

# Thermal Conductivities of Alternatives to CFC-11 for Foam Insulation

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The present work examines the thermal conductivities of several dilute vapors which are potential alternatives to chlorofluorocarbons (CFCs) for cellular-plastic foams. Thermal conductivity data are reported in the vapor phase, from 280 to 340 K, for trichlorofluoromethane (CFC-11), 1,1,1-trifluoro-2,2-dichloroethane (HCFC-123), 1-fluoro-1,1-dichloroethane (HCFC-141b), 1,1,2-trifluoroethane (HFC-143), 1,1,1,2,3,3,3-heptafluoropropane (HFC-227ea), 1,1,1,2,3,3-hexafluoropropane (HFC-236ea), 1,1,1,3,3,3-hexafluoropropane (HFC-236fa), 1,1,2,2,3-pentafluoropropane (HFC-245ca), 2-(difluoromethoxy)-1,1,1-trifluoroethane (HFE-245fa1), 2-(trifluoromethoxy)-1,1,1-trifluoroethane (HFE-263fb1), octafluorocyclobutane (RC318), 1,1,1-trifluoroacetone, and a 75 mol % cyclopentane + 25 mol % pentane mixture. These data were obtained with a transient hot-wire instrument and are estimated to have an uncertainty of  $\pm 2\%$  ( $2\sigma$  confidence) at these low densities.

## Introduction

Many cellular-foam insulation materials have relied on trichlorofluoromethane (CFC-11) as the gas used to fill the closed cells of the insulation. CFC-11 has many advantages for cellular-plastic foams: it is nontoxic and nonflammable and has a low thermal conductivity relative to that of air. However, the production of chlorofluorocarbons such as CFC-11 has been phased out (1995) under the Montreal Protocol. Hence, efficient, low-cost alternatives must be found. A foam insulation with air in its cells is less effective than a closed-cell foam with CFC-11 in the cells. The effectiveness of a foam-blowing agent as an insulator can be characterized by its thermal conductivity in the gas phase relative to the thermal conductivity of air (typically  $27 \text{ mW}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$  at 300 K) that it displaces. A cellular foam generally ages, with loss of thermal efficiency, as air diffuses into the closed cells and mixes with the blowing agent (typically CFC-11). Alternatives that are considered here include hydrochlorofluorocarbons (HCFCs), hydrofluorocarbons (HFCs), fluorinated ethers (HFEs), and several other chemicals with a low flammability and low vapor pressure. The thermal conductivity of the gas near atmospheric pressure is only one of many criteria that will ultimately determine which gases will be used in this application. Yet, the thermal conductivity of the gas used to fill the closed cells must be carefully considered, since it will directly influence the effectiveness of the cellular-plastic foam as a thermal insulation.

## Experimental Section

The purity of the samples studied was better than 99.7 mol %, with the exception of the cyclopentane + pentane mixture, where the sample composition was 75 mol % cyclopentane + 25 mol % pentane. The purity of each compound was verified by gas chromatography, and the

measured vapor was filled into the measurement cell from the vapor phase of each two-phase sample.

The measurements of thermal conductivity were obtained using a transient hot-wire instrument which has been described in detail.<sup>1</sup> The instrument has dual 12.7  $\mu\text{m}$  diameter hot wires that are made from platinum. The outer cavity around the hot wires is stainless steel and has a diameter of 9 mm. The instrument is capable of operation from 30 K to 340 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 0.01 K, and pressures are determined with a pressure transducer with an uncertainty of 7 kPa. The basic theory which describes the operation of the transient hot-wire instrument is given by Healy et al.<sup>2</sup> The 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

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

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 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,  $\Delta T_{\text{w}}$  is the measured temperature rise of the wire, and  $\delta T_i$  are corrections<sup>2</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 there are two significant corrections<sup>3,4,5,6</sup> which must be carefully considered. First, since the thermal diffusivity of the gas is much different from that of the wire, the correction for the finite wire

