



Full Length Article

Expanded fluid-based thermal conductivity model for hydrocarbons and crude oils



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ABSTRACT

Thermal conductivity data for crude (mainly heavy) oils and mixtures of crude oils and pure hydrocarbons were collected at temperatures from 20 to 125 °C and pressures up to 10 MPa using a hot wire apparatus. A criterion was established to screen out data that were affected by convection. The screened data and data from the literature were used to develop an Expanded Fluid (EF) based thermal conductivity model for pure hydrocarbons, crude oils and their mixtures. The proposed model is applicable across the entire phase diagram including the critical region; however, it does not predict the critical enhancement of thermal conductivity observed in the vicinity of the critical point. The model inputs are the density of the fluid, the pressure, the dilute gas thermal conductivity, the compressed state density, and three other fluid specific parameters. The gas thermal conductivity is calculated from a well-established correlation. The compressed state density is obtained from the literature or from fitting the EF viscosity model to viscosity data. The three fluid specific parameters are determined by fitting the model to thermal conductivity data. The model fits the data of 63 pure hydrocarbons at temperatures from –150 to 330 °C and pressures up to 200 MPa with average deviation of 4%, except in the vicinity of the critical point ($0.97 < \text{reduced temperature} < 1.1$). The model also fits the data of 7 different crude oils at temperatures and pressures up to 150 °C and 10 MPa to within 0.3% of the experimental data. Mass based mixing rules were proposed for the model parameters of mixtures. The data for 19 pure hydrocarbon binaries at atmospheric pressure were predicted with average deviation of 0.5% and that of 8 bitumen/solvent pseudobinaries, at pressures up to 10 MPa, was predicted with average deviation of 1.6%. The introduction of binary interaction parameters into the mixing rules halved the magnitude of the deviations.

1. Introduction

The prediction of the thermal conductivity of hydrocarbons and crude oils across the phase diagram is required for the design and simulation of heat transfer and non-isothermal mass transfer processes in refinery operations [1,2]. Thermal conductivity is also required in the simulation of reservoir recovery operations, such as Steam Assisted Gravity Drainage (SAGD) and *in situ* combustion [3,4]. A thermal conductivity model suitable for use in process and reservoir simulators must be continuous across the phase diagram, rapid to solve, and have a small number of adjustable parameters.

There are several thermal conductivity correlations that are suitable for pure hydrocarbons, petroleum distillation cuts, and crude oils. The great majority of these models are empirical and constrained to the liquid phase. Extensive reviews of these liquid phase models are provided by the API Technical Databook [5], Poling et al. [6], and Riazi [7]. In general, these correlations are only applicable to pure

hydrocarbons and distillation cuts (with average molecular weights below 300 g/mol) at temperatures below their boiling point at atmospheric pressure. In general, those models predict the thermal conductivity of pure hydrocarbons within 15% of experimental values [6]. The predictive capability of these correlations has not been tested for heavier hydrocarbons and distillation cuts.

Other models such as Corresponding States (CS) and a Peng-Robinson equation of state (PR-EoS) analog model are applicable to both gas and liquid phases. The Corresponding States (CS) model calculates the thermal conductivity of a fluid from two contributions: one arising from translational degrees of freedom and which follows the CS principle; the other arising from internal degrees of freedom and which must be calculated from other established correlations. The translational thermal conductivity is related to that of a reference fluid (methane) at the same reduced temperature and density [8] or reduced temperature and pressure [9]. Either shape factors or a rotational coefficient are used to correct for the non-correspondence of most fluids

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to the methane reference. Baltatu et al. [10] introduced another parameter to the CS model that accounts for the aromatic and naphthenic components in the petroleum fractions. This version of the model uses propane as the reference fluid. Although the CS model provides reliable predictions for light hydrocarbons and petroleum fluids, its predictive capabilities for heavier fluids are limited because these fluids correspond to methane, or propane, at temperatures below their freezing point where no liquid thermal conductivity data are available. Another limitation of this model is that it is computationally intensive requiring iterative calculations for the estimation of the reference fluid properties and shape factors.

The PR-EoS analog model [11] uses an expression similar to that of the original PR EoS but the molar volume is replaced by the thermal conductivity. The original expressions for the PR EoS parameters a and b are modified by including an empirical fluid specific parameter which is function of the reduced temperature and the reduced pressure. This function contains four coefficients that are calculated by fitting the model to experimental data. The PR-EoS analog model has been tested on pure alkanes, CO₂, N₂ and three light petroleum distillates with average deviations within 12% of experimental data. This method requires an iterative procedure to calculate the fluid thermal conductivity from the roots of the equation. It has been tested on hydrocarbons with molecular weights up to 255 g/mol but not on heavier, more aromatic components, or crude oils.

The Expanded Fluid (EF) concept is an alternative approach for predicting the fluid phase properties of hydrocarbons, light, and heavy oils. According to the EF concept, properties that depend on the spacing between molecules, such as viscosity and thermal conductivity, decrease monotonically as the fluid expands from a compressed state near the liquid-solid (or liquid-glass) phase transition to the dilute gas state. The compressed state is defined as the density at which the viscosity of a fluid tends to infinity [12]. The EF concept has been successfully used to develop a model for the viscosity of hydrocarbons, heavy oils, and their mixtures across the phase diagram [13,14]. This model has also been extended to predict the viscosity of crude oils characterized into pseudo-components based on a distillation assay [15]. This approach has not yet been applied to thermal conductivity.

