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Status of the Round Robin on the Transport Properties of R134a[1]

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The paper contains a status report on an international project coordinated by the Subcommittee on Transport Properties of Commission I.2 of the International Union of Pure and Applied Chemistry. The project has been conducted to investigate the large discrepancies between the results reported by various authors for the transport properties of R134a. The project has involved the remeasurement of the transport properties of a single sample of R134a in nine laboratories throughout the world in order to test the hypothesis that at least part of the discrepancy could be attributed to the purity of the sample. This paper provides an intercomparison of the new experimental results obtained to data in this project for the viscosity and the thermal conductivity in both gaseous and liquid phases. The agreement between the viscosity data from the laboratories contributing to the project was improved with several techniques, now producing consistent results. This suggests that the purity of the samples of R134a used in previous work was at least partly reponsible for the discrepancies observed. For the thermal conductivity in the liquid phase the results of the measurements are also more consistent than before, although not for all experimental techniques. Not all of the previous measurements suffered from

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significant sample impurities, so the present measurements on a consistent high-purity sample can be used to detect data sets which are outliers, possibly because of impurities. Identification of laboratories and techniques with systematic differences may require the examination of data for several fluids. The implications for future measurements of the transport properties of other refrigerants are significant.

KEY WORDS: R134a; dilute gas; refrigerant; saturation properties; 1,1,1,2-tetrafluoroethane; thermal conductivity; transport properties; viscosity.

1. INTRODUCTION

The damage to the onzone layer caused by the use of refrigerants containing chlorine was recognized some 10 years ago. This recognition prompted a search for replacement materials that were less harmful to the atmosphere and resulted in consideration of the group of hydrofluorocarbons (HFC's) as suitably materials since they contained no chlorine. Among this group of compounds preliminary studies indicated that R134a (1,2,2,2-tetrafluoroethane) was a suitable substitute material for R12 which had been used traditionally in domestic refrigeration. This identification prompted a series of measurements in many laboratories throughout the world of the thermophysical properties of this material including equilibrium and transport properties. For the thermodynamic properties, the studies that ensued provided results that suffered from only mild inconsistencies which were resolved by a group of collaborators working under the auspices of Annex 18 of the International Energy Agency. Their efforts have culminated in a definitive equation of state for R134a [1]. From the point of view of industrial design of refrigeration cycles, the thermodynamic properties of the refrigerant are more significant than the transport properties, so that work on the transport properties was initiated more slowly. In addition, it has transpired that the measurements suffered from far greater difficulties than had been expected.

At the Eleventh Symposium on Thermophysical Properties held in 1991, two independent reports of measurements of the viscosity of R134a were made and subsequently published by Diller et al. [2] and Fellows et al. [3] using different experimental techniques. The results of these two initial studies were very different; deviations of as much as 15% along the saturation line were apparent. This observation, combined with the increasing demand for viscosity and thermal conductivity data, prompted new studies of this and several other potential replacement refrigerants including R32 and R125.

In November of 1992, a meeting of some of those interested in the thermophysical properties of refrigerants, in particular the transport properties, was held in Ericeira, Portugal, to review the situation with respect to new measurements. The conclusion of that meeting was that discrepancies remained that greatly exceeded the experimental uncertainties claimed by individual authors. It was therefore agreed to launch a project, which initially involved nine laboratories, to elucidate the problems. At the time of writing, seven laboratories have reported new measurements on a uniform sample of R134a. The present paper compares the results obtained on this round-robin sample among themselves and with results of earlier work.

