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required to perform reservoir simulation during CO₂ injection. The knowledge of viscosity as a function of CO₂ composition, temperature and pressure is needed to predict diluted oil flow properties in porous media. With this aim in mind, an investigation of volumetric properties and viscosity of binary systems containing CO₂ and a hydrocarbon has been initiated in our laboratory. At the initial phase of this program, it was chosen to investigate the carbon dioxide + n-heptane binary mixture.

The present work aims at reporting experimental data of density and its derivative with respect to pressures of carbon dioxide + n-heptane system in the liquid state. For that purpose, density and speed of sound measurements were carried out in six mixtures ranging from 0 to 90% of CO₂ at pressure up to 70 MPa for two temperatures close to the critical temperature of CO₂ (303.15 and 313.15) K. Speed of sound was measured with a pulse echo technique working at 3 MHz whereas density was obtained from a U-tube densimeter.

FPF – O10 16:30 - 16:50 Monday

Room HS P1

NEW FORMULATION FOR THE VISCOSITY OF NORMAL BUTANE

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The exact knowledge of thermophysical properties of fluids with industrial importance is required for a more accurate basic design of compressors, gas turbines, and gas pipelines as well as cooling cycles and chemical processes. In contrast to the thermodynamic properties, the transport properties of normal butane, particularly in the region near to the critical point, are not sufficiently well-known. The current NIST standard database REFPROP 9.1 of Lemmon et al. (2013) recommends the viscosity correlation of Vogel et al. (1999), which is characterized by standard uncertainties of up to 3% in its range of validity applying an outdated equation of state of Younglove and Ely from 1987. Due to the fact that REFPROP 9.1 approves the reference equation of state of Bücker and Wagner (2006) for the thermodynamic properties of normal butane, there is a basic necessity for developing a new viscosity formulation for this working fluid.

Recently, very accurate viscosity measurements were performed by Herrmann and Vogel (2015) using a vibrating-wire viscometer in combination with a single-sinker densimeter. The standard uncertainty of these data was conservatively estimated to be 0.3%. Hence, they are considered to be primary data. In addition, viscosity measurements by Küchenmeister and Vogel (1998), whose results have already been used by Vogel et al. (1999) when generating their correlation, were re-evaluated (2017), so that they are qualified for being primary data, too.

Applying the new reference equation of state of Bücker and Wagner (2006) together with the improved data situation in the dense-gas region, a new viscosity formulation for normal butane was generated using the structure-optimisation method by Setzmann and Wagner (1989). The new formulation concept incorporates four contributions concerning the zero-density viscosity, the initial-density dependence, the near-critical region, and the higher-density terms of the

residual viscosity. The first two contributions are completely treated separately using little support of the kinetic theory. The critical-enhancement terms related to the near-critical region were slightly pre-treated with respect to the choice of some parameters. At the end, the bank of terms needed in the optimization procedure comprises only terms for the near-critical region and for the higher-density contribution. The latter terms are composed of a combination of double polynomials in the reduced density and reciprocal reduced temperature, some of them combined with a negative exponential function of the reduced density. The validity range of the new formulation extends from the melting line to temperatures of 650 K and to pressures of 100 MPa. The extrapolation and consistency behaviours of the formulation were investigated. Values calculated for the new viscosity formulation were compared with the primary data, used when developing the formulation, and with values resulting from the earlier viscosity correlations of Vogel et al. (1999) and of Quiñones-Cisneros and Deiters (2006).

FPF – O11 16:50 - 17:10 Monday

Room HS P1

PREDICTING THE VISCOSITY OF MULTICOMPONENT LIQUID MIXTURES USING AN MOLECULAR APPROACH

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In numerous industrial applications that involve the flow of fluids, knowledge of viscosity of the mixtures is an essential pre-requisite for good design and optimal operations. The present talk will focus on the mixtures containing *n*-alkane and aromatic species, as a precursor for developing molecular based methods [1-3] for predicting the viscosity of petroleum fluids; a rather challenging task, due in part to compositional complexity. For practical purposes the petroleum fluids are commonly described by a limited number of pseudo-components and there are many approaches to establish both the number and the characteristics of pseudo-components [4].

In this work, we present a simple molecular model, based on the extended hard-sphere approach [2,5-6], that relies on representing the viscosity of the multicomponent mixture by a viscosity of a single pseudo-component, which is characterized by an appropriate molecular weight. The initial validation of the predictive capability of the developed model is performed on compositionally well-defined liquid mixtures consisting of *n*-alkane and aromatic species, taking advantage of large number of experimental measurements available in literature. The results obtained indicate that the single pseudo-component representation works best for multicomponent mixtures composed of species with not too different molecular weight, as is the case in petroleum fluids. The model is capable of generating viscosities within 5% of the experimental value.

The observed good agreement with experimental data indicates that the proposed approach offers a tangible way forward in developing the molecular based model for predicting the viscosity of real crudes.