

# Measurement and Correlation of the Thermal Conductivity of Propane from 86 K to 600 K at Pressures to 70 MPa

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New experimental data on the thermal conductivity of propane are reported that allow improved correlations to be developed. Previous correlations have been limited by a lack of thermal-conductivity data for the vapor at temperatures below 300 K and liquid data near the critical point. In addition, significant discrepancies were noted in the high-temperature dilute-gas thermal conductivity. The present data cover the temperature range from the triple point at 85.5 K to 600 K and the pressure range 0.1 to 70 MPa. They are estimated to have an uncertainty of 1% for measurements removed from the critical point and at pressures above 1 MPa, which increases to 3% in the critical region and 4% at low pressures (<1 MPa). These new experimental data are used together with the previously available data to develop improved correlations for the thermal conductivity of propane. The thermal-conductivity correlation for propane is estimated to have an uncertainty of about 3% at a 95% confidence level, with the exception of state points near the critical point and the dilute gas, where the uncertainty of the correlation increases to 5%.

## Introduction

Accurate thermophysical property data are required for industrially important fluids such as propane to allow the design of efficient chemical processes and equipment. The large uncertainty associated with existing theoretical predictions for the transport properties has motivated efforts to develop empirical correlations to represent the transport properties as functions of temperature and pressure (or density). These correlations must be based on accurate experimental data that cover the entire fluid region of interest. An international effort has been coordinated by the IUPAC Subcommittee on Transport Properties to develop accurate correlations for such fluids. These correlations require a careful selection of the best available experimental data, based on a critical analysis of the measurement techniques and comparisons with other reliable data. New experimental data are reported here for the thermal conductivity of propane that were not available during development of the wide-range correlations proposed by Holland et al.<sup>1</sup> in 1979, Younglove and Ely<sup>2</sup> in 1987, and Ramires et al.<sup>3</sup> in 1996. The present work describes improved empirical correlations for the thermal conductivity of propane incorporating these new data.

Although desirable, a complete theoretical analysis of the available experimental data for the thermal conductivity of propane in the limit of zero density and in the critical region cannot yet be performed. The thermal conductivity of polyatomic molecules is strongly influenced by inelastic collisions and the exchange of energy between translational and the internal modes (rotation, vibration, electronic) during collisions. The propane molecule is characterized

by a significant number of excited degrees of freedom, including hindered rotations. Thus, the kinetic theory for the thermal conductivity in the dilute gas phase of polyatomic molecules is still approximate and the calculations are heuristic.<sup>4</sup> In the critical region, the absence of a crossover equation of state for propane does not allow an adequate theoretically based description of the thermal-conductivity enhancement and, thus, an empirical formulation was instead considered in this work.

Three wide-range empirical correlations for the thermal conductivity of propane have been published based on a comprehensive analysis of the data available to the authors. In 1979, Holland et al.<sup>1</sup> reported a correlation for propane at temperatures from 140 K to 500 K and at pressures up to 50 MPa, which has an estimated uncertainty<sup>2,3</sup> of 8% outside the critical region and 15% near the critical point. In 1987, Younglove and Ely<sup>2</sup> reported a correlation for propane at temperatures from 86 K to 600 K and at pressures up to 100 MPa that has an estimated uncertainty<sup>2</sup> of 5% outside the critical region and an uncertainty of 10% near the critical point. In 1996, Ramires et al.<sup>3</sup> reported a correlation for propane at temperatures from 192 K to 725 K and at densities up to 17 mol·L<sup>-1</sup> that has an estimated uncertainty<sup>3</sup> of 5% outside the critical region and an uncertainty of 10% near the critical point. The experimental data reported here, which cover a wide range of temperature and pressure, with improved coverage of the critical region and low-temperature vapor where almost no data existed, motivated the development of improved correlations. It will be shown that systematic deviations between thermal-conductivity data and the previous correlations<sup>1–3</sup> can be significantly reduced in the critical region and the dilute vapor.

## Experimental Section

The purity of the propane sample studied was verified to be 99.95 mol % by a gas-chromatographic mass-

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spectrometric method. The most significant impurity was ethane, at a concentration of 0.03 mol %. The measurements of thermal conductivity were obtained with two transient hot-wire instruments that have been described in detail.<sup>5,6</sup> Both instruments used dual hot wires 4  $\mu\text{m}$  in diameter and made from tungsten. The outer cavity around the hot wires is stainless steel and has a diameter of 9 mm. The low-temperature instrument is capable of operation from 30 K to 340 K at pressures to 70 MPa in the liquid, vapor, and supercritical gas phases. The high-temperature instrument is capable of operation from 250 K to 750 K at pressures to 70 MPa in the liquid, vapor, and supercritical gas phases. Temperatures are determined with a reference platinum resistance thermometer with an uncertainty of  $\pm 0.01$  K, and pressures are determined with a pressure transducer with an uncertainty of  $\pm 7$  kPa. The basic theory that describes the operation of the transient hot-wire instrument is given by Healy et al.<sup>7</sup> Each hot-wire cell is designed to approximate a transient line source as closely as possible, and deviations from this model are treated as corrections to the experimental temperature rise. The ideal temperature rise  $\Delta T_{\text{id}}$  is given by

$$\Delta T_{\text{id}} = \frac{q}{4\pi\lambda} \left[ \ln(t) + \ln\left(\frac{4a}{r_0^2 C}\right) \right] = \Delta T_w + \sum_{i=1}^{10} \delta T_i \quad (1)$$

where  $q$  is the power applied per unit length,  $\lambda$  is the thermal conductivity of the fluid,  $t$  is the elapsed time,  $a = \lambda/\rho C_p$  is the thermal diffusivity of the fluid,  $\rho$  is the density of the fluid,  $C_p$  is the isobaric specific heat capacity of the fluid,  $r_0$  is the radius of the hot wire,  $C = 1.781\dots$  is the exponential of Euler's constant,  $\Delta T_w$  is the measured temperature rise of the wire, and  $\delta T_i$  are corrections<sup>7</sup> to account for deviations from ideal line-source conduction.

