samples. σ decreases linearly with T and can reasonably be represented by the relations

$$\sigma = 0.0478 - 8.47 \times 10^{-5}T \quad \text{for API} = 31.52 \text{ (Sample I)} \quad (3a)$$

$$\sigma = 0.0434 - 7.18 \times 10^{-5}T \quad \text{for API} = 34.39 \text{ (Sample II)}. \quad (3b)$$

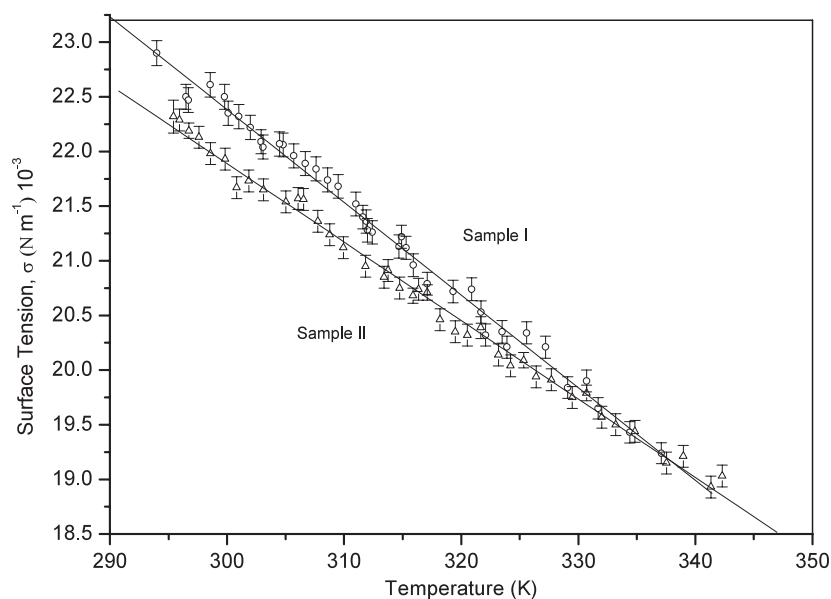


Figure 1. Variation of surface tension (σ) with temperature (T) for Samples I (API = 31.52) and II (API = 34.39).

The regression line fits the data adequately as indicated by the high value of the coefficient of determination R^2 (0.9918 for Sample I and 0.9933 for Sample II). In addition, the estimated parameters are statistically significant with p -values less than 10^{-4} . The values of σ of the two samples are very close to each other. Near room temperature they differ by about 2%, but at high temperature ($T = 343.15$ K) these values are very close to each other. It seems that the surface tension values are not very sensitive to API values as compared to viscosity values (see section 3). The absolute values of surface tension of crude oils are closer to those of other organic liquids dominant in carbon (C) and hydrogen (H) compositions. For example [7], σ for n -hexane and n -octane are found to be respectively 18.4×10^{-3} N m $^{-1}$ and 21.8×10^{-3} N m $^{-1}$ at 293.15 K. It may be noted that σ values for hydrocarbon liquids are much smaller than that of water, which is about 72.8×10^{-3} N m $^{-1}$ at 293.15 K.

The measured values of the temperature coefficient of surface tension, $(d\sigma/dT)$, i.e., -8.47×10^{-5} N m $^{-1}$ K $^{-1}$ for sample I and -7.18×10^{-5} N m $^{-1}$ K $^{-1}$ for sample II, are negative, as expected theoretically. The values of the coefficients are very much in the range of the values found [8, 9] for a wider class of liquid metals which are very well experimentally investigated. The coefficients for a large number of liquid metals are found to vary from -6×10^{-5} N m $^{-1}$ K $^{-1}$ to -50×10^{-5} N m $^{-1}$ K $^{-1}$. The σ values for such liquids are also found to be linear with temperature. Our observation of the linear dependence of the surface tension of crude oil on temperature is quite in agreement with the Eötvös law (see [9]), which can be expressed as

$$\sigma = \frac{B}{V^{2/3}}(T_C - T) \quad (4)$$

where B is a constant and V is the molar volume. At the critical temperature T_C , the distinction between liquid phase and gas phase disappears and hence the surface tension is reduced to zero. As a consequence, the surface tension of liquids must decrease with rising temperature. A discussion of the surface tension of crude oil with respect to its critical temperature will be presented in later work.

