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# THERMAL CONDUCTIVITY OF A POLYURETHANE FOAM FROM 95 K to 340 K

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U.S. Department of Commerce  
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U.S. DEPARTMENT OF COMMERCE, Malcolm Baldrige, Secretary

NATIONAL BUREAU OF STANDARDS, Ernest Ambler, Director



Thermal Conductivity of a Polyurethane Foam  
From 95 K to 340 K\*

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Values of thermal conductivity for a  $32 \text{ kg/m}^3$ ,  $\text{CCl}_3\text{F}$  blown polyurethane foam are presented in the temperature range from 95 to 340 K. These data were obtained using a guarded-hot-plate apparatus (ASTM C 177). The apparent conductivity is discussed in terms of contributions from radiation and solid and gas conduction. Heat transfer via gas convection is not significant when the cell diameters are less than approximately 3 mm; cell dimensions of the thermal conductivity specimen are on the order of 0.5 mm.

Key words: convection; foam; gas conduction; guarded-hot-plate; insulation; low temperature; radiation; solid conduction; thermal conductivity.

## 1. INTRODUCTION

Safe and economical storage, transportation, and use of cryogenic fluids depend to a large extent on materials used in construction of associated equipment. Thermal insulation and insulation systems are critical parts of essentially all equipment which must be used at low temperatures. Generally, the particular application dictates the generic type of thermal insulation or, at least, reduces the number of candidates. Some of the widely used types of insulation and concomitant thermal conductivity ranges are shown in figure 1. As seen in this figure, the evacuated insulations are the most thermally efficient. Most large-scale needs, however, preclude vacuum insulation on the grounds of cost and durability. The three classes of unevacuated insulation are reasonably competitive in thermal effectiveness at low temperatures; all are used, for instance, in large liquefied natural gas (LNG) and liquefied petroleum gas (LPG) storage tanks and tanker ships. The high strength-to-weight ratio of expanded plastics (foams),  $\sim 8 \text{ kPa/kg/m}^3$  compared to  $\sim 50$  for aluminum alloys and  $\sim 25$  for stainless steels, allows these insulations to be self supporting and to lend structural support to the system. Foam insulations are found in most recent LNG/LPG-oriented construction.

The particular material to be discussed below is a closed cell, rigid polyurethane (PU) foam of nominal density  $32 \text{ kg/m}^3$  ( $2 \text{ lb/ft}^3$ ). A foam is a composite consisting of a skeleton of solid materials and voids filled with various gases. The term "closed cell" is used to describe materials in which the voids form discrete chambers. A foam may be considered rigid when it is used below the glass transition temperature,  $T_g$ . This transition is of the second order and is characterized by a change in the specific heat of the resin. The transition occurs between 400 and 485 K for most PU resins. Polyurethane is a thermosetting material as opposed to a thermoplastic material (polystyrene

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is a thermoplastic material, and thus softens when heated). Thermosetting materials do not soften significantly at elevated temperatures, but rather decompose directly.

A property of primary importance in thermal insulations is, of course, thermal conductivity (as applied to cellular composites, the term apparent thermal conductivity is more correct since radiative and convective components are included along with the true conductive modes of heat transfer). Heat is transferred through foams via four mechanisms: (1) gas conduction,  $Q_g$ , (2) solid conduction,  $Q_s$ , (3) radiation,  $Q_r$ , and (4) convection,  $Q_c$ . The total heat flow,  $Q_T$ , via the four parallel modes is normally assumed to be additive

$$Q_T = Q_g + Q_s + Q_r + Q_c. \quad (1)$$

The dominant mode of heat transfer in an insulating foam is gas conduction until the temperature becomes low enough to liquefy the cellular gas. Solid conduction through the resin structure is small but not negligible (the solid conduction is a function of foam density). Radiation is similarly a small, but not negligible, component. Convective heat transfer is negligible for small-cell foams (cell diameter 3 to 4 mm) (Skochdopole, 1961); however, this component becomes critically important for cracked foams and slab installations where vertical cracks allow natural convection.