Both thermal conductivity and thermal diffusivity can be determined with the transient hot-wire technique, but only the thermal conductivity results are considered here. For gas-phase measurements, two significant corrections<sup>7-10</sup> must be carefully considered. First, since the thermal diffusivity of the gas is much different from that of the wire, the correction for the wire's finite radius becomes very significant. Second, the thermal diffusivity of the dilute gas varies inversely with the pressure, so it is possible for the transient thermal wave to penetrate to the outer boundary of the gas region during an experiment at low pressures.<sup>8-11</sup> The preferred method to deal with such corrections is to minimize them by proper design. For instance, the correction for finite wire radius can be minimized with wires of extremely small diameter (4 to 7)  $\mu\text{m}$  and penetration of the thermal wave to the outer boundary can be eliminated by use of a cell with an outer boundary of large diameter. However, such designs are often not optimum for a general-purpose instrument, where extremely fine wires may be too fragile and large outer dimensions may require too much of a scarce sample, particularly in the liquid phase.

The present transient hot-wire wires are optimum to minimize the corrections for the wire's finite radius during such dilute-gas measurements. However, measurement times must be carefully selected to minimize the correction for penetration to the outer boundary because of the relatively small diameter of the outer boundary. For the measurements reported here, only application of the full correction<sup>7</sup> for the finite wire dimensions was considered adequate. For a few of the measurements at the lowest pressures, the outer boundary was encountered during the one-second duration of the experiment and so the experiment time was reduced to minimize the magnitude of this

correction. The largest gas thermal diffusivity for any of the reported measurements was  $2 \times 10^{-5} \text{ m}^2 \cdot \text{s}^{-1}$ . This conclusion is consistent with previous work on light gases such as argon and nitrogen at 1 bar to 2 bar,<sup>9-11</sup> where the outer boundary was encountered at times below 1 s when the thermal diffusivity was greater than  $9 \times 10^{-6} \text{ m}^2 \cdot \text{s}^{-1}$ .

At these very low pressures, the steady-state hot-wire technique has the advantage of not requiring significant corrections. The working equation for the steady-state mode is based on a different solution of Fourier's law, but the geometry is still that of concentric cylinders. The solution can be found in standard texts for the case of constant thermal conductivity (see, for example, ref 12, page 114). This equation can be solved for the thermal conductivity of the fluid,  $\lambda$ ,

$$\lambda = \frac{q \ln\left(\frac{r_2}{r_1}\right)}{2\pi(T_1 - T_2)} \quad (2)$$

where  $q$  is the applied power per unit length,  $r_2$  is the internal radius of the outer cylinder,  $r_1$  is the external radius of the inner cylinder (hot wire), and  $\Delta T = (T_1 - T_2)$  is the measured temperature difference between the hot wire and its surrounding cavity.

For the concentric-cylinder geometry described above, the total heat flux per unit length  $q$  remains constant and is not a function of the radial position. Assuming that the thermal conductivity is a linear function of temperature, such that  $\lambda = \lambda_0(1 + b_\lambda T)$ , it can be shown that the measured thermal conductivity is given by  $\lambda = \lambda_0(1 + b_\lambda(T_1 + T_2)/2)$ . Thus, the measured thermal conductivity corresponds to the value at the mean temperature  $\bar{T}$  of the inner and outer cylinders, where

$$\bar{T} = (T_1 + T_2)/2 \quad (3)$$

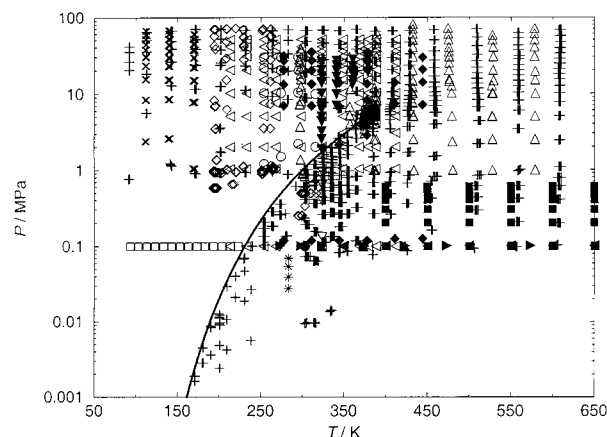
This assumption of linear temperature dependence for the thermal conductivity is valid only for experiments with small temperature rises. The density assigned to the measured thermal conductivity is taken from an equation of state for the temperature from eq 3 and the experimentally measured pressure. An assessment of corrections during steady-state hot-wire measurements is available.<sup>13</sup>

## Experimental Results

The results of measurements of the thermal conductivity of the vapor phase of propane are tabulated in the Supporting Information. The range of state points covered by the present measurements is shown in Figure 1 relative to the vapor-pressure curve of propane and the previous measurements that are available in the literature. There are a total of 393 steady-state measurements and 1380 transient measurements tabulated at temperatures from the triple point to 600 K with pressures to 70 MPa. The measurements are reported on the ITS-90 temperature scale and are estimated to have an uncertainty of  $\pm 1\%$  for transient measurements removed from the critical point and at pressures above 1 MPa, increasing to  $\pm 3\%$  in the critical region and at low pressures ( $< 1$  MPa) at a 95% confidence level. The steady-state vapor measurements are estimated to have an uncertainty of  $\pm 2\%$ . A summary of these measurements is given in Table 1 while the complete data set is available in the Supporting Information. Many of these vapor measurements were made at temperatures below the normal boiling point of propane. It should be

**Table 1. Summary of Present Measurements on Propane**

technique	temp range/K	pressure range/MPa	no. of points	estimated uncertainty
steady-state hot wire (low temperature)	170–300	0.004–0.35	158	±2%
steady-state hot wire (high temperature)	300–604	0.009–1.90	235	±2%
transient hot wire (low temperature)	89–300	0.10–70.0	340	±1–2%
transient hot wire (high temperature)	300–604	0.07–70.0	1040	±1–3%



**Figure 1.** Distribution of thermal conductivity data for propane reported here and available in the literature: +, present work; ○, Yata;<sup>15</sup> ●, Clifford et al.;<sup>18</sup> ×, Roder and Nieto de Castro;<sup>19,20</sup> △, Tufeu and Le Neindre;<sup>21</sup> ◇, Prasad et al.;<sup>22</sup> \*, Mann and Dickens;<sup>23</sup> ▼, Leng and Comings;<sup>24</sup> ▲, Smith et al.;<sup>25</sup> ◆, Carmichael et al.;<sup>26</sup> □, Brykov and Mukhamedzyanov;<sup>27</sup> ■, Aggarwal and Springer;<sup>28</sup> ▽, Zheng et al.;<sup>29</sup> open triangle pointing right, Vines and Bennett;<sup>30</sup> solid triangle pointing right, Senftleben;<sup>31</sup> open triangle pointing left, Ryabtsev and Kazaryan;<sup>32</sup> solid triangle pointing left, Ehya et al.<sup>33</sup>

emphasized that there are many difficulties associated with such measurements because of the small quantity of sample in the cell and possible contamination with air impurities and adsorbed material on the surface of the cell and wires.<sup>14</sup> The uncertainty of such vapor measurements is larger than that for the measurements in the liquid or vapor phase at higher reduced temperatures and pressures because of sample handling as well as relatively large corrections for transient measurements. An equation of state<sup>2</sup> is necessary during the data analysis to make the corrections to the measured temperature rise as described above.