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**Table 1. Summary of Gases Studied in This Work<sup>a</sup>**

chemical name	ASHRAE name	$T_{bp}/K$	MW	calculation models
trichlorofluoromethane	CFC-11	297.0	137.37	CS – REFPROP
octafluorocyclobutane	RC318	266.2	200.04	CS – REFPROP
1,1,1-trifluoro-2,2-dichloroethane	HCFC-123	301.1	152.93	MBWR – REFPROP
1-fluoro-1,1-dichloroethane	HCFC-141b	305.4	116.95	CS – REFPROP
1,1,2-trifluoroethane	HFC-143	277.2	84.04	CS – REFPROP
1,1,1,2,3,3,3-heptafluoroethane	HFC-227ea	256.8	170.04	CS – REFPROP
1,1,1,2,3,3,3-hexafluoropropane	HFC-236ea	279.7	152.04	CS – REFPROP
1,1,1,3,3,3-hexafluoropropane	HFC-236fa	272.1	152.04	MBWR – REFPROP
1,1,2,2,3-pentafluoropropane	HFC-245ca	298.4	134.05	CS – REFPROP
2-(difluoromethoxy)-1,1,1-trifluoroethane	HFE245fa1	302.4	150.05	CS – REFPROP
2-(trifluoromethoxy)-1,1,1-trifluoroethane	HFE263fb1	294.9	114.07	ECS – Supertrapp
1,1,1-trifluoroacetone		321.5	112.05	ECS – Supertrapp
75% cyclopentane + 25% pentane		304.5	70.13	ECS – Supertrapp

<sup>a</sup> The equation of state used is designated as the Carnahan–Starling (CS), modified Benedict–Webb–Rubin (MBWR), or extended corresponding states (ECS) formulation. The computer package used is designated as REFPROP<sup>8</sup> or Supertrapp.<sup>9</sup> The molecular weight is designated as MW in the table.

**Table 2. Thermal Conductivity Fit Coefficients and the Relative Standard Deviation of the Fit in Percent,  $\sigma$** 

chemical name	temp range/K	$a/(W \cdot m^{-1} \cdot K^{-1})$	$b/(W \cdot m^{-1} \cdot K^{-2})$	$\sigma$
trichlorofluoromethane	280–325	–7.31305E–03	5.13984E–05	1.54
octafluorocyclobutane	280–345	–1.05575E–02	7.54344E–05	0.84
1,1,1-trifluoro-2,2-dichloroethane	285–325	–1.36175E–02	8.01024E–05	1.96
1-fluoro-1,1-dichloroethane	300–335	–3.01317E–03	4.60624E–05	3.11
1,1,2-trifluoroethane	285–325	–1.37884E–02	9.17564E–05	1.78
1,1,1,2,3,3,3-heptafluoroethane	280–335	–1.20505E–02	8.53414E–05	1.15
1,1,1,2,3,3,3-hexafluoropropane	280–335	–4.71071E–03	6.38476E–05	1.55
1,1,1,3,3,3-hexafluoropropane	280–345	–1.34886E–02	8.74420E–05	0.97
1,1,2,2,3-pentafluoropropane	285–335	–1.54605E–02	9.62607E–05	2.19
2-(difluoromethoxy)-1,1,1-trifluoroethane	285–325	–4.39086E–03	5.95433E–05	1.95
2-(trifluoromethoxy)-1,1,1-trifluoroethane	285–325	–1.41303E–02	9.53310E–05	2.29
1,1,1-trifluoroacetone	280–345	–1.63865E–02	9.43158E–05	2.33
75% cyclopentane + 25% pentane	300–345	–5.21874E–02	1.99086E–04	1.82

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>4–6</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 by using wires of extremely small diameter [(5 to 7)  $\mu\text{m}$ ] and penetration of the thermal wave to the outer boundary can be eliminated by using 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.