2. BACKGROUND

The saturated-liquid viscosity data [2–8] which were available for R134a in 1992 are shown in the deviation plot in Fig. 1. For comparison, the viscosity correlation of Krauss et al. [9] is used as the baseline in Fig. 1, even though it is not recommended for use below 290 K for viscosity (dashed baseline in figure). The uncertainty in the correlation [9] is estimated to be ±5% as shown in Fig. 1. The results included in the figure were obtained by a variety of techniques including capillary viscometers, vibrating-wire viscometers, and one oscillating quartz-crystal viscometer. The maximum deviation between the data sets [2–8] is 33% at 340 K and far exceeds the mutual uncertainty claimed by the authors and the

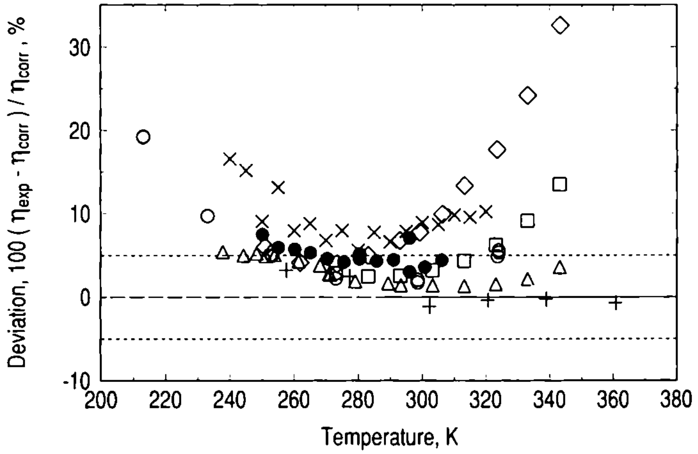

Fig. 1. Deviations of the viscosity of R134a in the liquid phase at saturation available in 1992 from the correlation of Krauss et al. [9]. ×, Diller et al. [2]; ○, Fellows et al. [3]; △, Oliveira and Wakeham [4]; ●, Ripple and Matar [5]; □, Kumagai and Takahashi [6]; +, Ruvinski et al. [7]; , Okubo et al. [8].

uncertainty of the baseline correlation. The results of Diller et al. [2] and Fellows et al. [3] appear to be systematically higher than the other results available in 1992. An examination [4] of the pressure dependence of the viscosity of R134a revealed a very much better agreement, although the absolute differences observed at saturation were maintained.

A review of the results for the dilute gas-phase viscosity [10] showed deviations of as much as 4% among the three data sets which were available at the time and obtained with two different types of viscometers. Again, this deviation substantially exceeded the mutual uncertainty claimed by the authors.

The data sets for the viscosity of R32 and R125 were also examined [11] to determine whether this problem was specific to R134a. Figure 2 contains the comparison among the three sets of results then available for R125 along the saturation line. The discrepancies exceed 15%. The representation of the data of Oliveira and Wakeham [11] has been used as the baseline, and the figure includes the results of Ripple and Matar [5] with a capillary viscometer, Oliveira and Wakeham [11] with a vibrating-wire viscometer, and Diller and Peterson [13] with an oscillating quartz-crystal viscometer. It appears that the problem is a general one which is not limited to the case of R134a.

Viscosity discrepancies of this magnitude are at best embarrassing and at worst damaging to those interested in the viscosity of liquids. Possible explanations include improper working equations for one or more

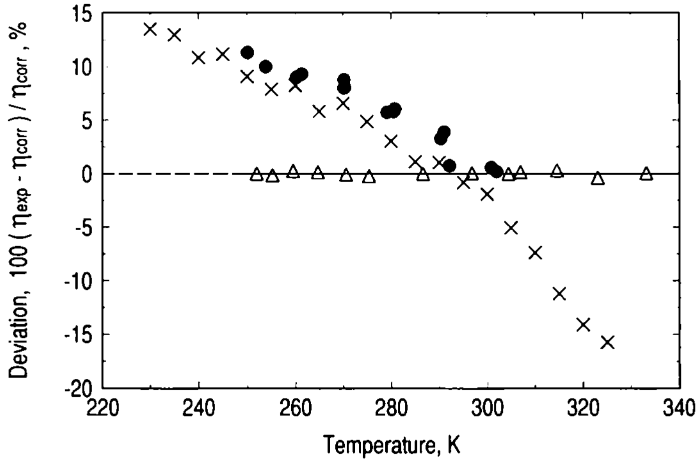

Fig. 2. Deviations of the viscosity of R125 in the liquid phase at saturation available in 1992 from the correlation of Oliveira and Wakeham [12]. △, Oliveira and Wakeham [12]; ●, Ripple and Matar [5]; ×, Diller and Peterson [13].