### Correlation Development

The thermal conductivity is represented as a sum of three contributions,

$$\lambda(\rho, T) = \lambda_0(T) + \Delta\lambda_r(\rho, T) + \Delta\lambda_c(\rho, T) \quad (4)$$

where  $\lambda_0$  is the dilute-gas thermal conductivity, which is dependent only on the temperature,  $\Delta\lambda_r$  is the residual thermal conductivity, and  $\Delta\lambda_c$  is the thermal-conductivity enhancement in the critical region.  $\Delta\lambda_r$  and  $\Delta\lambda_c$  depend on both density and temperature. This representation is useful, since it allows the theoretically based analysis of each contribution to be considered separately. This is particularly useful when the dilute-gas thermal conductivity and the thermal-conductivity enhancement in the critical region are examined.

To analyze the thermal conductivity in terms of density and temperature, the density of the fluid must be determined from the temperatures and pressures reported by each author. The modified Benedict–Webb–Rubin (MBWR) equation of state of Younglove and Ely<sup>2</sup> was used. This equation of state is valid at pressures up to 100 MPa and

**Table 2. Primary Experimental Data for the Thermal Conductivity of Propane**

ref	temp range/K, pressure range/MPa	technique <sup>a</sup>	no. of points	estimated uncertainty/%
15	254–315, 1–30	THW	16	±1
18	303, 0.1	THW	1	±1
19 and 20	110–300, 1–70	THW	400	±1.5
21	298–578, 1–70	CC	175	±2
22	192–320, 0.2–70	THW	128	±1.5

<sup>a</sup> THW, transient hot wire; CC, concentric cylinders.

**Table 3. Secondary Experimental Data for the Thermal Conductivity of Propane**

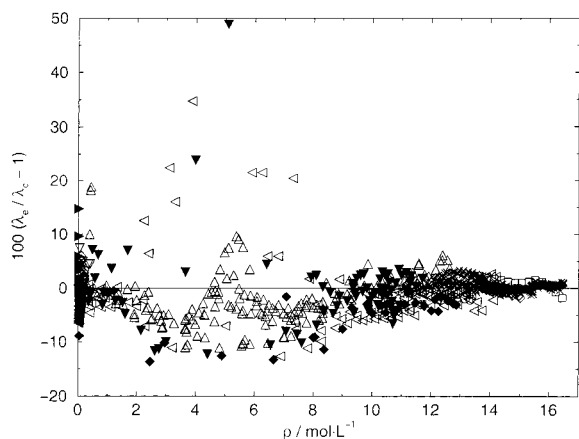
ref	temp range/K, pressure range/MPa	technique <sup>a</sup>	no. of points	estimated uncertainty/%
23	275–285, ~0.1	SSHW	6	±5
24	323–413, 0.1–30	CC	83	±5
25	323–423, 0.1	CC	3	±5
26	277–444, 0.1–35	SC	33	±5
27	93–223, 0.1	SSHW	14	±5
28	400–725, 0.1–0.6	CC	41	±3
29	323.75, 0.1–1	CC	6	±3
30	373–413, 0.1	SSHW	3	±5
31	273–673, 0.1	SSHW	8	±5
32	213–412, 0.1–50	SSHW	155	±5
33	300–1000, 0.1	SSHW	15	±5

<sup>a</sup> SSHW, steady-state hot wire; THW, transient hot wire; CC, concentric cylinders; SC, spherical cell.

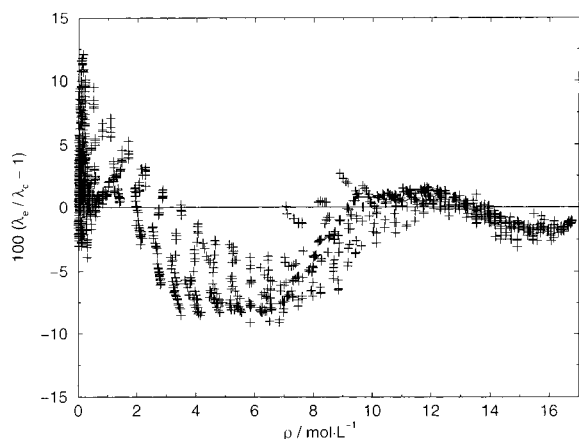
at temperatures between the triple point and 600 K. With the exception of the more recent results of Yata et al.,<sup>15</sup> all experimental data and the equation of state were reported prior to 1990, when the ITS-90 temperature scale was adopted. Therefore, the reported temperatures were converted to this scale according to the recommendations of IUPAC<sup>16</sup> and all calculations are based on the ITS-90 temperature scale. All densities are in moles per liter, temperatures are in kelvin, and stated uncertainties are at the 95% (2 $\sigma$ ) confidence level.

The available experimental data were initially grouped into categories of primary and secondary data according to the criteria described by Nieto de Castro et al.<sup>17</sup> However, this requirement of absolute measurements with uncertainties less than 2% excluded all of the data prior to 1982 except for one gas point and considered only four additional sets of data. Measurements of thermal conductivity made with relative instruments (calibrated through measurements with one or more reference fluids) have often been unreliable and have additional uncertainties that are poorly characterized. Consequently, it was decided to exclude all the relative measurements. A summary of the primary data for propane,<sup>15,18–22</sup> together with the ranges in temperature and pressure, the experimental technique, and the assigned uncertainty, is provided in Table 2. A summary of the secondary data for propane,<sup>23–33</sup> together with the ranges in temperature and pressure, the experimental technique, and the assigned uncertainty, is provided in Table 3.