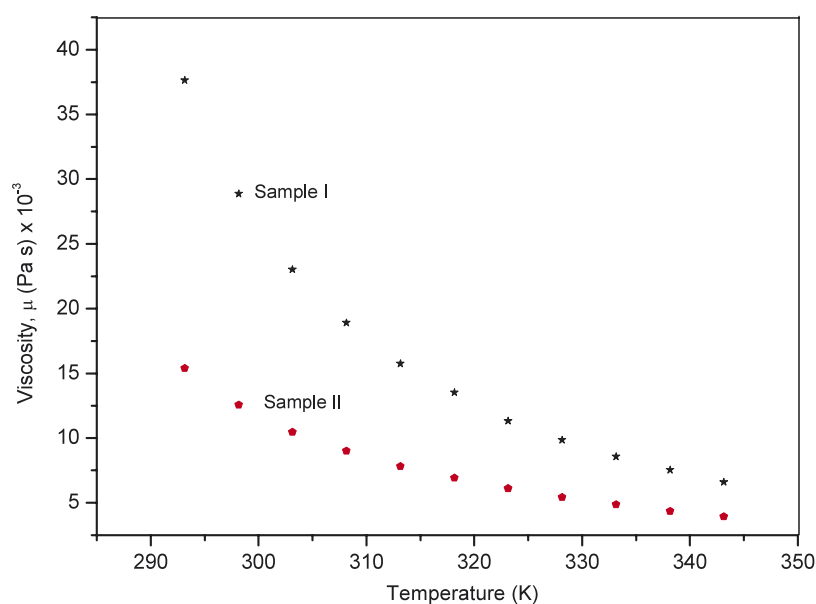


Figure 2. Variation of viscosity (μ) with temperature (T) for the two crude oil samples.

3. Experimental determination of viscosity, $\mu(T)$

The kinematic viscosity of the crude oil samples was determined using a Cannon–Fenske viscometer. The working of the viscometer is based on the principle that the average velocity of steady flow in a round tube depends inversely on viscosity. The viscometer determines the kinematic viscosity by timing the fluid flow through a capillary tube as it passes between two etched lines on the glass wall. In our experiment the viscometer was inserted into a constant temperature bath whose temperature was controlled to ± 0.1 °C using a Haake D8 circulation thermostat. To measure the efflux time, the thermostat was set at a desired temperature and the sample liquid was allowed to fall freely down past an upper mark measuring the time for the meniscus to pass the lower mark. The kinematic viscosity (μ') was determined by multiplying the measured transit time of the fluid column in seconds with the calibration constant. The error of the viscosity measurement is less than 0.35%. Finally the dynamic viscosity (μ) or simply the viscosity is obtained from the relation

$$\mu = \mu' \rho. \quad (5)$$

The density ρ in kg m^{-3} at a given temperature is determined from equation (2). The measured viscosities (Pa s) of the two samples are given in table 1. A plot of μ versus T is shown in figure 2. The error in the measurement of viscosity is too small to be shown in the plot. μ decreases exponentially as the temperature increases. A contrasting feature is the large difference in the values of the viscosity of the two samples. The viscosity of Sample I (API = 31.52) is about 145% higher than that of Sample II (API = 34.39) at room temperature, $T = 293.15$ K. This difference decreases to 67% when the temperature is raised to 343.15 K. We may recall that the two samples differ only by about 9% in their API values but this affects μ strongly. μ is found to be very sensitive to API values at low temperatures. Also the values of μ for crude oil samples are very large in comparison with water ($\mu_w = 1.002 \times 10^{-3}$ Pa s) at $T = 293.15$ K.