of the techniques, improper data analysis, carelessness on the part of the researchers, undetected equipment malfunctions, and differences in the chemical composition of the material studied.

For the thermal conductivity of R134a, the situation in 1992 was almost as serious in the liquid phase. Figure 3 contains a plot of the deviations of the thermal conductivity data [3, 7, 14–18] for liquid R134a along the saturation line from the correlation of Krauss et al [9], which has been employed as the baseline. The uncertainty in the correlation [9] is estimated to be ±5% for thermal conductivity as shown in Fig. 3. Below 300 K the agreement between the data and the correlation is within ±2.5%. This is interesting since the baseline correlation [9] is not considered valid below 240 K. The maximum deviations are about 20% in this case and occur near the critical temperature, but oddly this is the region where the correlation is considered valid. This value exceeds the mutual uncertainty claimed by the authors by almost an order of magnitude in some cases. In the vapor phase the few measurements available in 1992, mostly at low density, disagreed by as much as 15% [9]. Almost all of these data were obtained with the transient hot-wire method, although marginally different versions of the instrument were employed. These observations obviously threw doubt on the ability of some, if not all, of the

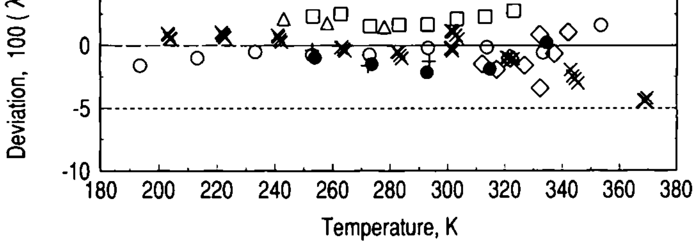

Fig. 3. Deviations of the thermal conductivity of liquid R134a along the saturation line available in 1992 from the correlation of Krauss et al. [9]. ×, Laesecke et al. [14]; △, Ross et al. [15]; □, Gross et al. [16]; ○, Ueno et al. [17]; ◇, Fellows et al. [3]; +, Ruvinski et al. [7]; ●, Yata et al. [18].

laboratories concerned, to make accurate thermal conductivity measurements, because either the experimental techniques were not understood, or the sample was not adequately pure, or both.

3. ROUND-ROBIN PROJECT

The issues raised in the study of R134a and other refrigerants such as R125 have potentially far wider consequences because it appeared that it was not possible to determine accurately even the viscosity of a liquid under ambient conditions (a task frequently assigned to undergraduate students). The threat to the credibility of future measurements of viscosity and thermal conductivity was significant. It is essential to identify the source or sources of the discrepancies between the results of various authors. As one part of this effort the problem of the purity of the samples must be addressed. Almost all experimenters had used samples supplied by different companies and the purity, as supplied, was not always specified. Furthermore, water was a potential impurity in samples of R134a, which even in trace amounts could have significant consequences for the viscosity of liquids and could also have a serious effect upon the electrical measurements associated with the transient hot-wire technique. The only way in which sample purity could be investigated in a satisfactory manner was if various laboratories undertook measurements of the viscosity and/or thermal conductivity of R134a on the *same sample* with their own equipment. It was impractical to circulate the same sample among different laboratories since it is very time-consuming and there is significant danger of losing or contaminating the sample as it circulates among laboratories. The present authors decided that a single source of supply of R134a should be selected and sufficient samples taken from that single source of supply to provide each laboratory with its own cylinder.

