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Viscosity and density measurements on liquid n-tetradecane at moderately high pressures

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ABSTRACT

The main aim of the work is to study the viscosity and density of compressed normal tetradecane in the region of pressures from saturation to 10 MPa, where the available literature data are scarce. New measurements of the viscosity of n-tetradecane (n-C_{14}) along eight isotherms in the range (283–358) K and at pressures up to 70 MPa, have been performed using the vibrating wire technique in the forced mode of operation. Density measurements have also been performed along nine isotherms in the temperature range from (283 to 373) K and pressures from (0.1 to 70) MPa. The vibrating wire viscosity results were correlated with density, using a modified hard-spheres scheme. The root mean square (rms) deviation of the data from the correlation is less than 0.32% and the maximum absolute relative deviation is less than 1.0%. The expanded uncertainty of the present viscosity data is estimated as ±1.5% at a 95% confidence level. The density results were correlated with the temperature and pressure using a modified Tait equation. The expanded uncertainty of the present density data is estimated as ±0.2% at a 95% confidence level. The isothermal compressibility and the isobaric thermal expansion were calculated by differentiation of the modified Tait equation. The uncertainties of isothermal compressibility and the isobaric thermal expansion are estimated to be less than ±1.7% and ±1.1%, respectively, at a 95% confidence level. The results are compared with the available literature data.

1. Introduction

Thermal energy storage systems are very important to achieve significant energy savings in order to get sustainability in its environmental, economic and social aspects. The storage of energy in adequate forms is a present day challenge to engineers and scientists. In particular, storing thermal energy is presently requiring further research in order to enable its large scale application. Among the materials in use for storing thermal energy, phase change materials (PCMs) are particularly interesting. Paraffins and their mixtures are a group of substances with potential for use as phase change materials (PCM) [1,2] near room temperature. Having in view large-scale applications of potential PCMs, namely involving the use of solar energy [3], the knowledge of their physical properties assumes an obvious relevance. Normal tetradecane (n-C_{14}) has a melting point near 6 °C [4] which makes it suitable for some "low temperature" energy storage applications, like v.g., cold water tanks. The development of models for the description of energy charge and discharge of PCMs, eventually contained in solid structures (v.g., capsules), require thermophysical properties of both the solid and liquid phases of the PCMs. Among other applications, the development of two-phase, moving boundary models, may require accurate data on the thermophysical properties of the PCM's, namely, their density, viscosity, and thermal conductivity, possibly at pressures above atmospheric pressure, v.g., in the case of containment in capsules.

The thermal conductivity of n-alkanes, including n-C_{14}, have been subject of rigorous measurements since, at least, the 1980's (vd., for instance Calado et al. [5]). However, this might eventually not be the case of viscosity and density measurements at moderately high pressures. In particular, the available viscosity results covering the range from (0.1–10) MPa, which is of great importance

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for many applications, are restricted to just one set of measurements [6]. In fact, as far as the authors are aware, only one set of viscosity values could be found in that pressure interval [6]. The present article has therefore a definite goal to provide information on the viscosity and density of n-C_{14} at moderately high pressures, where measurement results are scarce. Ultimately it is aimed to develop a reference viscosity correlation with density for n-C_{14}, which may prove useful for the development of estimation techniques for the properties of paraffin mixtures in use for thermal energy storage. It is believed that this goal is consistent with work that is being developed in the International Association for Transport Properties (IATP).

The present article follows the lines discussed in several recently published works regarding the strategy, the type of property correlation methods [7–12] and the experimental measurement technique mainly used by the present authors, namely, the vibrating wire method [13–16].

2. Materials and methods

2.1. Materials

The vibrating wire sensor was calibrated with toluene, supplied by Sigma Aldrich, having a nominal purity of 99.8%, without further treatment, except for drying to a water content of ca. 14 $\mathrm{mg \cdot kg^{-1}}$ and degassing by helium spraying.

The n-C_{14} sample was obtained from Merck KGaA, batch S7150867 (normal tetradecane GR for synthesis), with a minimum purity of 99%, used without further treatment except for drying to a water content less than 13 $\mathrm{mg \cdot kg^{-1}}$ and degassing by helium spraying. Drying of both toluene and n-C_{14} samples was performed using molecular sieves (Riedel-deHaën, 0.4 nm). The water content was checked using a Karl–Fischer 831 KF Coulometer from Metrohm.