The effect of aging and gas permeability on the thermal conductivity must be considered when foams are used in facilities with long lifetimes. As mentioned above, the total heat transfer through a foam is very dependent upon the cellular gas content. The gas content is, in turn, dependent on age, permeability, and how the foam is shielded from the atmosphere. The rates of diffusion of gases into and out of PU foams are indicated in table 1. As seen in this table, PU would tend to lose a heavy fluorocarbon gas, such as  $CCl_3F$ , very slowly but would absorb air approximately 15 times as rapidly. If unprotected, air will diffuse into PU foam until the partial pressure of air is equal inside and out; this, combined with the slower loss of heavier fluorocarbon fill gases will result in a net positive pressure. Eventually the equilibrium gas content will be air at ambient pressure.

Table 1. Permeability of gases through PU films (Scott and Roff, 1971)

Gas	Permeability, $10^{-15} \frac{\text{cm}^3 \text{ gas} \cdot \text{mm}}{\text{s} \cdot \text{cm}^2 \cdot \text{Pa}}$	
	Polyester	Polyether
$CCl_3F$	2.2	
$N_2$	20.3	36.8
$O_2$	80.3	112 to 360
$CO_2$	30.0	1050 to 3000
Air	32.3	

## 2. APPARATUS

The apparatus used to make the thermal conductivity ( $k$ ) measurements is shown schematically in figures 2 and 3. The path of the cooling fluid is shown in figure 2 and the details of the specimen configuration are shown in figure 3. This type of system is commonly known as a guarded-hot-plate and is described in the American Society for Testing and Materials Standard C 177 (ASTM, 1978). This method allows an absolute determination of  $k$  and is considered to be the most accurate method available for insulations. The particular apparatus used to make the  $k$  measurements reported here is described in detail by Smith, Hust, and Van Poolen (1981); a very brief description is included below.

The basic operation of this type of apparatus involves supplying a measured amount of power,  $Q$ , from the main heater plate to the two specimens and measuring the resulting steady-state temperature difference. The thermal conductivity is then given by

$$k = \frac{\Delta X}{2A} \cdot \frac{Q}{\Delta T} \quad (2)$$

where  $\Delta X$  is the sample thickness,  $A$  the area of the metered section of the main heater plate, and  $\Delta T$  is the steady-state mean temperature difference across the specimens. The absolute temperatures and temperature differences are measured with type K thermocouples.

The accuracy of the  $k$  results is dependent on the accuracy of measurement of the parameters shown in eq (2) and on establishment of unidirectional (vertical in this case) heat flow in the metered area of the specimens. The inner-guard heater plate is controlled at the temperature of the main heater in order to minimize radial heat flow in the metered area. Somers and Cyphers (1951) and ASTM C 177 indicate that when the diameter-to-thickness ratio of a specimen is greater than 4, the errors in  $k$  due to edge losses should be less than 1%. The metered diameter of the foam specimens was 10.2 cm and the thickness was 2.54 cm.

The most critical and difficult parameters to determine are the specimen temperature differences ( $\Delta T$ );  $\Delta T$  averaged 10 K for the PU foam specimens. The correctness of  $\Delta T$  measurements depends on plate-to-specimen thermal contact, on unidirectional heat flow, and on thermocouple calibration and referencing sources of error. The diverse materials which can be tested in this type of system cause a wide range of plate-to-specimen contact situations. Tye and Spinney (1976) found that embedding the thermocouple wires in the specimen, as opposed to the measuring plates, resulted in higher calculated conductivities. This effect is due to better specimen-to-thermocouple contact which results in smaller measured  $\Delta T$  and larger  $k$  (eq (2)). They also found that the effect of thermocouple placement was dependent on the conductivity of the specimens. As the conductivity of the specimen increases, the relative effect of an air gap in the specimen-to-plate interface becomes larger.

Thermocouple placement used in obtaining the data reported below consisted of cementing the wires into machined grooves in the measuring plates (plates adjacent to the specimens shown in figure 3) so that the top of the wires were flush with the plate surfaces. The plate emissivity was 0.82.

The accuracy of measurements made with this system on fiberglass and fiberboard reference materials is discussed by Smith, Hust, and Van Poolen (1981). In a qualitative sense the plate-to-specimen contact should be similar for low density foams and the fibrous materials tested by Smith. Based on the percentage errors given by Smith for each parameter in eq (2) and the magnitude of these parameters for the PU foam experiments, uncertainties in the PU foam data are estimated to be 0.2 mW/m·K random and 0.4 mW/m·K systematic near room temperature; near 80 K the random and systematic components are 0.1 mW/m·K.