One company agreed to provide the single source of R134a and filled nine cylinders from that same source, using procedures for pharmaceutical materials to ensure the cleanliness of the cylinders before use. The purity of the batch was confirmed by gas chromatography for organic impurities and the Karl-Fischer test for water. The purity of the batch was greater than 99.9%, and the principal impurity was R134 at a concentration of 850 ppm. Water was present at a concentration of 6 ppm. These nine cylinders of material were distributed in July 1993 to nine laboratories throughout the world that had volunteered to participate in the project in 1992. The laboratories were asked to conduct measurements of the transport properties using their preferred technique on the sample, taking care to maintain the purity of the sample and to recover the sample after use for

Table I. Summary of the Measurements of Transport Properties Made on the Round-Robin Sample of R134a

Primary contact	Viscosity measurements	Thermal Conductivity measurements
M. J. Assael, Aristotle University assael@aliakmon.cperi.forth.gr	Vibrating wire (liquid) Polimatidou (saturation, compressed)	Transient hot-wire (liquid–Ta wire) Karagiannidis (saturation, compressed)
Y. Nagasaka, Keio University nagasaka@mech.keio.ac.jp		Transient hot-wire (liquid–Pt wire) Yamada (saturation, compressed)
C. A. Nieto de Castro, Universidade de Lisboa qcastro@cc.fc.ul.pt		Transient hot-wire (liquid–Pt wire) Gurova (saturation, compressed)
R. A. Perkins, NIST Thermophysics Division perkins@bldr.nist.gov		Transient hot-wire (liquid–Ta wire) Perkins (Saturation, compressed)
K. Ström, Chalmers University of Technology	Höppler falling body (liquid) Ström (compressed)	
E. Vogel, Universität Rostock eckhard.vogel@chemie.uni-rostock.d400.de	Oscillating disk (gas) Wilhelm (dilute limit)	
W. A. Wakeham, Imperial College w.wakeham@ic.ac.uk	Vibrating wire (liquid) Oliveira and Fenghour (saturation) Wilhelm (compressed)	

subsequent analysis to ensure that the purity had not been degraded. All of the laboratories also agreed to use the equation of state for R134a reported by Tillner-Roth and Baehr [1]. Since the work of all of these laboratories has been entirely voluntary, it has not proved possible for all of them to carry out all of the measurements intended at this stage. The next section therefore provides a comparison of the results available to date. The new measurements on the round-robin sample are summarized in Table I. None of the data on the round-robin sample have been published, but the primary contact listed in Table I can provide additional details and the individual data points.

4. COMPARISONS FOR THE ROUND-ROBIN SAMPLE

The comparisons between the new experimental data are made simpler by the adoption of a uniform baseline for the comparison. We adopt here the representation of the transport properties of R134a provided by Krauss et al. [9] as used in Figs. 1 and 3 for the earlier data. The fact that this correlation has large uncertainties, owing to the very discrepancies among the data upon which it is based and identified above, is unimportant for this purpose. We have also extrapolated the correlation of Krauss et al. [9] downward in temperature from its original lower limits of 290 K (viscosity) and 240 K (thermal conductivity) to 200 K. Again, this extrapolation has no consequences for the comparison which is the principal matter of interest here.

4.1. Liquid-Phase Viscosity at Saturation

Using the round-robin sample the liquid-phase viscosity at saturation has been measured independently by Oliveira and Fenghour at Imperial College in London and Polimatidou at the University of Thessaloniki in Greece. The two sets of measurements have been performed with similar vibrating-wire viscometers, although there are differences in the cell geometry and electronics between the viscometers in the two locations. Both of these groups estimate that the viscosity measurements have an accuracy of ±0.5%, largely as a result of the need for calibration against standard reference values. Figure 4 compares the results of the two sets of measurements with the correlation of Krauss et al. [9]. Over the temperature range 240 to 340 K the maximum spread of the results is approximately ±1%, although the results all lie substantially above the correlation of Krauss et al. [9]. The spread of results among the measurements on the round-robin sample is considered consistent with the