The need for improved correlations for the thermal conductivity of propane is evident when the literature data and the present results are compared with the previous

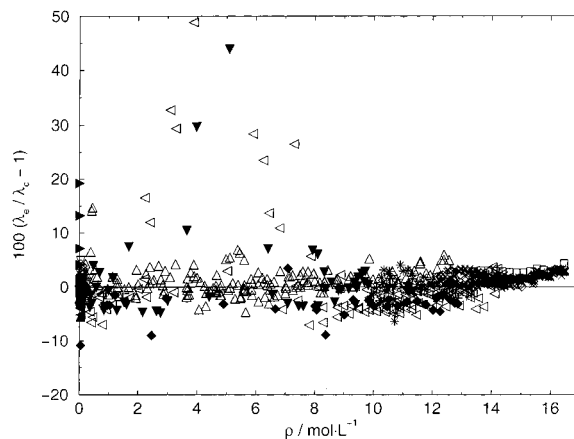


**Figure 2.** Relative deviations between literature values for the thermal conductivity of propane and the correlation of Younglove and Ely;<sup>2</sup> ○, Yata;<sup>15</sup> ●, Clifford et al.;<sup>18</sup> ×, Roder and Nieto de Castro;<sup>19,20</sup> △, Tufeu and Le Neindre;<sup>21</sup> ◇, Prasad et al.;<sup>22</sup> \*, Mann and Dickins;<sup>23</sup> ▼, Leng and Comings;<sup>24</sup> ▲, Smith et al.;<sup>25</sup> ◆, Carmichael et al.;<sup>26</sup> □, Brykov and Mukhamedzyanov;<sup>27</sup> ■, Aggarwal and Springer;<sup>28</sup> ▽, Zheng et al.;<sup>29</sup> open triangle pointing right, Vines and Bennett;<sup>30</sup> solid triangle pointing right, Senftleben;<sup>31</sup> open triangle pointing left, Ryabtsev and Kazaryan;<sup>32</sup> solid triangle pointing left, Ehya et al.<sup>33</sup>

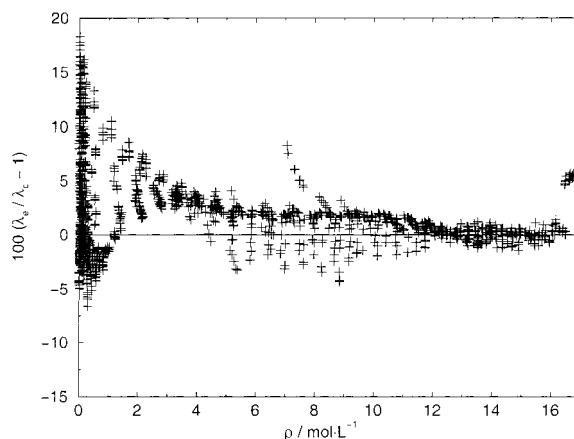


**Figure 3.** Relative deviations between the present measurements for the thermal conductivity of propane and the correlation of Younglove and Ely.<sup>2</sup>

correlations. The relative deviations between the literature data and the correlation of Younglove and Ely<sup>2</sup> are shown in Figure 2. In the critical region, deviations range from 10% to 50%. The relative deviations between the present measurements and the correlation of Younglove and Ely<sup>2</sup> are shown in Figure 3. Near the critical density (5.000 mol·L<sup>-1</sup>), the bulk of the literature data and the present measurements are lower than the correlation<sup>2</sup> by 10%. At temperatures above 450 K, the dilute-gas thermal conductivity data are systematically higher than the correlation<sup>2</sup> by 10% to 15%. The relative deviations between the literature data and the correlation of Ramires et al.<sup>3</sup> are shown in Figure 4. In the critical region, deviations again range from 10% to 50% for a few isolated data points. The relative deviations between the present measurements and the correlation of Ramires et al.<sup>3</sup> are shown in Figure 5. The correlation of Ramires et al.<sup>3</sup> does much better than the correlation of Younglove and Ely<sup>2</sup> at high temperatures near the critical density. It is also systematically lower than the present dilute-gas measurements and the data of Senftleben<sup>31</sup> and Tufeu and LeNeindre<sup>21</sup> by 10% to 15% at temperatures above 450 K. Thus, improved correlations



**Figure 4.** Relative deviations between literature values for the thermal conductivity of propane and the correlation of Ramires et al.<sup>3</sup> ○, Yata;<sup>15</sup> ●, Clifford et al.;<sup>18</sup> ×, Roder and Nieto de Castro;<sup>19,20</sup> △, Tufeu and Le Neindre;<sup>21</sup> ◇, Prasad et al.;<sup>22</sup> \*, Mann and Dickins;<sup>23</sup> ▼, Leng and Comings;<sup>24</sup> ▲, Smith et al.;<sup>25</sup> ◆, Carmichael et al.;<sup>26</sup> □, Brykov and Mukhamedzyanov;<sup>27</sup> ■, Aggarwal and Springer;<sup>28</sup> ▽, Zheng et al.;<sup>29</sup> open triangle pointing right, Vines and Bennett;<sup>30</sup> solid triangle pointing right, Senftleben;<sup>31</sup> open triangle pointing left, Ryabtsev and Kazaryan;<sup>32</sup> solid triangle pointing left, Ehya et al.<sup>33</sup>

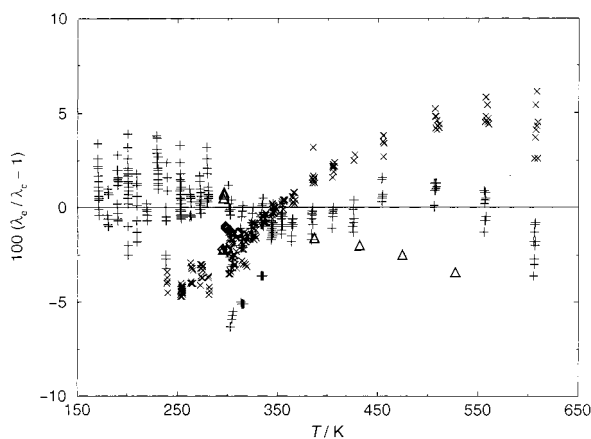


**Figure 5.** Relative deviations between the present measurements for the thermal conductivity of propane and the correlation of Ramires et al.<sup>3</sup>

should be developed for the thermal conductivity of propane.

