



Full Length Article

Viscosity of a $[x\text{CH}_4 + (1 - x)\text{C}_3\text{H}_8]$ mixture with $x = 0.8888$ at temperatures between (203 and 424) K and pressures between (2 and 31) MPa



Fernando F. Czubinski^a, Saif Z.S. Al Ghafri^{b,*}, Thomas J. Hughes^b, Paul L. Stanwix^b, Eric F. May^b

^a POLO-Research Laboratories for Emerging Technologies in Cooling and Thermophysics, Department of Mechanical Engineering, Federal University of Santa Catarina, 88040900 Florianópolis, SC, Brazil

^b Fluid Science and Resources Division, School of Mechanical and Chemical Engineering, University of Western Australia, 35 Stirling Highway, Crawley, WA 6009, Australia

ARTICLE INFO

Keywords:

Viscosity
Methane
Propane
Mixture
Vibrating wire
Viscosity models

ABSTRACT

Viscosity measurements of $[x\text{CH}_4 + (1 - x)\text{C}_3\text{H}_8]$ with $x = 0.8888$ are reported for temperatures between (203 and 424) K and pressures between (2 and 31) MPa using a vibrating wire viscometer clamped at both ends and operating in a steady-state mode. Reliable operation over this range of conditions required a detailed set of calibration and validation measurements using pure reference fluids. Most previous viscosity determinations with vibrating wire instruments have determined the important vacuum damping parameter Δ_0 from a single measurement and assumed it was temperature independent. Here we extended the calibration procedure beyond measurements in vacuum and helium (to determine the wire radius) to include low density methane ($\rho \leq 1.2 \text{ kg}\cdot\text{m}^{-3}$) from (223 to 420) K. Using viscosity values for these reference fluids linked to ab initio calculations revealed Δ_0 had a temperature dependence below about 350 K, increasing from 2.04×10^{-5} at 372 K to 5.79×10^{-5} at 223 K. Subsequent validation measurements with pure N_2 , He and CH_4 at pressures to 30 MPa confirmed the estimated standard relative uncertainty in viscosity of less than 2.5%. The binary mixture measurements were compared with literature data and the predictions of four models including two corresponding states based approaches (ECS and ST), a semi-theoretical model (VW) based on an extended hard-sphere scheme derived from the Enskog equation, and a model (LJ) based on molecular dynamics simulations of Lennard Jones fluids. The ECS and ST models exhibited systematic relative deviations from the data of up to -5% at $150 \text{ kg}\cdot\text{m}^{-3}$ and -10% at $300 \text{ kg}\cdot\text{m}^{-3}$, respectively. The LJ and VW models provided far better ($< 4\%$) representations of the data over their entire range, with the VW model able to represent all the measurements within 3%, which is comparable to their experimental uncertainty.

1. Introduction

Natural gas is a complex, methane-dominant multi-component mixture of light hydrocarbon species and impurities which typically include hydrogen sulfide, carbon dioxide and nitrogen. Accurate knowledge of the thermophysical properties of natural gas mixtures is important for process simulation and equipment design. Viscosity is one of the most important thermophysical properties required as it is needed, for example, when estimating equipment pressure drops and for use in heat transfer correlations. For design and simulation, reliable models that can accurately predict a fluid's viscosity over a wide range of temperature, pressure and composition are therefore desirable. Such models, however, need to be checked against experimental data to

verify their accuracy and moreover, are usually developed through regression to high quality measurements of key binary mixtures. The methane + propane system is one of the most important constituent binary mixtures for natural gas and its accurate representation over a wide range of conditions is essential to the performance of any model aiming to represent industrially relevant, multi-component natural gas mixtures.

Over the last few years, May and co-workers [1–4] have measured the viscosity of constituent natural gas binaries over a wide range of conditions as part of a program supported by the Gas Processors Association, using a vibrating wire viscometer (VWV) clamped at both ends. Measurements have typically spanned the temperature range (200 and 424) K at pressures to 31 MPa, and the acquired data have been

* Corresponding author.