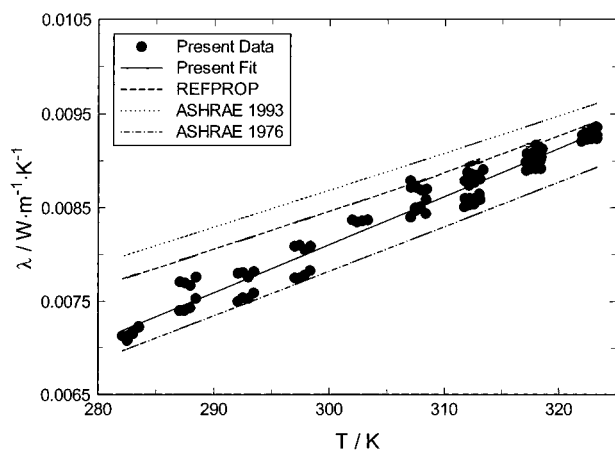
Given that the present transient hot-wire cell is less than optimum for such dilute gas measurements, it remains to apply the relatively large corrections carefully to obtain accurate thermal conductivity results. For our relatively large wires, it was found that only application of the full correction<sup>2</sup> for the finite wire dimensions was adequate. The outer boundary was not encountered during the 1 s duration of the experiment, since there is no curvature apparent at longer times for any of the gases measured. The largest gas thermal diffusivity for any of the reported measurements was  $6 \times 10^{-6} \text{ m}^2 \cdot \text{s}^{-1}$ . This conclusion is consistent with previous work on light gases such as argon and nitrogen at (1 to 2) bar,<sup>5,6</sup> where the outer boundary was encountered at times below 1 s when the thermal diffusivity was greater than  $9 \times 10^{-6} \text{ m}^2 \cdot \text{s}^{-1}$ .

## Results

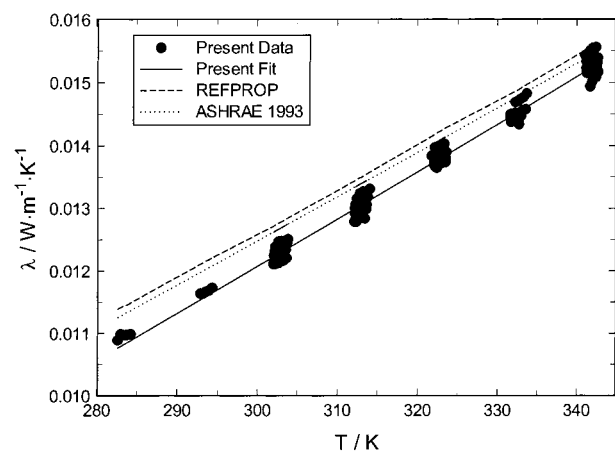
The results of measurements of the vapor phase of 13 gases are tabulated in the Supporting Information. The measurements are reported on the ITS-90 temperature

scale and are estimated to have an uncertainty of  $\pm 2\%$  at a 95% confidence level. A summary of these measurements is given in Tables 1 and 2. The uncertainty of the regression slope at 95% confidence, and hence the thermal conductivity according to eq 1, is given by the parameter STAT for each measurement reported in the Supporting Information and is typically less than 1%. Many of these measurements were made at temperatures below the normal boiling point of the compound. It should be emphasized that there are many difficulties associated with such measurements because of the small quantity of sample in the cell and of possible contamination with air impurities and adsorbed material on the surface of the cell and wires.<sup>7</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 is necessary during the data analysis to make the corrections to the measured temperature rise as described above. Models for most of the gases studied are available in the REFPROP<sup>8</sup> computer database. The exceptions to this were HFE263fb1, 1,1,1-trifluoroacetone, and the cyclopentane + pentane mixture, whose properties were calculated using a modified version of the SUPERTRAPP<sup>9</sup> computer database.

In Table 2 the thermal conductivity  $\lambda_0$  of each dilute gas is represented by a linear function of temperature  $T$ ,  $\lambda_0 = a + bT$ . The measurements used for the fits are near the saturation pressure at temperatures below the normal boiling point and at 1 bar for pressure above the boiling point. The standard deviation of the data relative to the fit is also shown in Table 2. This uncertainty generally increases for the materials with higher boiling temperatures.