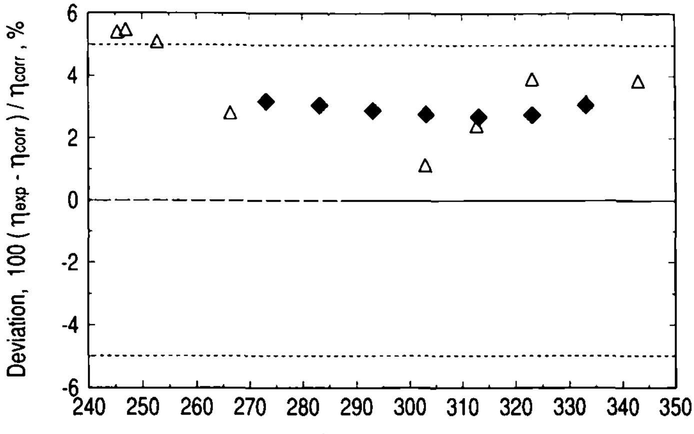

Fig. 4. Deviations of the measurements of the viscosity of liquid R134a along the saturation line measured on the round-robin sample from the correlation of Krauss et al. [9]. ◆, Polimatidou; △, Oliveira and Fenghour.

claimed mutual uncertainty of these two instruments. It is noteworthy that the results from these two laboratories are also consistent with their earlier results on different samples. This indicates that the purity of the samples used in these previous vibrating-wire studies was comparable to that of the present round-robin sample and did not affect the measured viscosity. Figures 1 and 4 taken together reveal that the new data on the round-robin sample generally fall in the middle of all the previous data, with the exception of the torsional crystal results of Diller et al. [2] and the capillary results of Fellows et al. [3], which are both systematically higher. In these two cases, impurities could have introduced a measurement error or could have altered the actual viscosity of the fluid. When both Fig. 1 and Fig. 4 are considered together and the two data sets mentioned above are considered outliers [2, 3], there is agreement to within ±3% using both capillary and vibrating-wire viscometers for the saturated-liquid viscosity of R134a from 240 K to 320 K with both the round-robin sample and the samples used in previous work around the world. Above 340 K the scatter in the previous data increases to ±5%.

4.2. Liquid-Phase Viscosity at Elevated Pressures

The viscosity of the round-robin sample of R134a at elevated pressures has been measured along isotherms by Wilhelm in London at 300 and 320 K and by Polimatidou in Thessaloniki at 273, 293, 313, and 333 K,

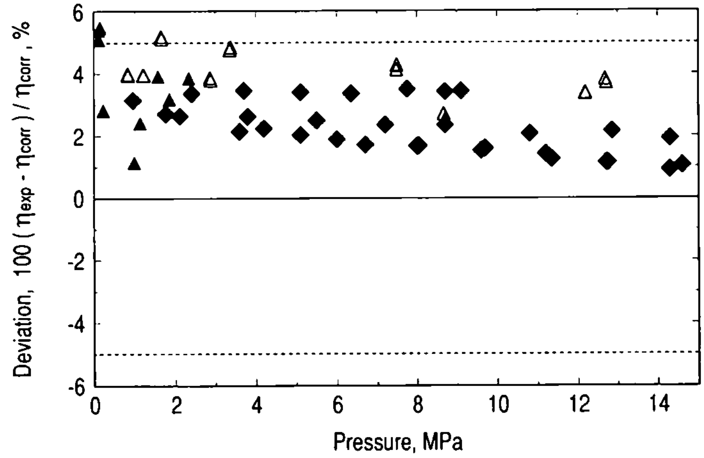

Fig. 5. Deviations of the measurements of the viscosity of liquid R134a at elevated pressure measured on the round-robin sample from the correlation of Krauss et al. [9]. ◆, Polimatidou; △, Wilhelm; ▲, Ström.

