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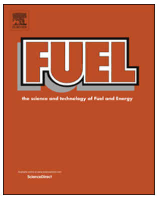

Full Length Article

Ultrasonic shear wave reflectometry applied to the determination of the shear moduli and viscosity of a viscoelastic bitumen

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ABSTRACT

Ultrasonic shear wave reflectometry is widely used in process control to measure the dynamic shear viscosities of Newtonian liquids. We apply the technique to study the temperature dependence of produced bitumen: a non-Newtonian ultra-heavy 6° API hydrocarbon with a room temperature viscosity of $\sim 10^3$ Pa·s. The experimental apparatus employs a delay line made of polyetheretherketone (PEEK) whose shear impedance more closely matches that of the bitumen allowing for greater sensitivity but at the cost of long equilibration wait times. The temperature dependent values of the complex shear modulus and consequent estimates of the steady flow shear viscosity both follow close to an Arrhenius-like behaviour over the range of 10 °C–50 °C, but there are significant discrepancies between the ultrasonic and more conventional spindle viscosities. These may reflect differences between the particle displacements, the strains, and the strain rates associated with each measurement technique. It is in the nature of bitumen to shear thin at high shear rate using ultrasonic technique. Regardless, these measurements do show that the shear wave reflectometry does provide information on the changes in the viscoelastic bitumen with temperature that may be useful in for purposes of its monitoring during production and processing.

1. Introduction

In a Newtonian fluid, the shear stress is linearly proportional to the strain rate via the proportionality constant of the dynamic viscosity. Many low molecular weight liquids and gases, such a molten metals and inorganic salts, and organic and inorganic liquids and gases, exhibit Newtonian viscous behavior [1]. In contrast, the stress observed in most foams, emulsions, colloidal suspensions and polymeric solutions depend nonlinearly on strain rate; and these are known as non-Newtonian fluids. The apparent viscosity in these depends on the strain being manifest as the phenomena of shear thinning (apparent viscosity decreases with shear rate) or shear thickening (apparent viscosity increases with shear rate).

Bitumen is the generic term used by the American Petroleum Institute to collectively describe ultra-heavy hydrocarbon oils with densities higher than 1000 kg/m^3 and dynamic shear viscosities at the in situ temperature (11 °C) as high as $\sim 10^4$ Pa·s [2]. The extreme viscosities of bitumen make economic extraction difficult as it cannot flow easily through permeable rock or pipelines. The bitumen used in this study, for example, is natural sample produced from the Grosmont Formation of northern Alberta, Canada where thermal recovery processes are used to reduce the natural oil's viscosity allowing it to flow. Understanding the rheological behavior of these oils and how it changes with temperature [3–10] and pressure [11,12] is key to the design of efficient extraction processes. Therefore, having advance knowledge of these changes in viscosity is of practical importance in the design of efficient recovery techniques, among other needs.

Knowledge of bitumen's rheological behavior, too, is necessary to properly interpret surface time-lapse geophysical data [13,14] where the changes in seismic wave speeds will for the most part depend on the fluid properties [15]. When considering seismic wave propagation at sufficiently low temperatures ($< \sim 50$ °C), the bitumen may have a nonnegligible shear modulus [16] that will contribute to the overall frequency-dependent rock elastic properties [17]. This information could be vital to the proper interpretation of the angle of incidence variations in seismic reflectivity in attempts to measure in situ attenuation directed towards pore fluid identification e.g., [18–20] although a great deal of care must be taken to properly define this problem e.g., [21–23]. Consequently, our main motivation for this work stems from a need for values of the complex shear modulus G^*. That said, such information too may be applied to study the bitumen's rheology at high frequencies.

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Reliable determination of bitumen viscosity alone remains problematic for many reasons. Round-robin blind measurements from differing laboratories on carefully collected and presumably uniform samples of the same bitumen have given values differing by more than a factor of two from 36 Pa·s to 72 Pa·s at 20 °C [24,25]. Considering only shear viscosity, however, likely masks more complex rheological behavior.

Bazyleva et al. [6] emphasized three critical issues on the viscosity data for such complex multicomponent and multiphase fluid bitumen: the geographical origin of the sample, the environment of the experiments, and the awareness of the limitation or associated error of the techniques. They noted that shear rate has the most substantial impact on the rheological measurements. The complex viscosity of non-Newtonian Athabasca bitumen at the low-temperature is reported to vary as much as three orders or magnitude with shear rate. Besides, bitumen tends to have a broad range of relaxation spectrum due to the presence of various sizes of molecules [10,26,27]. Hence, the relaxation time, the ratio of viscosity and shear modulus (real) of the medium at high frequency, also depends on the temperature [28].

These issues highlight the need to better understand the mechanical behavior of bitumen and to estimate viscosity and shear modulus. Towards this end, we describe the further development of an ultrasonic shear wave reflectometer. This method, first described by Mason et al., [29], has been used quantitatively and qualitatively to measure the dynamic shear properties of a variety of complex fluids with applications to the setting of concrete [30] and asphalt [31], foodstuffs such as honey [32] and bread doughs [33], polymers [34], silicate melt [35], and various other compounds [36–41] including hydrocarbon oils [42] [43]. Aside from Han et al.,'s [16,44,45] measurements focused on the estimation of a shear wave speed, to our knowledge this technique has not been applied to bitumen although there have recently been attempts to model similar responses using vegetable oil shortening with compressional wave reflectivity [46].