**Zero-Density Limit.** The zero-density or dilute-gas limit is determined from the extrapolation of a particular transport property, held at constant temperature, to zero density.<sup>4</sup> Consequently, dilute-gas transport properties cannot be directly measured. Reliable extrapolation of low-density vapor data requires that the data span a reasonable range of density. This is not possible at the lowest temperatures, where the vapor pressure of propane is of the order of 0.1 MPa and there is an insufficient density range covered to allow extrapolation to zero density. It was assumed here that at low densities, less than 1 MPa, the density dependence is linear with a constant slope. Thus, the available thermal conductivity data for the vapor phase over the full range of temperature, at pressures less than 1 MPa or the vapor pressure at any given temperature, were fit with a polynomial in reduced temperature with a constant linear density dependence to allow extrapolation to the true zero-density dilute-gas limit.

Given the need for vapor thermal conductivity data at low temperatures and pressures, it was decided that the



**Figure 6.** Relative deviations between the present steady-state (+) and transient (x) measurements for the thermal conductivity of propane and the dilute-gas thermal conductivity calculated with eq 5. Deviations are shown between the dilute-vapor data:  $\Delta$ , Tufeu and LeNeindre;<sup>21</sup>  $\diamond$ , Prasad et al.<sup>22</sup>

**Table 4. Coefficients for the Representation of the Thermal Conductivity of Propane in the Limit of Zero Density, Eq 5**

$A_1/(\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1})$	$-1.247\ 78 \times 10^{-3} \pm 2.0727 \times 10^{-4}$
$A_2/(\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1})$	$8.163\ 71 \times 10^{-3} \pm 4.4489 \times 10^{-4}$
$A_3/(\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1})$	$1.993\ 74 \times 10^{-2} \pm 2.2096 \times 10^{-4}$

present steady-state measurements were the most reliable and should form the basis for the dilute-gas thermal conductivity of propane. The present thermal conductivity data from steady-state measurements were correlated with a polynomial, quadratic in temperature,

$$\lambda_0(T) = A_1 + A_2(T/T_c) + A_3(T/T_c)^2 \quad (5)$$

where  $\lambda_0$  is the dilute-gas thermal conductivity,  $T$  is the absolute temperature, and  $T_c$  is the critical temperature. Table 4 contains the coefficients  $A_i$  together with their uncertainties at a level of confidence of one standard deviation. The maximum deviation of the primary experimental data is 4%. The deviations of the present measurements and the primary data from eq 5 are presented in Figure 6. Good agreement is found between the present steady-state measurements and the steady-state measurements of Tufeu and Le Neindre<sup>21</sup> and the transient measurements of Prasad et al.<sup>22</sup> Good agreement is also found between the present transient measurements (uncertainty of  $\pm 3\%$  for dilute vapor) and the present steady-state measurements (uncertainty  $\pm 2\%$ ). The transient measurements are up to 5% higher than the steady-state measurements at 600 K. This difference is systematic and has been observed for other hydrocarbons and alternative refrigerants. No theoretical basis for this difference is available, but steady-state measurements at low-density are considered more reliable, since significant corrections are required for the transient measurements at these conditions. The uncertainty of the dilute-gas thermal conductivity calculated with eq 5 is estimated to be  $\pm 4\%$  at a level of confidence of 95%.

**Residual Thermal Conductivity.** The thermal conductivities of pure fluids exhibit an enhancement over a large range of densities and temperatures around the critical point and become infinite at the critical point. This behavior can be described by models that produce a smooth crossover from the singular behavior of the thermal conductivity asymptotically close to the critical point to the

nonsingular background values far away from the critical point.<sup>34–37</sup> The contribution of critical enhancement to the thermal conductivity in eq 4 can be isolated

$$\lambda(\rho, T) = \bar{\lambda}(\rho, T) + \Delta\lambda_c(\rho, T) \quad (6)$$

where  $\bar{\lambda}(\rho, T) = \lambda_0(T) + \Delta\lambda_r(\rho, T)$  is the background contribution and  $\rho$  is the fluid density.

To assess the critical enhancement either theoretically or empirically, we need to evaluate the background contribution, which is the sum of the contributions of the dilute-gas and residual-thermal-conductivity terms. The procedure adopted during this analysis was somewhat different, since the regression software, ODRPACK V. 2.01<sup>38</sup> allows a fit of all the primary data simultaneously to the residual thermal conductivity and the critical enhancement, while maintaining the parameters already obtained from the fit of the dilute-gas thermal-conductivity data.

The residual thermal conductivity was represented by a polynomial in temperature and density

$$\Delta\lambda_r(\rho, T) = \sum_{i=1}^5 (B_{i,1} + B_{i,2}(T/T_c))(\rho/\rho_c)^i \quad (7)$$

where  $\rho$  is the density and  $\rho_c$  is the critical density.

**Empirical Critical Enhancement.** For applications that are relatively distant from the critical region, the critical enhancement is represented by the following empirical expression,

$$\Delta\lambda_c(\rho, T) = \frac{C_1}{C_2 + |\Delta T_c|} \exp[-(C_3 \Delta\rho_c)^2] \quad (8)$$

where  $\Delta T_c = (T/T_c) - 1$  and  $\Delta\rho_c = (\rho/\rho_c) - 1$ . This equation does not require accurate information on the compressibility and specific heat of propane in the critical region, as does the theory of Olchowy and Sengers.<sup>34–37</sup> The critical point is that of Younglove and Ely,<sup>2</sup> with  $T_c = 369.82$  K (369.85 K on IPTS-68) and  $\rho_c = 5.000$  mol·L<sup>-1</sup> (220.3 kg·m<sup>-3</sup>). The coefficients of eq 5 were fixed while the coefficients of eqs 7 and 8 were fit with ODRPACK V. 2.01<sup>38</sup> to the present transient and steady-state data for the thermal conductivity of propane. Table 5 lists the optimum coefficients for eqs 7 and 8 together with their uncertainties at a level of confidence of one standard deviation. Figure 7 shows deviations of the experimental data from the surface correlation of eqs 5, 7, and 8 incorporating this empirical equation for the representation of the critical enhancement. Around the critical region, despite the lack of a crossover equation of state, the primary data are generally reproduced within  $\pm 10\%$ , with the two near-critical isotherms of Tufeu and LeNeindre<sup>21</sup> exhibiting the largest deviations ( $> 20\%$ ). The present transient and steady-state measurements are represented to within 4% at a level of confidence of 95%.