both with vibrating-wire viscometers. Again, the estimated accuracy of the two sets of measurements is ±0.5%. Ström in Gothenburg has also made measurements with a Höppler, falling-body viscometer with an estimated accuracy of ±2%. The results of Ström are at temperatures of 260, 268, 278, 288, 298, 308, and 318 K. Figure 5 compares the results of the three sets of measurements with the correlation of Krauss et al. [9] as a function of pressure. The comparison is confined to pressures below 15 MPa where the sets of data overlap. The deviations between the two vibrating-wire viscometers and the falling-body viscometer are generally within ±3%, and the pressure dependence is observed is similar. All of the data agree within their mutual uncertainty and are consistent with the saturated-liquid values.

4.3. Viscosity in the Dilute Gas

The only set of measurements on the vapor-phase viscosity of the round-robin sample so far available was made by Wilhelm in the oscillating-disk viscometer at the University of Rostock. The measurements were performed on R134a at pressures such that the values derived are those appropriate to the dilute-gas limit (zero density) over the temperature range 290 to 450 K. It is estimated that the accuracy of the viscosity data is ±0.5%. Although a comparison of the values with others on the round-robin sample is not possible, it is interesting to compare the results with the

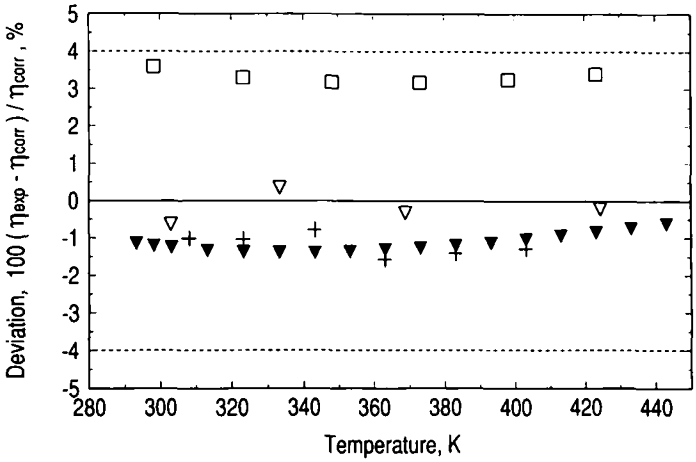

Fig. 6. Deviations of the measurements of the viscosity of R134a in the dilute-gas state from the correlation of Krauss et al. [9]. ▼, Wilhelm; +, Dowdell and Matthews [10]; ∇, Nabizadeh and Mayinger [19]; □, Takahashi et al. [20].

previous results available in the literature [10, 19, 20]. Figure 6 shows these deviations relative to the correlation of Krauss et al. [9]. The data of Dowdell and Matthews [10] obtained with a capillary viscometer and the data of Nabizadeh and Mayinger [19] obtained with an oscillating-disk viscometer agree with the new oscillating-disk measurements to within their mutual uncertainty. The oscillating-disk data of Takahashi et al. [20] are systematically 4% higher than the present results of Wilhelm. The sample of R134a used by Takahashi was probably impure since all of these instruments generally agree within their mutual uncertainty for dilute-gas viscosity.