**Figure 1.** Thermal conductivity of CFC-11 from 280 K to 325 K.

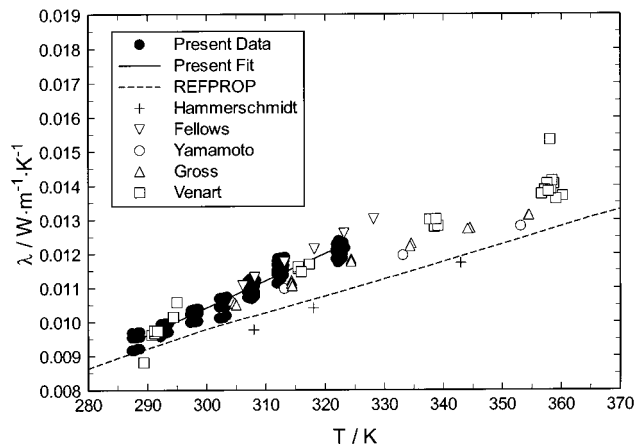


**Figure 2.** Thermal conductivity of RC318 from 280 K to 345 K.

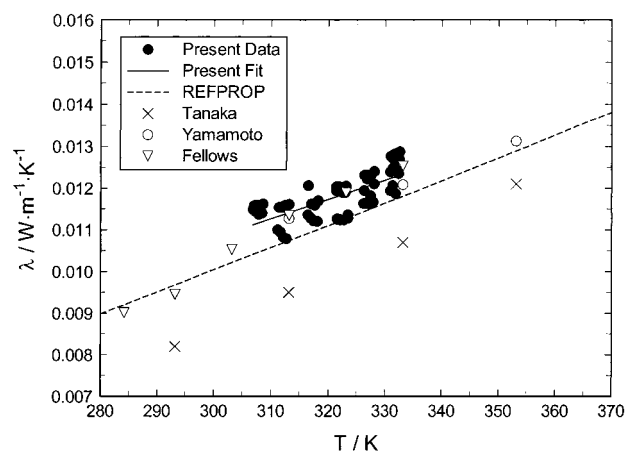
## Discussion

It is useful to compare the results reported here with results obtained by other independent experimental instruments and techniques to assess their accuracy. This is possible for four of the compounds measured here: CFC-11, RC318, HCFC-123, and HCFC-141b. Thermal conductivity has been widely studied for CFC-11, but there are discrepancies between data sets and recommendations<sup>10–12</sup> based on these data sets. The 1976 recommendations<sup>11</sup> of the American Association of Heating, Refrigerating, and Air-Conditioning Engineers (ASHRAE) are about 10% lower than the 1993 ASHRAE correlation.<sup>10</sup> The 1993 correlation<sup>10</sup> is based on the recommendations of Altunin et al.<sup>12</sup> The present data and correlation are shown to fall between these two correlations in Figure 1. The predictions of REFPROP<sup>8</sup> are in good agreement with the present data and also fall between the two ASHRAE recommendations. The ASHRAE correlation of Liley and Desai<sup>10</sup> is estimated to have an uncertainty of  $\pm 5$  to 6% for the thermal conductivity of CFC-11, and the present data agree within this uncertainty.

The ASHRAE recommendations of Liley and Desai<sup>10</sup> for the thermal conductivity of RC318 follow the standard reference data paper of Krauss and Stephan.<sup>13</sup> In this case, the agreement between the present data and the ASHRAE recommendations is much better, as shown in Figure 2. The predictions of REFPROP<sup>8</sup> are slightly higher than the ASHRAE correlation.<sup>10</sup> The ASHRAE reference correlation<sup>10</sup> for the thermal conductivity of RC318 is estimated to have an uncertainty of  $\pm 6\%$ , and the present data agree within this uncertainty. Thus, the present data agree for both



**Figure 3.** Thermal conductivity of HCFC-123 from 285 K to 360 K.

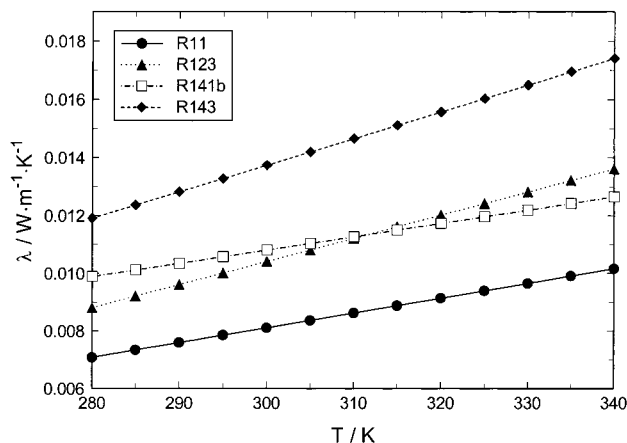


**Figure 4.** Thermal conductivity of HCFC-141b from 280 K to 360 K.

CFC-11 and RC318 appear to be reliable, on the basis of comparisons with existing recommendations.