### 3. MATERIAL

The material tested in this study is a nominal 32 kg/m<sup>3</sup> (2 lb/ft<sup>3</sup>) polyurethane foam (PU) designated as GM30. Our supply of this material was obtained from the NBS Office of Standard Reference Materials (OSRM), Washington, DC. The OSRM distributed this and other expanded plastics for the Products Research Committee (1980). These materials were commercially produced and designated by OSRM as General Materials. Our bulk supply of GM30 was in the form of a 0.1 x 1.2 x 1.8 m slab. The thermal conductivity tests were conducted on specimens taken from the circular area indicated in figure 4. The chemical formula for the PU resin in GM30 is C<sub>10.47</sub>H<sub>11.99</sub>O<sub>2.38</sub>N, the molecular weight is 189.71, and the composition by weight is (%) C, 66.2; H, 6.3; O, 20.2; and N, 7.4 (Products Research Committee, 1980). The orientation of the elongated cell axis for the material used in the physical properties tests was determined optically from statistical evaluation of microphotographs. The ratios of cell height to cell width for the principal orthogonal planes of the test specimens were: x/y = 1.33 ± 0.24, z/x = 1.02 ± 0.18, z/y = 1.40 ± 0.26. The ± values represent estimates of one standard deviation. The cell orientation relative to the orthogonal axes of the bulk slab are shown in figure 5. The average intercepted cell cross sectional areas in the principal orthogonal planes were found using the method described by Schael (1967) and are shown in table 2.

Table 2. Average intercepted cell area for two specimens of PU, 32 kg/m<sup>3</sup> foam

Specimen	XY plane	XZ plane	YZ plane
GM30-2	1.27x10 <sup>-3</sup> cm <sup>2</sup> /cell	1.21x10 <sup>-3</sup> cm <sup>2</sup> /cell	1.25x10 <sup>-3</sup> cm <sup>2</sup> /cell
GM30-3	1.02x10 <sup>-3</sup>	1.32x10 <sup>-3</sup>	1.11x10 <sup>-3</sup>

The cellular surface to volume ratios for the specimens listed in table 2 are 67.7 and 69.6 cm<sup>-1</sup> for specimens GM30-2 and GM30-3 respectively.

The composition of the gas in the foam cells, table 3, was determined by mass spectrometry at the beginning of the thermal conductivity tests.



Table 3. Gas content of GM30, PU foam as determined using mass spectrometry

Gas	Mole %
CCl <sub>3</sub> F	71.2
N <sub>2</sub>	6.1
O <sub>2</sub>	21.8
A	0.5
CO <sub>2</sub>	0.3
CO	0.1

#### 4. EXPERIMENTAL RESULTS

Thermal conductivity as a function of temperature, for GM30 PU foam, is shown in table 4 and figure 6. These data were determined over a period of 18 days in the guarded-hot-plate system described earlier. The numbers given in table 4 represent averages of 3 or more determinations of  $k(T)$ ; the determinations were separated by a minimum of 15 minutes. Each determination is plotted in figure 6, but the resolution is such that, in every case, the set appears as a single point.

Table 4. Average thermal conductivity as a function of average temperature for a 32 kg/m<sup>3</sup> PU foam

$\bar{T}$ (K)	k (mW/m·K)	$\bar{T}$ (K)	k (mW/m·K)
95.10	11.4	256.76	22.6
106.89	13.0	258.17	22.8
120.39	14.6	264.67	21.6
132.04	15.8	270.65	20.4
153.89	18.2	274.67	20.1
169.54	19.9	275.74	19.9
175.29	20.7	280.78	20.0
185.78	21.7	284.90	20.5
202.20	23.1	290.79	21.2
214.63	23.8	297.00	21.6
216.82	23.8	297.95	21.8
220.54	23.9	301.97	22.2
239.67	23.8	318.03	24.5
239.91	23.8	332.38	26.2
248.97	23.6	332.94	26.0
		340.22	27.8

The sequence of  $k$  determinations was not in order of increasing or decreasing temperatures, e.g., one determination might be made at an average specimen temperature ( $\bar{T}$ ) of 120 K while the next determination might be made at  $\bar{T} = 280$  K.

The specimen environment throughout the 18 day testing period was dry nitrogen gas. The gas surrounding the specimen was maintained at a slight positive pressure of 260 to 660 Pa (2 to 5 mm Hg) relative to atmospheric pressure.