The objectives of this study were: 1) to develop a simple and fast convergence fluid phase thermal conductivity model based on the Expanded Fluid (EF) concept for pure hydrocarbons and heavy oils represented as single components; and, 2) to extend the model to mixtures through simple mass based mixing rules. The following steps were taken to develop an EF based thermal conductivity model: 1) assemble experimental thermal conductivity and density data of pure hydrocarbons from assorted chemical families including alkanes, cyclics and aromatics from the NIST Database [16]; 2) find a mathematical relationship between fluid expansion (inverse of density) and thermal conductivity applicable to hydrocarbons; 3) measure the thermal conductivity and density of crude oils, and; 4) test the proposed model on this dataset. The following steps were taken to develop the model mixing rules: 1) collect pure hydrocarbon binary thermal conductivity data from the literature; 2) measure the thermal conductivity of mixtures of heavy oil and solvents; and, 3) develop a set of mass based mixing rules that best fits the mixture data.

2. Experimental methods

2.1. Materials and sample preparation

The following dead (gas free) crude oil samples were used in this study: WC-B-B3, WC-B-A3(1), WC-B-A3(2), WC-B-A1, CO-B-B1, EU-HO-A1, ME-CV-A1, and a deasphalted sample of WC-B-B3 (WC-B-B3-DAO). WC, CO, EU and ME correspond to the oil producing regions of Western Canada (WC), Colombia (CO), Europe (EU) and Middle East (ME), respectively. B, HO and CV indicate bitumen, heavy oil, or conventional oil, respectively, and the third term indicates the source reservoir and

Table 1

Specific gravity (SG), atomic hydrogen-to-carbon (H/C) ratio, molecular weight (MW), viscosity, μ , and thermal conductivity, λ , both at 20 °C and atmospheric pressure, pentane precipitated asphaltene (C5-Asph.) content, and toluene insoluble (TI) content of the oil samples used in this study.

Sample	SG	H/C	MW g/mol	μ at 20 °C mPa·s	λ at 20 °C mW m ⁻¹ K ⁻¹	C5-Asph. wt%	TI wt%
WC-B-B3	1.020	1.473	558	150,000	137.0	22	0.68
WC-B-A3(1)	1.010	1.453	550	356,000	138.3	18	0.55
WC-B-A3(2)	1.009	1.453	550	300,000	139.6	18	0.55
CO-B-B1	1.000	1.473	577	158,000	136.1	22	0.74
WC-B-A1	0.996	1.577	585	72,800	137.8	16	0.51
EU-HO-A1	0.968	1.596	475	5040	129.0	7	0.31
ME-CV-A1	0.872	1.756	475	18.1	125.0	3.8	0.03
WC-B-B3-DAO	0.984	1.533	483	1600	133.1	0	0

sample number. The WC-B-A3(1) and WC-B-A3(2) samples were recovered from the same reservoir but had different density, viscosity and thermal conductivity. Selected physical properties and the asphaltene and toluene insoluble contents of the samples are summarized in Table 1. The preparation of deasphalted oil (DAO) samples as well as the determination of asphaltene and toluene insoluble material was described in detailed elsewhere [15].

The solvents used for the preparation of the diluted bitumen samples were *n*-pentane (purity of 99.5%), *n*-heptane (purity of 99.5%), *n*-tetradecane (purity of 99.5%), toluene (purity of 99.5%) and cyclohexane (purity of 99%). The *n*-pentane, *n*-heptane, *n*-tetradecane, toluene and cyclohexane were obtained from VWR. Additionally, *n*-pentane, *n*-heptane, *n*-tetradecane toluene and deionized ultrafiltered water (obtained from Fisher Scientific Chemicals) were used for the calibration and validation of the thermal conductivity apparatus utilized in this study.

Mixtures of gas free crude oil (dead oil) and liquid solvents (*n*-pentane, *n*-heptane, *n*-tetradecane, toluene and cyclohexane) were prepared at ambient temperature and atmospheric pressure. A known mass of dead oil and solvent (approximately 200 g in total) were placed in a beaker equipped with a lid and mixed continuously in a rotary mixer at 6 rpm until a homogenous mixture was observed. The mixture was considered homogeneous when its density was no longer changing with mixing time, typically after 12 h. Once the mixture was homogeneous, its final mass was determined in order to recalculate the solvent concentration to take into account any evaporation (usually < 1 wt%) which was assumed to be only from the solvent.

2.2. Thermal conductivity measurements

2.2.1. Apparatus

An apparatus was commissioned and built to measure the thermal conductivity of liquid samples using the transient hot wire technique. The physical principle behind the hot wire technique is the transfer of a constant heat flux during a short period of time from a thin platinum (Pt) wire to an infinite acting medium of constant physical properties. The platinum wire, which serves as heating element and thermometer, is immersed into the medium in a cylindrical configuration that gives a radial heat flux. The rate at which the temperature of the wire changes is a function of the thermal conductivity of the medium. The thermal conductivity of the medium is determined from the analytical solution of the transient one-dimensional Fourier equation [17,18]:

$$\Delta T_w = \frac{q}{4\pi\lambda} \left[\ln \left(\frac{4\alpha t}{r_o^2} \right) - \gamma \right] \quad (1)$$

where ΔT_w is the temperature change of the wire, λ and α are the thermal conductivity and thermal diffusivity of the medium, respectively, q is the heat per unit length, r_o is the radius of the wire, t is the time, and γ is the Euler constant ($\gamma = 0.5772\dots$).

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