The thermal conductivities of HCFC-123 and HCFC-141b have been recently measured by several authors using both transient and steady-state techniques. The thermal conductivity of HCFC-123 is shown in Figure 3 along with the data of five other researchers.<sup>14–18</sup> The thermal conductivity of HCFC-141b is shown in Figure 4 along with the data of three other researchers.<sup>15,17,19</sup> For both gases, REFPROP<sup>8</sup> predictions are lower than the measured transient results<sup>15–18</sup> while the measured steady-state results<sup>14,19</sup> are lower still. The transient data agree to within their mutual uncertainties for both HCFC-123 and HCFC-141b.

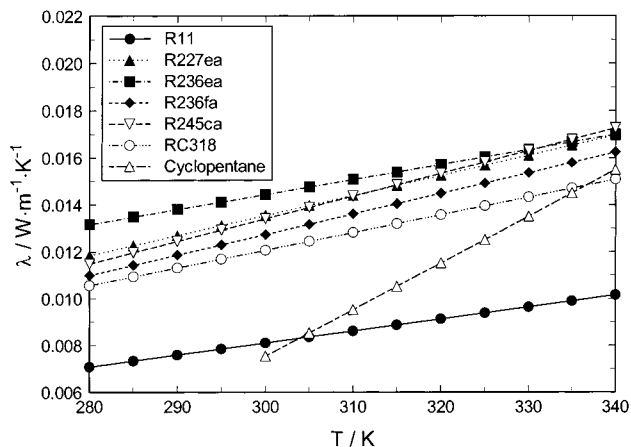
It is useful to examine the techniques used by each of the authors in an attempt to understand the discrepancies between the measurement techniques. In the case of HCFC-123, the results of Hammerschmidt<sup>14</sup> are the only steady-state data. For HCFC-141b, the results of Tanaka et al.<sup>19</sup> are the only steady-state data. Hammerschmidt<sup>14</sup> used a parallel-plate instrument, while Tanaka et al.<sup>19</sup> used a concentric-cylinder instrument. Both steady-state instruments are relative devices and were calibrated with noble gases. Both steady-state instruments have an estimated uncertainty of  $\pm 2\%$ , yet both instruments are systematically lower than the transient measurements by (8 to 10%). This systematic difference is quite significant and larger than the combined uncertainty of the transient and steady-state data sets for both gases.



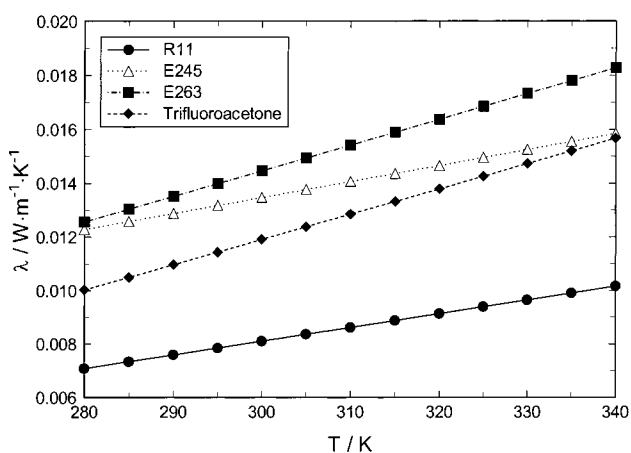
**Figure 5.** Thermal conductivity of the ethane-derived halocarbons relative to that of CFC-11.