**Simplified Crossover Model.** The theoretically based crossover model proposed by Olchowy and Sengers<sup>34–36</sup> is complex and requires solution of a quartic system of equations in terms of complex variables. A simplified crossover model has also been proposed by Olchowy and Sengers.<sup>37</sup> The critical enhancement of the thermal conductivity is given by

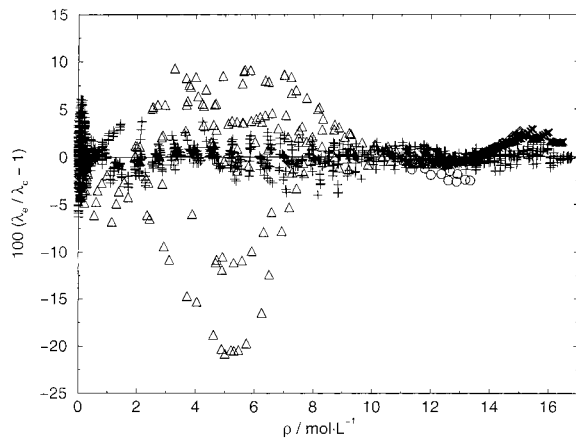
$$\Delta\lambda_c = \frac{\rho C_p R k_b T}{6\pi\eta\xi} (\bar{\Omega} - \bar{\Omega}_0) \quad (9)$$

**Table 5. Coefficients for the Representation of the Residual Thermal Conductivity of Propane in Eq 7 with the Empirical Critical Enhancement of Eq 8**

$\Delta\lambda_r$			$\Delta\lambda_c$	
	$B_{i,1}/(\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1})$	$B_{i,2}/(\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1})$	$C_i$	
$i = 1$	$-3.511\ 52 \times 10^{-2} \pm 1.4977 \times 10^{-3}$	$4.691\ 95 \times 10^{-2} \pm 1.4392 \times 10^{-3}$	$i = 1$	$(3.664\ 86 \times 10^{-4} \pm 3.0597 \times 10^{-5})/\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$
$i = 2$	$1.708\ 90 \times 10^{-1} \pm 4.5444 \times 10^{-3}$	$-1.486\ 16 \times 10^{-1} \pm 3.7486 \times 10^{-3}$	$i = 2$	$-2.216\ 96 \times 10^{-3} \pm 2.4663 \times 10^{-3}$
$i = 3$	$-1.476\ 88 \times 10^{-1} \pm 4.5872 \times 10^{-3}$	$1.324\ 57 \times 10^{-1} \pm 3.4307 \times 10^{-3}$	$i = 3$	$2.642\ 13 \times 10^0 \pm 8.6017 \times 10^{-2}$
$i = 4$	$5.192\ 83 \times 10^{-2} \pm 1.7262 \times 10^{-3}$	$-4.856\ 36 \times 10^{-2} \pm 1.2398 \times 10^{-3}$		
$i = 5$	$-6.186\ 62 \times 10^{-3} \pm 2.1698 \times 10^{-4}$	$6.604\ 14 \times 10^{-3} \pm 1.5740 \times 10^{-4}$		

**Table 6. Coefficients for the Representation of the Residual Thermal Conductivity of Propane in Eq 7 with the Simplified Crossover Critical Enhancement of Eqs 9 to 12**

$\Delta\lambda_r$			$\Delta\lambda_c$
	$B_{i,1}/(\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1})$	$B_{i,2}/(\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1})$	$\bar{q}_D^{-1}$
$i = 1$	$-3.695\ 00 \times 10^{-2} \pm 1.5105 \times 10^{-3}$	$4.827\ 98 \times 10^{-2} \pm 1.4568 \times 10^{-3}$	$(7.166\ 35 \times 10^{-10} \pm 2.669\ 87 \times 10^{-11})/\text{m}$
$i = 2$	$1.486\ 58 \times 10^{-1} \pm 4.9570 \times 10^{-3}$	$-1.356\ 36 \times 10^{-1} \pm 3.8798 \times 10^{-3}$	
$i = 3$	$-1.199\ 86 \times 10^{-1} \pm 5.1882 \times 10^{-3}$	$1.175\ 88 \times 10^{-1} \pm 3.6186 \times 10^{-3}$	
$i = 4$	$4.124\ 31 \times 10^{-2} \pm 1.9559 \times 10^{-3}$	$-4.369\ 11 \times 10^{-2} \pm 1.2884 \times 10^{-3}$	
$i = 5$	$-4.869\ 05 \times 10^{-3} \pm 2.4400 \times 10^{-4}$	$6.160\ 79 \times 10^{-3} \pm 1.5893 \times 10^{-4}$	

**Figure 7.** Relative deviations between the present measurements (+) and the primary literature data: O, Yata;<sup>15</sup> ●, Clifford et al.;<sup>18</sup> ×, Roder and Nieto de Castro;<sup>19,20</sup> Δ, Tufeu and Le Neindre;<sup>21</sup> and ◇, Prasad et al.<sup>22</sup> for the thermal conductivity of propane and the correlation with the empirical expression for the critical enhancement of eqs 5, 7, and 8.

with

$$\bar{\Omega} = \frac{2}{\pi} \left[ \left( \frac{C_p - C_v}{C_p} \right) \arctan(\bar{q}_D \xi) + \frac{C_v}{C_p} \bar{q}_D \xi \right] \quad (10)$$

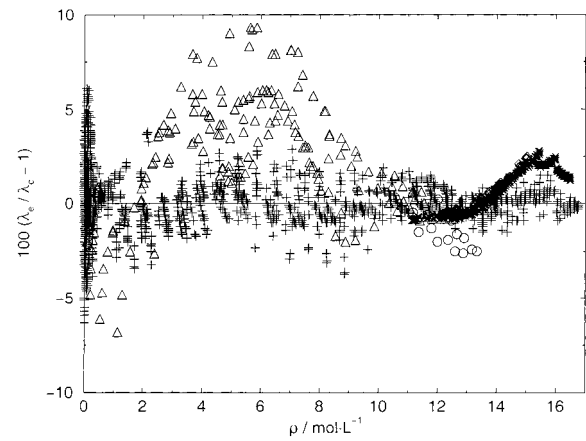
and

$$\bar{\Omega}_0 = \frac{2}{\pi} \left\{ 1 - \exp \left[ - \frac{1}{(\bar{q}_D \xi)^{-1} + (\bar{q}_D \xi \rho_c / \rho)^2 / 3} \right] \right\} \quad (11)$$