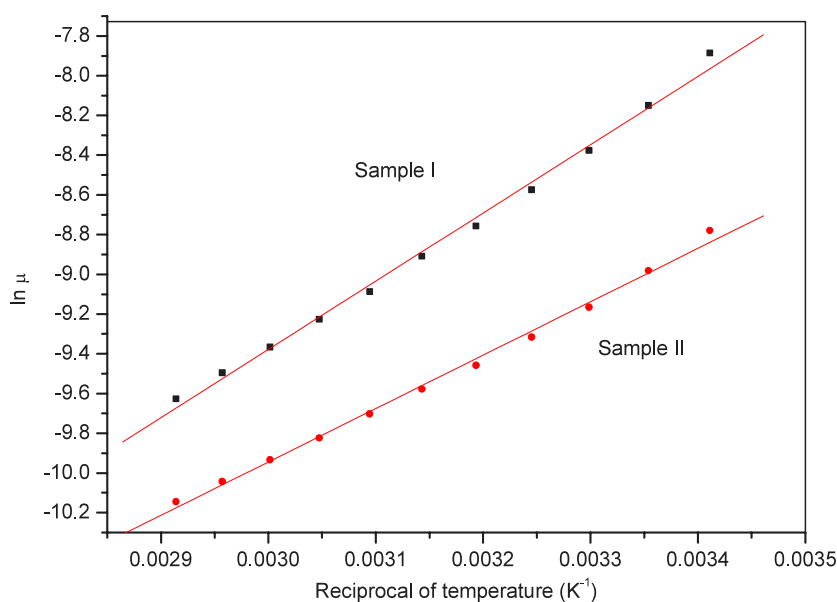


Figure 3. $\ln \mu$ versus reciprocal of temperature (T).

4. Activation energy

The processing of crude oil, in a major way, depends on chemical reaction. The activation energy is a measure of the minimum energy necessary for a specific chemical reaction to occur. The repulsive energy emerging due to overlap of electron clouds needs to be overcome by the heat of the system. The Arrhenius type equation gives the quantitative basis for the evaluation of activation energy. The activation energy (H_μ) and the viscosity (μ) are related as

$$\mu = A \exp\left(\frac{H_\mu}{RT}\right) \quad (6)$$

where A is a constant and R is the gas constant. Equation (6) has been very successful in explaining the temperature dependence of viscosity and hence in the determination of activation energy. Equation (6) was further improved [10] to include the free volume effect as suggested by Cohen and Turnbull [11]. They emphasized that for the viscosity of liquids, two events must simultaneously occur before a molecule can undergo a diffusive jump: (i) the molecule must attain sufficient energy to break the existing bond, and then (ii) there must be an empty site to accommodate the diffusing atom. The activation energy (H_μ) obtained from equation (6) involves both of these effects. However, the contribution due to breaking of bonds is likely to be higher than the energy of the free volume.

We have used our experimentally determined values of μ as a function of T in equation (6) to determine H_μ . Taking the natural logarithm of equation (6), one has

$$\ln \mu = \ln A + \left(\frac{H_\mu}{R}\right) \frac{1}{T}. \quad (7)$$

A plot of $\ln \mu$ against T^{-1} , shown in figure 3, yields straight line behaviour for both the samples. The goodness of the fit is demonstrated through the high value of the coefficient of determination R^2 (0.9952 for Sample I and 0.9959 for sample II). The p -values in both cases