All of the transient instruments were absolute devices and required no calibration. As discussed above, the transient hot-wire technique has two corrections which can get quite large for dilute-gas measurements. If all the instruments had similar geometry and there were an error in these corrections, this could explain some of the discrepancy. The steady-state instruments on the other hand do not require such large corrections but are susceptible to parasitic heat losses that become more significant for gases with very low thermal conductivity, such as these refrigerant gases. The diameter of the transient hot wires varied significantly between the various researchers. Yamamoto et al.<sup>17</sup> used the smallest wire (5  $\mu\text{m}$  in diameter). Venart,<sup>18</sup> Fellows et al.,<sup>15</sup> and the present work used 12.5  $\mu\text{m}$  diameter wires. Gross et al.<sup>16</sup> used the largest wire, with a 17  $\mu\text{m}$  diameter. Thus, the wire diameter varied by a factor of 3 between these four researchers and the finite wire radius correction varied by a factor of 9 ( $3^2$ ). Yet, all of the transient researchers report the same result to within their combined uncertainty. Several of the papers are not clear on the outer boundary dimensions,<sup>15,17,18</sup> but Gross et al.<sup>16</sup> had an outer boundary of 27 mm relative to the outer boundary in the present measurements of 9 mm. This is a variation of a factor of 3, so errors due to the outer boundary should be apparent and much more pronounced for the present measurements. The transient measurements compared in Figures 3 and 4 have significant differences in cell geometry while the measurements remain consistent with each other. It is concluded that the present transient hot-wire measurements of refrigerant thermal conductivity are reliable on the basis of agreement of each experiment with the instrument theory of eq 1 (tabulated as STAT in the Supporting Information) and by comparison with other independent measurements using apparatus with different techniques and cell geometries.

The correlations for the dilute-gas thermal conductivities of each of the 13 gases can be used to compare the effectiveness of each gas relative to the CFC-11, which must ultimately be replaced both as a refrigerant and a foam-blowing agent. These correlated lines are shown in Figures 5–7. Figure 5 shows results for the ethane derivatives HCFC-123, HCFC-141b, and HFC-143 relative to CFC-11. Both HCFC-123 and HCFC-141b have lower thermal conductivities than that of HFC-143 and also have higher boiling points that more closely match that of CFC-11. Figure 6 shows results for the fluorinated propane derivatives and also includes results for a cyclopentane + pentane mixture, which has been suggested as a likely replacement for foam-blowing applications in Europe. The



**Figure 6.** Thermal conductivity of the fluorinated propane derivatives, fluorinated cyclic alkanes, and 75% cyclopentane + 25% pentane compared with that of CFC-11.



**Figure 7.** Thermal conductivity of the oxygenated series relative to that of CFC-11.

fluorinated propanes HFC-227ea, HFC-236ea, HFC-236fa, and HFC-245ca have thermal conductivities which are nearly equal. The cyclic materials RC318 and the cyclopentane + pentane mixture have thermal conductivities that are slightly lower than those of the linear propane derivatives. Finally, Figure 7 shows results for the oxygenated gases HFE245fa1, HFE263fb1, and 1,1,1-trifluoroacetone. The thermal conductivities of these gases are nearly equal to those of the propane type gases, with that of 1,1,1-trifluoroacetone being slightly lower than those for the ethers. It is interesting to note that the data for 1,1,1-trifluoroacetone are about 10% higher than the recommendations of Vargaftik et al.<sup>20</sup> for acetone vapor.

## Conclusions

Correlations are presented for the dilute-gas thermal conductivities of 13 gases that have potential for use in expanded cellular foams. These thermal conductivity data are estimated to have an uncertainty of  $\pm 2\%$ . Reasonable agreement is found between the data available in the literature for CFC-11, RC318, HCFC-123, and HCFC-141b. However, it is alarming that steady-state measurements from two independent laboratories appear to be systematically lower than transient measurements for both HCFC-123 and HCFC-141b. The cyclopentane + pentane mixture, HCFC-123, and HCFC-141b have the lowest thermal conductivities of the alternative gases studied near 300 K. All of the gases studied have dramatically lower thermal conductivities than those (typically  $27 \text{ mW}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$  at 300

K) for the air that they typically displace in a cellular-foam insulation and so remain potentially attractive for foam-blowing applications. None of the gases studied has a thermal conductivity that is as low as that of the chlorofluorocarbon, CFC-11, which must be withdrawn from use due to its high ozone-depletion effect.

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### Supporting Information Available:

Tabulated experimental data, complete with the observed relative uncertainty of each measurement based on the complete theory of the transient hot-wire instrument (available in both pdf and ACSII formats). This material is available free of charge via the Internet at <http://pubs.acs.org>.

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