4.4. Thermal Conductivity in the Liquid Phase at Saturation

Measurements of the thermal conductivity of R134a in the liquid phase at saturation have been made with the transient hot-wire technique by Karagiannidis in Thessaloniki using a double-wire cell with anodised tantalum wires for electrical insulation (uncertainty claimed, ±0.5%), by Perkins at NIST in Boulder using the same method (uncertainty claimed, ±1.0%), by Gurova in the University of Lisbon using a double-wire cell with bare platinum wires operated with a polarization voltage (uncertainty claimed, ±0.5%), and by Yamada at Keio University using a cell with a single bare platinum wire with potential leads (uncertainty claimed, ±0.5%). Figure 7 compares the results of these measurements with the representation of Krauss et al. [9]. The measurements of Yamada by means of the single-wire cell with potential taps is shown to be systematically low by up to 5% at temperatures above 220 K. This new data set is also inconsistent with the previous data set from this laboratory [17] on a different sample as shown in Fig. 3. Yamada noted very significant electrical conduction through their sample of the round-robin R134a during their measurements. This electrical conduction increased dramatically with temperature and may be the source of the error at high temperatures since their hot wires are not electrically insulated. Since very pure refrigerants normally do not have significant electrical conduction, there probably was some impurity in their cell during the measurements. It is not yet clear whether the sample bottle for the Keio sample was contaminated or whether the Keio measurement cell was contaminated. The contents of the Keio sample are being analyzed for impurities and a sample is being sent from the NIST bottle for another set of measurements. Excluding the Yamada data set, the spread of values is generally about ±1.5%, which is consistent with the claimed uncertainties and is a significant improvement over the situation illustrated in Fig. 3 at higher temperatures.

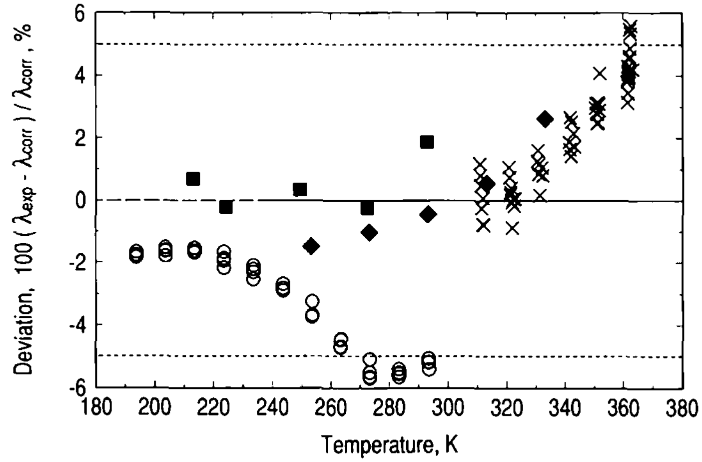

Fig. 7. Deviations of the measurements of the thermal conductivity of liquid R134a along the saturation line measured on the round-robin sample from the correlation of Krauss et al. [9]. ◆, Karagiannidis, ×, Perkins; ■, Gurova; ○, Yamada.

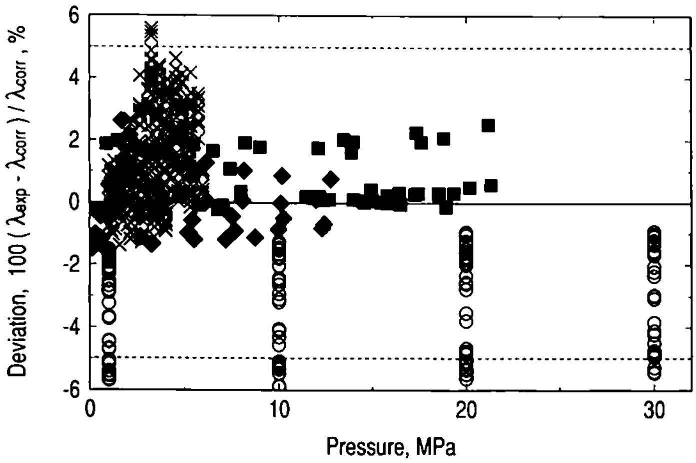

Fig. 8. Deviations of the measurements of the thermal conductivity of liquid R134a at elevated pressure measured on the round-robin sample from the correlation of Krauss et al. [9]. ◆, Karagiannidis, ×, Perkins; ■, Gurova; ○, Yamada.