In eqs 9–11,  $k_B$  is Boltzmann's constant,  $\bar{\eta}$  is the background viscosity, and  $\bar{q}_D$  is the effective wavenumber cutoff determined by fitting thermal conductivity data in the critical region. The correlation length  $\xi$  is given by

$$\xi = \xi_0 \left[ \frac{P_c \rho}{\Gamma \rho_c^2} \right]^{\nu/\gamma} \left[ \frac{\partial \rho(T, \rho)}{\partial P} \Big|_T - \left( \frac{T_r}{T} \right) \frac{\partial \rho(T_r, \rho)}{\partial P} \Big|_T \right]^{\nu/\gamma} \quad (12)$$

The coefficients of eq 5 were fixed while the coefficients of eq 7 and eqs 9–12 were fit with ODRPACK V. 2.01<sup>38</sup> to the present transient and steady-state data for the thermal conductivity of propane. This crossover model requires the universal constants  $R = 1.03$ ,  $\nu = 0.063$ , and  $\gamma = 1.239$ ,

**Figure 8.** Relative deviations between the present measurements (+) and the primary literature data [O, Yata;<sup>15</sup> ●, Clifford et al.;<sup>18</sup> ×, Roder and Nieto de Castro;<sup>19,20</sup> Δ, Tufeu and LeNeindre;<sup>21</sup> and ◇, Prasad et al.<sup>22</sup>] for the thermal conductivity of propane and the correlation with the simplified theoretical expression<sup>37</sup> for the critical enhancement of eqs 5, 7, and 9–12.

the system-dependent amplitudes  $\Gamma = 0.0496$  and  $\xi_0 = 1.94 \times 10^{-10}$  m, critical-point properties including the critical pressure  $P_c = 4.247\ 66$  MPa, and a reference temperature where the critical enhancement is negligible,  $T_r = 3/2 T_c = 554.73$  K. Table 6 lists the optimum coefficients for eq 7 and eqs 9–12, together with their uncertainties at a level of confidence of one standard deviation. Figure 8 shows a plot of deviations of the primary experimental data from the surface correlation calculated with this simplified theory for the representation of the critical enhancement. Around the critical region, despite the lack of a crossover equation of state, the data are reproduced within  $\pm 5\%$  at a level of uncertainty of 95%, although a systematic trend is observed in that region for the two near-critical isotherms of Tufeu and LeNeindre.<sup>28</sup>

**Full Crossover Model.** The full crossover equations proposed by Olchowy and Sengers<sup>34–36</sup> were also fit to the propane data. The critical enhancement of the thermal conductivity is given by

$$\Delta\lambda_c = \frac{\rho C_p R k_b T}{6\pi \bar{\eta} \xi} (\Omega - \Omega_0) \quad (13)$$

with

**Table 7. Coefficients for the Representation of the Residual Thermal Conductivity of Propane in Eq 7 with the Full Crossover Critical Enhancement of Eqs 13 to 19**

	$\Delta\lambda_r$		$\Delta\lambda_c$
	$B_{i,1}/(\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1})$	$B_{i,2}/(\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1})$	$q_{D-1}$
$i = 1$	$-3.615\ 22 \times 10^{-2} \pm 1.6134 \times 10^{-3}$	$4.679\ 24 \times 10^{-2} \pm 1.5546 \times 10^{-3}$	$(6.281\ 80 \times 10^{-10} \pm 2.6500 \times 10^{-11})/\text{m}$
$i = 2$	$1.659\ 14 \times 10^{-1} \pm 5.2523 \times 10^{-3}$	$-1.430\ 28 \times 10^{-1} \pm 4.1350 \times 10^{-3}$	
$i = 3$	$-1.403\ 09 \times 10^{-1} \pm 5.4692 \times 10^{-3}$	$1.260\ 92 \times 10^{-1} \pm 3.8516 \times 10^{-3}$	
$i = 4$	$4.882\ 10 \times 10^{-2} \pm 2.0580 \times 10^{-3}$	$-4.637\ 39 \times 10^{-2} \pm 1.3729 \times 10^{-3}$	
$i = 5$	$-5.781\ 46 \times 10^{-3} \pm 2.5663 \times 10^{-4}$	$6.393\ 52 \times 10^{-3} \pm 1.6969 \times 10^{-4}$	

$$\Omega = \frac{2}{\pi} \left( \frac{1}{1 + y_\gamma} \right) \left[ y_D - \sum_{i=1}^4 \left( \frac{a_3 z_i^3 + a_2 z_i^2 + a_1 z_i + a_0}{\prod_{j=1, j \neq i}^4 (z_i - z_j)} \right) F(z_i, y_D) \right] \quad (14)$$

and

$$\Omega_0 = \frac{1 - \exp\{-[(q_D \xi)^{-1} + (q_D \xi \rho_c / \rho)^2 / 3]^{-1}\}}{\frac{\pi}{2} [1 + y_\alpha (y_D + y_\delta) + y_\beta (1 + y_\gamma)^{-1}]} \quad (15)$$

The correlation length  $\xi$  is again given by eq 12 as for the simplified crossover model, but  $q_D$  is the effective wavenumber cutoff ( $\bar{q} \neq q_D$ , since each effective wavenumber cutoff is model dependent) and is determined by a fit of thermal conductivity data in the critical region. The coefficients  $a_i$  in eq 14 are given by

$$\begin{aligned} a_0 &= y_\gamma^2 - y_\alpha y_\gamma y_\delta \\ a_1 &= y_\alpha y_\gamma y_D \\ a_2 &= y_\gamma - y_\beta - y_\alpha y_\delta \\ a_3 &= y_\alpha y_D \end{aligned} \quad (16)$$

In eq 14, the  $z_i$  are the two real and two complex-conjugate roots of the equation