Before introduction in the vibrating wire measurement cells, the liquid samples were filtered with a porous ceramic filter, with (16–40) μm porosity.

Table 1 shows the characterization of the liquid samples used for the present work, namely the toluene sample used in the calibration of the vibrating wire sensor and of the n-C_{14} sample used in the present viscosity and density measurements.

2.2. Vibrating U-tube density measurements

The density of n-C_{14} was measured from (283 to 373) K and pressures from (0.1 to about 70) MPa, along nine isotherms, using a vibrating U-tube DMA HP model densimeter from Anton Paar, using an Anton Paar DMA 5000 model as a reading unit. The experimental procedure was followed in accordance to Brito e Abreu et al. [17], with the calibration previously obtained by those authors [17], for temperatures up to 363 K. This calibration was extended for temperatures up to 423 K, using water with reference data from Wagner and Pruss [18]. The density range was also extended in this calibration. For this purpose 2-2-4 trimethyl pentane (isooctane) was used as a reference, with data from Pádua et al. [19].

The repeatability of the present density measurements is better than 0.1 $\mathrm{kg \cdot m^{-3}}$. The estimated overall maximum uncertainty of the density measurements is ±0.2%. This estimate is based on previous sensitivity studies [17], taking into account the particular characteristics of the present experiments.

2.3. Vibrating wire viscosity measurements

The viscosity, η, of n-C_{14} was measured along eight isotherms in the range in the range (283–358) K and at pressures up to 70 MPa. The vibrating wire instrument used for the n-C_{14} viscosity measurements was described elsewhere [10]. The working equations and the technique used for the measurements, are based on the treatment of the complex voltage measured at the wire ends, as described previously [20,21]. The electronic instrumentation and the acquisition system of the raw data have been described by Diogo et al. [10]. The temperature was measured with a 100 Ω platinum resistance thermometer, calibrated by EIA, Portugal, with an overall estimated uncertainty of ±0.05 K. During each run the temperature was stable within ±0.005 K. The repeatability of the viscosity measurements, at a 95% confidence level, is better than ±0.3% and their estimated expanded uncertainty is ±1.5%. These estimates are based on previous sensitivity studies [15,16,20–23] and checked in the present work by comparison with data obtained using an independent method, namely, a suspended-level capillary technique.

2.4. Capillary viscosity measurements

Independent measurements of the viscosity of n-C_{14} at atmospheric pressure were made with an Ubbelohde viscometer type 541 01/0a, manufactured and calibrated by SI Analytics GmbH, using an automatic Schott ViscoSystem® AVS 440 measuring unit. Its calibration constant has an uncertainty of ±1.2%, at a confidence level of 95% [24]. The temperature of the thermostatic oil bath was measured with a 100 Ω platinum resistance thermometer, calibrated by EIA, Portugal, with an overall estimated uncertainty of ±0.05 K [10]. The experimental setup and procedure were previously described by Diogo et al. [25]. Based on previous sensitivity studies [11,20,25], the overall uncertainty of these viscosity measurements was estimated as ±1.6%.

3. Density and derived properties

3.1. Density measurements

The results of the measurements of the density, ρ, of n-C_{14} in our laboratory obtained with an Anton Paar DMA HP densimeter, performed at pressures up to 70 MPa, and at nine temperatures from (283 to 373) K are shown in Table 2. The measurements have an estimated uncertainty of ±0.2% at a 95% confidence level. It should be noted that the (283, 288 and 293) K isotherms contain a restricted range of pressures, due to the low freezing pressures of n-C_{14} at these temperatures.

Table 1
Characterization of the samples used in this work.

Liquid	Source of Sample	Water content ($\mathrm{mg \cdot kg^{-1}}$)	Purity (Mass Fraction)
Toluene	Riedel-de Haën	14[a]	99.8%[b]
2,2,4-trimethyl pentane	Merck KGaA	–	99.5%[b]
n-Tetradecane	Merck KGaA	13[a]	99%[b]

[a] Water content of the samples after dried.

[b] Mass fraction purity reported by the supplier.