4.5. Thermal Conductivity of the Liquid Under Pressure

Transient hot-wire measurements of the thermal conductivity of R134a on the round-robin sample have been conducted by Karagiannidis in Thessaloniki and Perkins at NIST in Boulder using cells with double anodized-tantalum wires, Gurova at University of Lisbon using a cell with double bare platinum wires, and Yamada at Keio University using a cell with a single platinum wire and potential leads. Figure 8 compares the selected results of all of these measurements with the representation of Krauss et al. [9] as a function of pressure along several isotherms. The three sets of measurements made with the two-wire cells are mutually consistent within a band of approximately ±3%. In this case the larger uncertainty is due to the deficiencies of the correlation [9] used as a baseline but is still acceptable since it is within the ±5% uncertainty band claimed for the correlation. The correlation is increasingly low as the temperature is increased above 300 K, as shown in Fig. 7 at saturation. As with the saturation measurements, the measurements made with the single wire by Yamada lie systematically 4 to 5% below the other results near 300 K.

4.6. Thermal Conductivity in the Dilute Gas

There have been no new measurements of the vapor phase thermal conductivity of R134a with the round-robin sample. This is unfortunate because the recent analysis of Krauss et al. [9] indicates that the discrepancies between the results of various authors amount to as much as 20% as shown in Fig. 9 of Ref. 9. It is not yet possible to say anything about this problem.

5. SUMMARY

On the basis of the foregoing material, it is concluded that measurements made of the dilute gas viscosity by Wilhelm on the round-robin sample are consistent with the previous data of Dowdell and Matthews [10] and Nabizadeh and Mayinger [19], who both used different samples of R314a. Purity was probably not a problem with these two viscosity data sets, and capillary measurements [10] agree with oscillating-disk measurements (present work and Ref. 19) within the mutual uncertainty of the measurements. It would be desirable in the future to have other researchers measure the dilute gas viscosity of the round-robin sample of R134a. No measurements of the dilute-gas thermal conductivity are available for comparison at this time.

The very large discrepancies that characterized early liquid-phase viscosity results have not been repeated when an identical sample has been used. There is agreement within mutual uncertainty between liquid viscosity measurements made by means of the vibrating-wire technique and the falling-body technique using the round-robin sample. Additionally, the viscosity data obtained for the round-robin sample and the previous data using the capillary technique [5–8] agree to within ±3% at temperatures below 320 K. Also, the laboratories involved in the round-robin test of the viscosity have reproduced their earlier results within their estimated uncertainty. This would suggest that, while purity may have been, in part, responsible for the differences between the results observed earlier, there may be additional problems associated with the working equations or the operation of some of the instruments. Problems with measurement techniques will likely become more evident through examinations of results for several fluids.

During the round-robin project, each researcher has taken extreme care during the measurements and the subsequent data analysis, using a carefully prepared sample of high purity. Despite these precautions, systematic (temperature-dependent) deviations are observed with a magnitude of 5% in thermal conductivity measured by the transient hot-wire technique. Understanding of this discrepancy is crucial to the round-robin project and the credibility of the transient hot-wire technique. It is obviously desirable to have researchers participate using a variety of experimental techniques including perhaps a steady-state technique and light scattering, so it is unfortunate that, at present, results are available from only three variants of the transient hot-wire technique. If the Yamada data set is excluded because of the electrical conduction problem reported in the Keio sample, the spread of values is generally about $\pm 1.5\%$, which is consistent with the claimed uncertainties of each of the researchers.

The round-robin project will continue for some time to allow other contributors to complete their measurements. This project will continue to examine problems in the measurement of the transport properties of refrigerants with future emphasis on the gas-phase thermal conductivity, which has proved rather more difficult to measure than expected. The completion of this work should enhance the optimal application of new refrigerant materials, increase the understanding of the fluid state for a unique group of polar compounds, and establish the credibility and uncertainty of the techniques which are available to measure the transport properties of fluids.

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