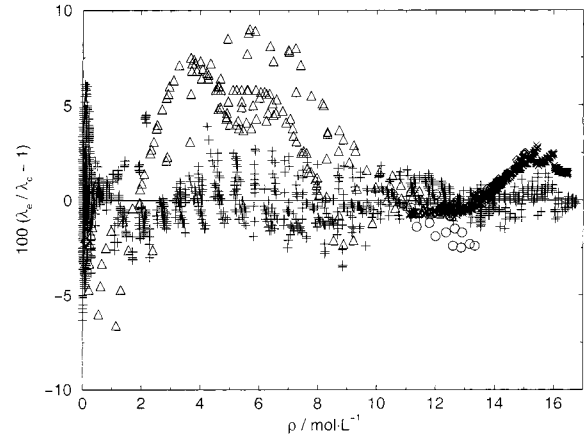
$$z^4 + y_\alpha y_D z^3 + (y_\gamma + y_\beta + y_\alpha y_\delta) z^2 + y_\alpha y_\gamma y_D z^2 + y_\alpha y_\gamma y_\delta z = 0 \quad (17)$$

The function  $F$  in eq 14 is given by

$$F(x, y_D) = \frac{1}{(1 - x^2)^{1/2}} \ln \left[ \frac{1 + x + (1 - x^2)^{1/2} \tan(y_D/2)}{1 + x - (1 - x^2)^{1/2} \tan(y_D/2)} \right] \quad (18)$$

The auxiliary functions are given by

$$\begin{aligned} y_D &= \arctan(q_D \xi) \\ y_\delta &= \frac{\arctan[q_D \xi / (1 + q_D^2 \xi^2)^{1/2}] - y_D}{(1 + q_D^2 \xi^2)^{1/2}} \\ y_\alpha &= \rho k_B T / 8 \pi \bar{\eta}^2 \xi \\ y_\beta &= \bar{\lambda} / \bar{\eta} (C_p - C_v) \\ y_\gamma &= C_v / (C_p - C_v) \\ y_\eta &= (y_\delta + y_\beta / y_\alpha) / y_D \quad \text{and} \\ y_v &= y_\gamma y_\delta / y_D \end{aligned} \quad (19)$$



**Figure 9.** Relative deviations between the present measurements (+) and the primary literature data [O, Yata;<sup>15</sup> ●, Clifford et al.;<sup>18</sup> ×, Roder and Nieto de Castro;<sup>19–20</sup> △, Tufeu and LeNeindre;<sup>21</sup> ◇, Prasad et al.<sup>22</sup>] for the thermal conductivity of propane and the correlation with the full theoretical expression<sup>34–36</sup> for the critical enhancement of eqs 5, 7, and 13–19.

The coefficients of eq 5 were fixed while the coefficients of eq 7 and eqs 13–19 were fit with ODRPACK V. 2.01 to the present transient and steady-state data for the thermal conductivity of propane. This crossover model requires the universal constants  $R = 1.03$ ,  $z = 0.063$ , and  $\gamma = 1.239$ , the system-dependent amplitudes  $\Gamma = 0.0496$  and  $\xi_0 = 1.94 \times 10^{-10}$  m, and a reference temperature where the critical enhancement is negligible,  $T_R = 3/2 T_c = 554.73$  K. Table 7 lists the optimum coefficients for eq 7 and eqs 13–19 together with their uncertainties at a level of confidence of one standard deviation. Figure 9 shows a plot of deviations of the experimental data from the surface correlation calculated with this theory for the representation of the critical enhancement. It can be seen that, around the critical region and despite the lack of a crossover equation of state, the data are reproduced within  $\pm 5\%$  at a level of uncertainty of 95%, although a systematic trend is observed in that region for the two near-critical isotherms of Tufeu and LeNeindre,<sup>21</sup> similar to the case of the simplified model.

## Discussion

Three alternative correlations are developed for the thermal conductivity of propane that are valid over the temperature range from 86 K to 600 K with densities up to  $17 \text{ mol}\cdot\text{L}^{-1}$  ( $750 \text{ kg}\cdot\text{m}^{-3}$ ). These three correlations differ only in the expression used to account for the enhancement of the thermal conductivity in the critical region. The simple empirical expression is easy to implement and fits the present data quite well but is shown to have difficulty in dealing with the near-critical isotherms of Tufeu and LeNeindre.<sup>21</sup> A simplified crossover theory<sup>37</sup> for the enhancement of the thermal conductivity is also relatively easy to implement, fits the present data well, and also predicts the enhanced thermal conductivity of the near-

critical isotherms of Tufeu and LeNeindre.<sup>21</sup> The full crossover theory<sup>34–36</sup> for the enhancement of the thermal conductivity does not appear to offer improved performance over the simplified theory and is significantly more difficult to implement. The current limitation in theoretically accounting for the critical enhancement of propane is the lack of a crossover equation of state that is valid in the critical region. It is possible that the full theory would offer improved performance when used with such a crossover equation of state.

All three of the alternative correlations described above are shown to provide a significantly improved representation of the thermal conductivity of propane relative to literature correlations.<sup>1–3</sup> On the basis of simplicity and performance, the correlation based on the simplified crossover theory for the thermal conductivity enhancement in the critical region is recommended relative to the other two formulations. The simplified crossover model fits all of the present steady-state and transient data, as well as the primary literature data, within  $\pm 5\%$  at a level of uncertainty of 95%. This is particularly impressive, since the primary data sets, some of which are much closer to the critical point, were not used during the fitting process.

## Conclusions

A total of 393 steady-state and 1380 transient hot-wire measurements of the thermal conductivity of propane are reported at temperatures from 86 K to 600 K with pressures to 70 MPa. These data are estimated to have an uncertainty of  $\pm 1\%$  for measurements removed from the critical point and at pressures above 1 MPa, increasing to  $\pm 3\%$  in the critical region and  $\pm 4\%$  at low pressures ( $< 1$  MPa) at a 95% confidence level. This is a significant extension to the literature data that are available for the thermal conductivity of propane in terms of both the range of temperature and pressure studied and the reduction of uncertainty. Three alternative correlations are developed on the basis of this new data, valid over the temperature range from 86 K to 600 K with densities up to  $17 \text{ mol}\cdot\text{L}^{-1}$  ( $750 \text{ kg}\cdot\text{m}^{-3}$ ). The simplified crossover model is recommended, since it fits all of the present steady-state and transient data, as well as the primary literature data, for the thermal conductivity of propane within  $\pm 5\%$  at a level of uncertainty of 95% at temperatures from 86 K to 600 K with pressures to 70 MPa, while remaining relatively simple to implement.

## Supporting Information Available:

Tabulated experimental data (43 pages). This material is available free of charge via the Internet at <http://pubs.acs.org>.

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