E-mail address: saif.alghafri@uwa.edu.au (S.Z.S. Al Ghafri).

compared with various predictions, including those made with the extended corresponding states (ECS) model [5] implemented in the widely-used software package REFPROP 9.1 [6]. Locke et al. [1] and then Stanwix et al. [2] measured $[x\text{CH}_4 + (1-x)\text{C}_3\text{H}_8]$ mixtures with $x \approx 0.95$ over this range of conditions using the same VWV as the present work. In the work of Locke et al. [1], viscosities were evaluated for $x = 0.9452$, pressures in the range of (0.61 to 6.97) MPa and at temperatures of 280 and 298 K, which correspond to densities in the range (4.679 to 64.289) $\text{kg}\cdot\text{m}^{-3}$. Stanwix et al. [2] measured viscosities of this mixture for $x = 0.949$, temperatures between (200 and 423) K and pressures between (10 and 31) MPa, corresponding to densities of (120 to 360) $\text{kg}\cdot\text{m}^{-3}$. Both sets of results showed a systematic deviation of about -5% in the predictions of the ECS model around $\rho \approx 160 \text{ kg}\cdot\text{m}^{-3}$, which is, coincidentally, near the critical density of pure methane (but not of the mixture).

To our knowledge, the only other high pressure viscosity data available in the literature for this system are the measurements of Giddings et al. [7]; Huang et al. [8]; and Bicher and Katz [9]. The data from Giddings et al. [7] were obtained using a capillary viscometer for temperatures from (311 to 411) K, pressures in the range (0.1 to 55.1) MPa and x in the range (0.22 to 0.79), corresponding to densities in the range (0.6 to 506) $\text{kg}\cdot\text{m}^{-3}$. Huang et al. [8] used a falling cylinder viscometer to study this mixture at temperatures between (153 and 311) K, pressures in the range (3.4 to 34) MPa, and x in the range (0.22 to 0.75), corresponding to densities in the range (34 to 645) $\text{kg}\cdot\text{m}^{-3}$. Bicher and Katz [9] used a rolling ball viscometer to study this mixture at temperatures between (298 and 498) K, pressures in the range (0.1 to 34.4) MPa, and x in the range (0.2 to 0.8), corresponding to densities in the range of (0.5 to 513) $\text{kg}\cdot\text{m}^{-3}$.

However, the precision of the literature data is not generally sufficient to confirm whether the systematic deviation from the ECS model observed around $\rho \approx 160 \text{ kg}\cdot\text{m}^{-3}$, is attributable to an error in the data or is a deficiency of the model. Accordingly, to help establish the origin of this systematic deviation, and to extend significantly the available data set for this crucial natural gas constituent binary, we set out to measure a system that was significantly richer in propane and to compare the results obtained with two other state-of-the-art viscosity models [10–13].

Fig. 1 shows the location in the (p,T) plane of the measurements conducted in this work for a $[x\text{CH}_4 + (1-x)\text{C}_3\text{H}_8]$ mixture with $x = 0.8888$, relative to its phase envelope, which was calculated using the GERG-2008 EOS [14], implemented in REFPROP 9.1. The measurements cover the ranges (200 to 425) K and (2 to 31) MPa, with a total of 57 viscosity data being acquired from (10 to 64) $\mu\text{Pa}\cdot\text{s}$ at densities from (13 to 390) $\text{kg}\cdot\text{m}^{-3}$. The measurements were mainly conducted in the high pressure (≥ 10 MPa) region.