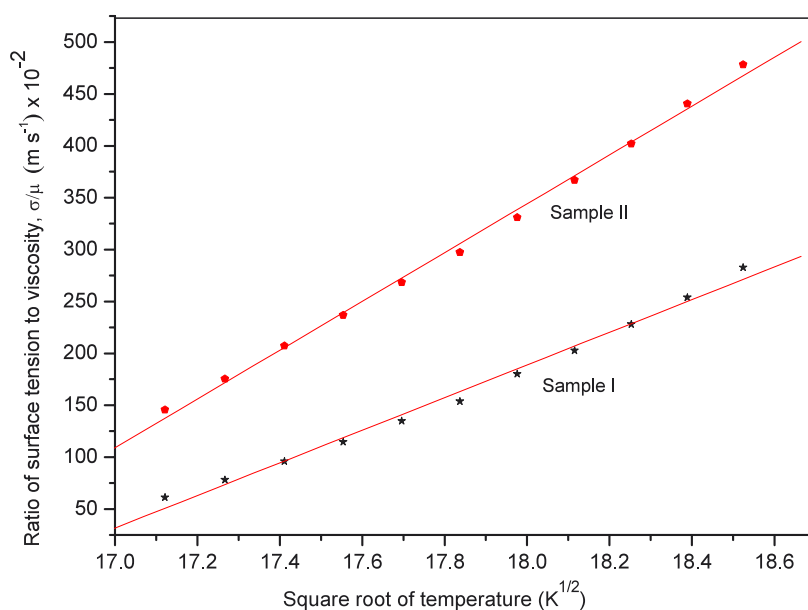


Figure 4. Ratio of surface tension to viscosity (σ/μ) versus square root of temperature ($T^{1/2}$).

are less than 10^{-4} . The activation energies determined from the slopes are

$$H_{\mu} = (28.6 \pm 0.7) \text{ kJ mol}^{-1} \text{ (Sample I, API} = 31.52)$$

$$H_{\mu} = (22.6 \pm 0.5) \text{ kJ mol}^{-1} \text{ (Sample II, API} = 34.39).$$

The two samples differ in their activation energy values by about 28%. Larger API has resulted in smaller activation energy.

5. Ratio of surface tension to viscosity

The striking similarity between the integral expressions of surface tension [4] and viscosity [5] indicates that the two physical properties are related. March [3] has shown that the surface tension and the viscosity of a liquid are related to each other through its atomic mass (M), Boltzmann's constant (k_B) and the absolute temperature (T) by the equation

$$\frac{\sigma}{\mu} \left(\frac{M}{k_B T} \right)^{1/2} = \text{constant.} \quad (8)$$

For a given liquid, one has

$$\frac{\sigma}{\mu} \propto T^{1/2}. \quad (9)$$

The ratio σ/μ against square root of the temperature for the two liquids is plotted in figure 4. Both samples exhibit a linear relationship between σ/μ and $T^{1/2}$. The gradient for Sample II with higher API is slightly larger than that of Sample I with lower API.

Often the factor $(k_B T/M)^{1/2}$ in equation (8) is interpreted as the thermal velocity of atoms in liquids. March [3] opined that one should consider replacing this factor, i.e., the thermal velocity, by a velocity reflecting the collective behaviour which may be the velocity of sound (v_l). Since the velocity of sound in the same crude oil samples is known (1406.8 and

1391.9 m s^{-1}) from our previous measurements [2], the constant of equation (7) is determined as 4.3×10^{-4} for Sample I and 0.1×10^{-4} for Sample II at room temperature. Obviously, the values of $\sigma/(\mu v_l)$ are quite different for the two samples at room temperature. The constant is further found to depend on temperature. This needs to be further investigated by taking examples of a wider class of other liquids.

6. Product of isothermal compressibility and surface tension

Several researchers [12–14] have investigated the physical significance of the product of isothermal compressibility (χ) and surface tension (σ) of liquids. According to the fluctuation theory (see [4]), the product of χ and σ determines the effective surface thickness (l),

$$\chi\sigma = l. \quad (10)$$

Equation (10) essentially characterizes the density profile of the liquid by a single parameter l . We have used the values of σ and experimentally [1] determined χ to determine l , which is found to be around 0.17 \AA for both the samples and almost remains independent of temperature in the range investigated. Equation (10) was successfully applied to determine [15] l for liquid metals near the melting temperature; it varies from 0.15 to 0.48 \AA .