## 5. DISCUSSION

As mentioned earlier, heat is conducted through a foamed material via solid and gaseous conduction, by radiation, and by convection. The cell size of the PU foam tested here is too small to allow natural convection to contribute to the total heat flow (Skochdopole, 1961).

The published thermal conductivity data for nominal 32 kg/m<sup>3</sup> PU foams at low temperatures are shown in figure 7. A strict quantitative analysis of the contribution of each of the three modes of heat transfer is not possible for the literature data because insufficient information is available. It is possible, however, to establish in general terms that gas conduction is the principal mode and that the NBS data reflect the expected conductivity based on the known gas content.

Radiant heat flow through foam insulators is due to transmission and absorption and reradiation. Resins used for making foams are partially transparent in the 2 to 30 μm range of wavelengths (Skochdopole, 1961). Sections associated with thermal insulations are normally optically thick (thickness  $\gg$  photon mean free path), however, so that direct transmission will be small. The effective  $Q_r$  is expected to be proportional to  $T^3$  and for nominal 32 kg/m<sup>3</sup> PU foams at ambient temperature is equivalent to  $k_r \approx 4 \text{ mW}/(\text{m}\cdot\text{K})$ .

Solid conduction takes place through the cell walls and membranes of the foam. Since PU resins are made up of disordered (noncrystalline) materials, there is little lattice conduction at low temperatures and the conductivity is nearly a linearly decreasing function of temperature. The heat conducted by the solid phase can be estimated by

$$k_s = V_s k_{ss} \quad (3)$$

where  $V_s$  is the relative volume of resin in the foam and  $k_{ss}$  is the thermal conductivity of the non-expanded polymer. Using this and  $k$  (ambient temp.) = 133 mW/(m·K) (Nadeau, Waszeciak, and Sayigh, 1967),  $k_s = 4 \text{ mW}/(\text{m}\cdot\text{K})$ , since the resin only occupies approximately 3% of the volume of a 32 kg/m<sup>3</sup> (2 lb/ft<sup>3</sup>) foam. This contribution must be further reduced by a steric factor (Hammond, 1968), i.e., the portion of the solid that is perpendicular to the direction of heat flow cannot contribute directly to the conduction. This factor depends on the orientation of the cells relative to the direction of heat flow and is on the order of 0.66 to 1.

The remaining mode for heat transfer is conduction in the gas occupying the foam cells. As will be shown below, this is the largest component. Figure 8 indicates the conductivity of the gases typically found in an insulating foam; a combination of CCl<sub>3</sub>F, O<sub>2</sub>, and N<sub>2</sub> was present in the foam tested here (table 3). When a mixture of gases is present,  $k_g$  can be estimated by using

$$k_g = k_{\text{mixture}} = \sum_{i=1}^n X_i k_i \quad (4)$$

where thermal conductivities of the individual species are given by  $k_i$ ,  $X_i$  represents the corresponding mole fraction, and  $n$  is the number of species present. A computation of  $k_g$  at ambient temperature using the gas composition given in table 3, yields  $k_g = 12.9 \text{ mW}/(\text{m}\cdot\text{K})$ .

It is not rigorously correct to add the conductivities due to the three modes of heat transport (the heat transfer modes are not independent of one another). Results obtained using this approach have, however, been found to approximate  $k$  of rigid polyurethane foams (e.g., Ball, Hurd, and Walker, 1970, and Levy, 1966).

$$k_T = k_g + k_r + k_s \quad (5)$$

If one uses eq (5) and the values of  $k_g$ ,  $k_s$ , and  $k_r$  obtained above,  $k_T = 20.9 \text{ mW}/(\text{m}\cdot\text{K})$  at 290 K. The experimental value from figure 6 is  $21.1 \text{ mW}/(\text{m}\cdot\text{K})$ . This close agreement tends to verify the composition of gases in the NBS specimen as determined via mass spectrometer.