The predictions of four models were compared with the experimental data. Two of these, the ECS and SuperTrap (ST) models, are based on the corresponding states theory [5], and implemented in the

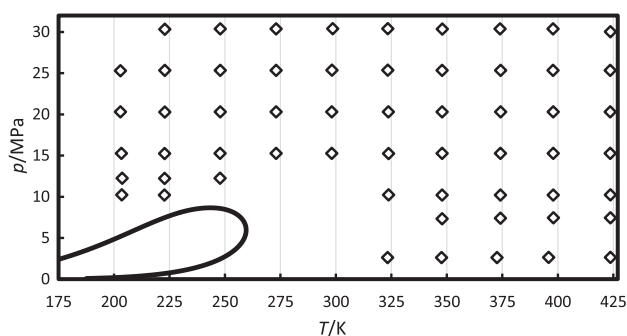


Fig. 1. Pressure and temperature conditions of the binary mixture viscosity data measured in this work $[x\text{CH}_4 + (1-x)\text{C}_3\text{H}_8]$ with $x = 0.8881$, together with its phase envelope calculated using the GERG-2008 EOS.

REFPROP 9.1 [6] and MultiFlash 4.4 [15,16] software packages, respectively. The third model, LJ, is a predictive model derived from molecular simulations combined with a corresponding states scheme, where the molecules are treated as spheres interacting through the Lennard-Jones potential, combined with a van der Waals one fluid approximation [10,11]. The fourth model, VW, is a semi-theoretical approach based on hard-sphere molecules in which the viscosity of a mixture is related to the viscosity of each individual component at the temperature and molar volume of interest. The pure fluid evaluation makes use of correlations fitted to experimental data for the core-size and the roughness factor at a reduced temperature and pressure [12,13].

To conduct the planned measurements, it was first necessary to replace the wire used in the apparatus previously (which had broken) and re-calibrate the viscometer. Previously, this process involved a direct measurement of the wire resonance when under vacuum to determine the apparatus parameter known as the vacuum damping, Δ_0 , which quantifies the loss on resonance in the absence of any viscous damping. Then the wire radius, r , was determined by measurements with helium. Normally these two parameters, which are quite strongly correlated when simultaneously determined by non-linear regression of a resonance peak measured when the wire is immersed in a fluid, are assumed to be independent of temperature. However, in this work we observed that Δ_0 varied significantly at temperatures below 350 K, and we detail here the method by which the temperature dependence of Δ_0 was determined.

The remainder of this paper is structured as follows. First, brief descriptions of the materials, apparatus and method are presented. Then the details of the calibration procedure and the results of the subsequent pure fluid validation measurements are described. The results of the mixture measurements are shown together with a comparison of the new data with results from the literature. Finally, we assess the performance of four viscosity models in terms of their ability to represent the new data.

2. Experimental section

2.1. Materials and apparatus overview

Table 1 presents the source and purities of the component fluids studied in this work. The binary mixture $[x\text{CH}_4 + (1-x)\text{C}_3\text{H}_8]$ with $x = (0.8888 \pm 0.0003)$ was prepared gravimetrically, as described in Refs. [1,2]. The uncertainty of the mole fraction was determined from contributions arising from the mass measurements, mole fraction purity of the component gases, and dead volume in the valve located at the top of the mixture preparation cylinder.

The details and measurement principles of the doubly-clamped vibrating wire viscometer (VWV) apparatus have been described in detail elsewhere [1,2]. Accordingly, only a brief description is provided here, including some minor modifications to both the apparatus and method used.

Single-phase fluids were transferred from the vessel (cylinder) in which they were prepared gravimetrically to the VWV by means of an ISCO model 260D syringe pump, which was then also used to control pressure during the measurements. Pressure was monitored with an

Table 1

Source and purities of chemicals used in this work. All purities are as specified by the supplier and no further purification was applied.

Compound	Supplier	Mole Fraction Purity
Helium	Coregas	0.99999
Nitrogen	Coregas	0.99999
Methane	BOC	0.99995
Propane	Air Liquide	0.99995

Download English Version:

<https://daneshyari.com/en/article/6631060>

Download Persian Version:

<https://daneshyari.com/article/6631060>

[Daneshyari.com](https://daneshyari.com)