Further, Egelstaff and Widom [6] showed that the product of χ and σ of a liquid near its triple point may be perceived to be a fundamental length (L) characteristic of the liquid,

$$\chi\sigma = 0.07L. \quad (11)$$

L is often interpreted as the correlation length. Near the critical point the deviation of this physical property from its value in the bulk phase vanishes proportionally to the damping factor $\exp(-|z|/L)$, where z is the distance from the interface. Therefore $(\chi\sigma/0.07)$ provides a rough measure of the thickness of the surface layer. For both samples, equation (11) suggests $L \approx 2.4 \text{ \AA}$. From experimental data on 30 liquids (organic liquids, simple non-metallic liquids, water, molten salts and liquid metals), it was shown [6] that L varies in the range $2\text{--}6 \text{ \AA}$. Obviously crude oil samples fall in the category which are endowed with a thin surface layer.

7. Shear wave velocity and attenuation parameter

The propagation of ultrasound waves in real fluids involves a specialized solution of the Navier–Stokes equation where not only longitudinal but also shear waves may propagate through a viscous medium. Equations for the velocity of shear waves (v_s) and the attenuation factor (k) have been derived [16] in terms of viscosity, density and angular frequency, which may be written respectively

$$v_s = \left(\frac{2\mu\omega}{\rho} \right)^{1/2} \quad (12)$$

and,

$$k = \left(\frac{\omega\rho}{2\mu} \right)^{1/2} \quad (13)$$

where v_s is the shear wave velocity in cm s^{-1} , ω is the angular frequency in rad s^{-1} , ρ is the density in g cm^{-3} , μ is the viscosity in poise and k is the attenuation factor in cm^{-1} . It is evident from equation (12) that shear wave propagation, unlike longitudinal wave propagation, depends on the frequency (ω). Besides the Navier–Stokes approach, one can also determine the propagation velocity from the dispersion (ω versus q , where q is the wavenumber)

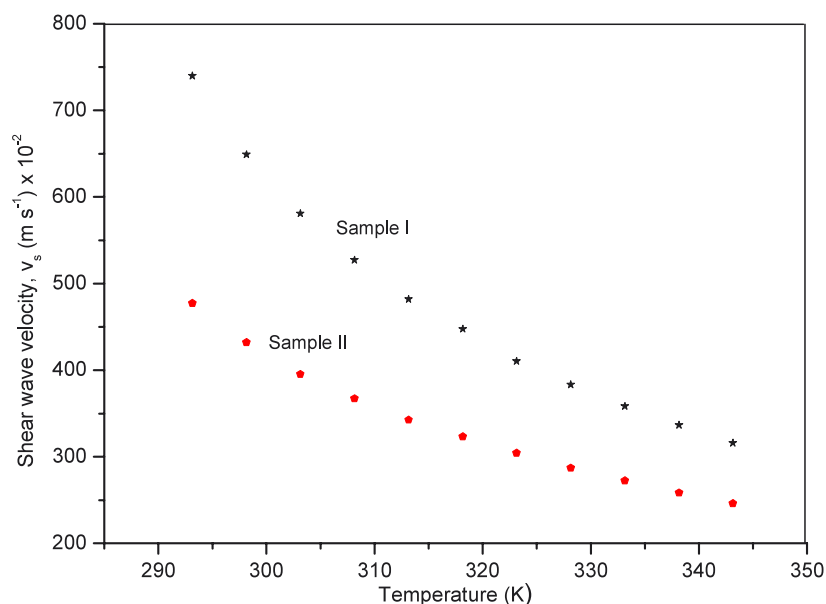


Figure 5. Shear wave velocity (v_s) versus temperature (T).

curves for longitudinal and transverse phonons. Bhatia and Singh [17] showed that for pairwise interactions one has $v_s \propto q$, or $v_s \propto \sqrt{\omega_s}$, where ω_s is the frequency of the shear waves. Viscous waves are highly damped due to relaxation effects. Usually the frequencies of transverse phonons are quite smaller than those of longitudinal phonons. For smaller wavevector (q), few or no excitations exist for transverse phonons. We have used our values of μ and ρ in equations (12) and (13) to determine the shear wave velocity (v_s) and attenuation factor (k) for a typical shear wave of frequency $\approx 10^5$ Hz. The computed values of v_s and k are given in table 1. Their variation with temperature is shown in figures 5 and 6 respectively. At room temperature, values of v_s and k are

$$v_s = 7.39 \text{ m s}^{-1} \quad \text{and} \quad k = 8.50 \times 10^4 \text{ m}^{-1} \quad \text{for Sample I}$$

$$v_s = 4.77 \text{ m s}^{-1} \quad \text{and} \quad k = 13.17 \times 10^4 \text{ m}^{-1} \quad \text{for Sample II.}$$