This paper begins with a brief overview of the theory of extracting shear properties from the complex shear wave reflection coefficient. We describe the construction of an ultrasonic shear wave reflectometer and its testing at modest temperatures using well known fluids. This is followed a detailed description of our bitumen sample and measurements that incorporate a novel signal processing scheme. We interpret the observed complex shear wave reflection coefficients and comment on the meaning of the measurements given that the analysis relies on the assumed rheological behaviour of the material.

2. Theory

Our initial goal of our measurements is to obtain values of the complex shear modulus G^* that may be applied to modelling of wave speeds in rocks saturated with bitumen. This data, however, can also constrain the rheological behaviour of bitumen. This is accomplished in two stages by first experimentally determining the fluid's shear modulus G^* that, second, must then be interpreted using an appropriate rheological model. Here only simple models are discussed given the preliminary nature of this study the intent of which is to develop the methodology.

2.1. Physical property relationships

In general, the shear properties of any material of known density ρ may be described equivalently by the complex shear modulus G^*, velocity c^*, the viscosity η^*, or impedance Z^*. Here we will employ measurements of Z^* to provide information on the other parameters; and it is instructive to review these relationships that are summarized in Table 1.

The shear modulus for any material may be written as a complex number $G^* = G' + iG''$ where G' and G'' are real numbers respectively referred to as the storage and loss moduli. Their values are generally frequency dependent. They are constants only for the bounding cases of a perfect Hookean elastic solid with $G'' \equiv 0$ on one hand and a Newtonian fluid with $G' \equiv 0$ on the other. In the following we implicitly assume that all parameters are frequency dependent. Following Dixon et al. [47], if the material has density ρ, one can similarly describe a complex shear phase velocity $c^* = c' + ic''$ and the relationship

$$G' + iG'' = \rho[(c'^2 - c''^2) + 2ic'c''] \tag{1}$$

Alternatively, the phase velocity $c^* = \omega/k^*$ where $k^* = k' + ik''$ is the complex wavenumber with

$$k' = \frac{\omega c'}{c'^2 + c''^2} \quad \text{and} \quad k'' = \frac{-\omega c''}{c'^2 + c''^2} \tag{2}$$

through solution of the wave equation allows the wave displacement amplitude $A(x,t)$ to be described through its propagation

$$A(x, t) = A_o \exp(-k''x)\exp(i(k'x - \omega t)) \tag{3}$$

where A_o is the initial amplitude, that will have a measurable wave speed $v = \omega/k'$ and attenuation k''. The subsequent concept of the skin or penetration depth $\delta = 1/k''$ that is in studies of electromagnetic wave propagation a measure of the distance from the surface of the material at which the amplitude has decayed to $A(\delta,t)/A_o = 1/e$ (~36.79%) and the intensity to $1/e^2$ (~13.53%). The quality factor $Q = G'/G''$ is a normalized measure of the wave's attenuation that is also often useful [49].

In principle, G^* could be determined by direct measurement of wave speed and attenuation (e.g., Barlow and Subramanian, [50]) but due to difficulties in propagating shear waves into fluids this is usually not practical. To overcome this difficulty, it is important to note that a material's mechanical properties, independent of any assumed rheology, may also be expressed in terms of its complex impedance $Z^* = \rho c^* = R + iX$, where R and X are respectively the shear resistance and reactance in mechanical Ohm units of $N{\cdot}s/m^3$, ρ is the density and c^* is the complex phase velocity. The complex shear impedance is related to the complex shear modulus as,

$$Z^* = (\rho G^*)^{1/2} \tag{4}$$

or rearranging

$$G^* = \frac{(Z^*)^2}{\rho} = (R + iX)^2/\rho \tag{5}$$

Hence, in principle G^* may be determined from knowledge of the density and complex impedance with real

$$G'(\omega) = \frac{R^2 - X^2}{\rho} \tag{6}$$

and imaginary components

$$G''(\omega) = \frac{2RX}{\rho} \tag{7}$$

with these equations being applicable for any rheology [50]. These relations are exploited using shear wave reflectometry as described below.

Alternatively, the material can be considered in terms of its complex viscosity $\eta^* = \eta' + i\eta''$ [51,52] with the frequency dependent dynamic viscosity $\eta' = G''/\omega$, which collapses to the static or steady-flow viscosity η at the lowest frequencies, and an elastic component $\eta'' = G'/\omega$ with

$$|\eta^*| = \frac{|G^*|}{\omega} \tag{8}$$

Exploration of these relationships for a simple Maxwell rheology discussed later but constructed with an instantaneous shear modulus G_∞ and steady flow viscosity η illustrates the expected changes in the components of G^* and η' with increasing normalized frequency (Fig. 1a) and in the mechanical shear resistance R and reactance X (Fig. 1b)