Under similar conditions of room temperature and atmospheric pressure, the shear wave velocity of crude oil is quite large in comparison to that of water, $v_s(\text{water}) = 1.1 \text{ m s}^{-1}$. However, the values of attenuation factor for the crude oil samples are much smaller than that of water ($56 \times 10^4 \text{ m}^{-1}$).

The shear wave velocity plotted against temperature in figure 5 indicates that it decreases exponentially with increasing temperature. The gradient for sample I (lower API value) is quite large near room temperature in comparison to that of sample II (higher API value). As the temperature increases, the gradients flatten out significantly. But the attenuation factor (figure 6) increases linearly with temperature for both samples.

8. Summary and conclusion

Surface tension and viscosity for two crude oil samples (API = 31.52 and 34.39) were determined experimentally as a function of temperature ranging from room temperature up to

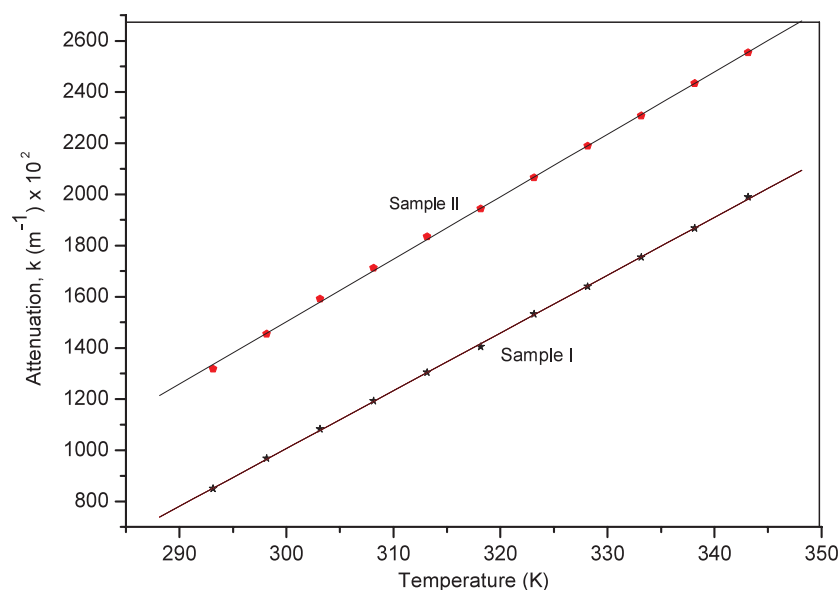


Figure 6. Attenuation parameter (k) versus temperature (T).

343.15 K. Hence we could investigate the connection between surface, transport and thermo-elastic properties over a wide range of temperature in a multicomponent liquid hydrocarbon. The surface tension is found to decrease linearly with temperature whereas the viscosity exhibits Arrhenius type variation. This has been used to estimate the activation energy; the values for the two samples differ by about 28%. Larger API has resulted in smaller activation energy.

The temperature dependent measured values of viscosity and density are further used to evaluate the shear wave velocity and the attenuation factor following simple visco-elastic equations. The shear velocity decreases exponentially with increasing temperature but the attenuation factor is found to increase linearly with temperature.

The ratio ($\frac{\sigma}{\mu}$) of surface tension and viscosity for both the samples is found to vary as the square root of temperature. The product ($\chi\sigma$) of the isothermal compressibility and the surface tension, which is often characterized as a fundamental or correlation length of the surface of a liquid, yields a constant value of about 2.4 Å for both crude oil samples and remains almost independent of temperature.

The results suggest that a good correlation exists between surface, transport and the thermo-elastic properties of liquid hydrocarbons.

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