As temperature decreases, the contribution of radiation decreases as  $T^3$  and the contribution of solid conduction decreases as  $T$ . The contribution due to gas conduction, however, is a complex function of temperature and composition. Figure 9 illustrates the conductivity of three gas mixtures as a function of temperature. The shape of the 33, 40, and 47 mole percent  $\text{CCl}_3\text{F}$  data are similar to that seen in figure 7 for the total, apparent conductivity of nominal  $32 \text{ kg}/\text{m}^3$  PU foams. The variability in the PU data of different experimenters, shown in figure 7, may be attributed to different gas content at the time of measurement (smaller differences no doubt also occur due to experimental errors). These differences cannot be quantitatively analyzed since specific gas contents are not known for specimens used by other experimenters.

The heat transfer observed in a practical application of PU foam insulation is approximated by the summation of the integrals of the modes of conduction. The integral approach is necessary since  $k_i$  is a non-linear function of temperature and is dependent on the temperature distribution through the thickness of the foam. As mentioned above,  $Q_r$  and  $Q_s$  are small and can be approximated by simple functions of  $T$ . To the first order, the heat flow due to these modes is  $Ak_s\Delta T$  and  $k_r\Delta T$ , where  $A$  is a constant containing the steric factor and the volume percent resin. These relationships do not account for the fact that solid conduction will be increased slightly when the cell gas condenses or that the gas does absorb radiant energy and this absorption changes with temperature. The gas conduction component, on the other hand, must be divided into temperature zones due to the phase changes involved. While both  $\text{CCl}_3\text{F}$  (or other fill gas) remains in the vapor phase a mixture of air and fill gas must be considered so that  $k_g = \int_{T_1}^{T_{HI}} k_{\text{mixture}} dT$  where  $T_1$  represents the temperature where the fill gas has essentially all condensed leaving only air in the cells. This temperature is seen to be at approximately 220 K in figure 9. The variation in experimental data shown in figure 7 is also seen to decrease at about this temperature. This would be expected since the conductivity below this temperature is primarily due to air. At temperatures below  $T_1$ , but higher than the air condensing

temperature ( $T_2$ ),  $k_g = \int_{T_2}^{T_1} k_{air} dT$ . When  $T < T_2$ , the foam is in a cryopumped state so that  $k_T = k_S + k_r$  and, since  $k_r$  is small,  $k_T \sim k_S$ .

This temperature-zone behavior of the gas conduction can be qualitatively illustrated by considering an aged  $CCl_3F$  blown PU foam with one face at 19 K (-425°F) and the other at 300 K (80°F). Beginning at the 300 K face, gas conduction will be due to the mixture of air and  $CCl_3F$ . The conductivities of several possible mixtures are shown in figure 10. If the mixture is assumed to be 50 mole percent air and 50 mole percent  $CCl_3F$ , the conductivity will decrease along this constant composition line until the temperature is reached where  $CCl_3F$  begins to condense. The actual temperature at which this occurs depends on the partial pressure of the  $CCl_3F$  in the cells. This corresponds to the 278 K inflection in the thermal conductivity of the NBS PU specimen (figs. 6 and 7) and the  $k_g$  inflection shown between 275 and 295 K in figure 9. As the temperature is lowered, the mole fraction of  $CCl_3F$  gas will decrease until essentially only gaseous air is present. This decreasing  $CCl_3F$  gas content causes the observed conductivity to have a negative slope in the range  $230 \leq T \leq 280$  K. The second regime of gas conduction is encountered as the distance to the cold wall is decreased. The gas conduction here is entirely due to air and decreases smoothly until the components of air begin to condense. The third regime begins at this temperature. The cellular-gas pressure continues to decrease as the cold wall is approached and air condenses. The gas conductivity decreases, as would be expected from the conductivity of air shown in figure 8. At ~50 K, the pressure in the cells will be approximately 133 Pa (1 mm Hg) and the mean free path of the gas molecules will approximate the cell diameter. Under these conditions,  $k_g$  will be a function of pressure. Pressure within the cells and the concomitant gas conduction continue to decrease as the cold wall is approached. Solid conduction and radiation remain as the principal modes of heat transfer in the very low (cryopumped) temperature section of the foam.

## 6. CONCLUSIONS

The effective or apparent thermal conductivity of a rigid, fluorocarbon blown foam is strongly dependent upon the composition of gas in the foam cells. For temperatures encountered in LNG/LPG installations a complex, double inflected conductivity exists. The observed insulation quality, or conductivity, will reflect the changing gas characteristics as the temperature changes in the body of the foam. It appears that the effective conductivity can be modeled well enough to allow users and producers of PU foam to base designs on density and measured gas content.

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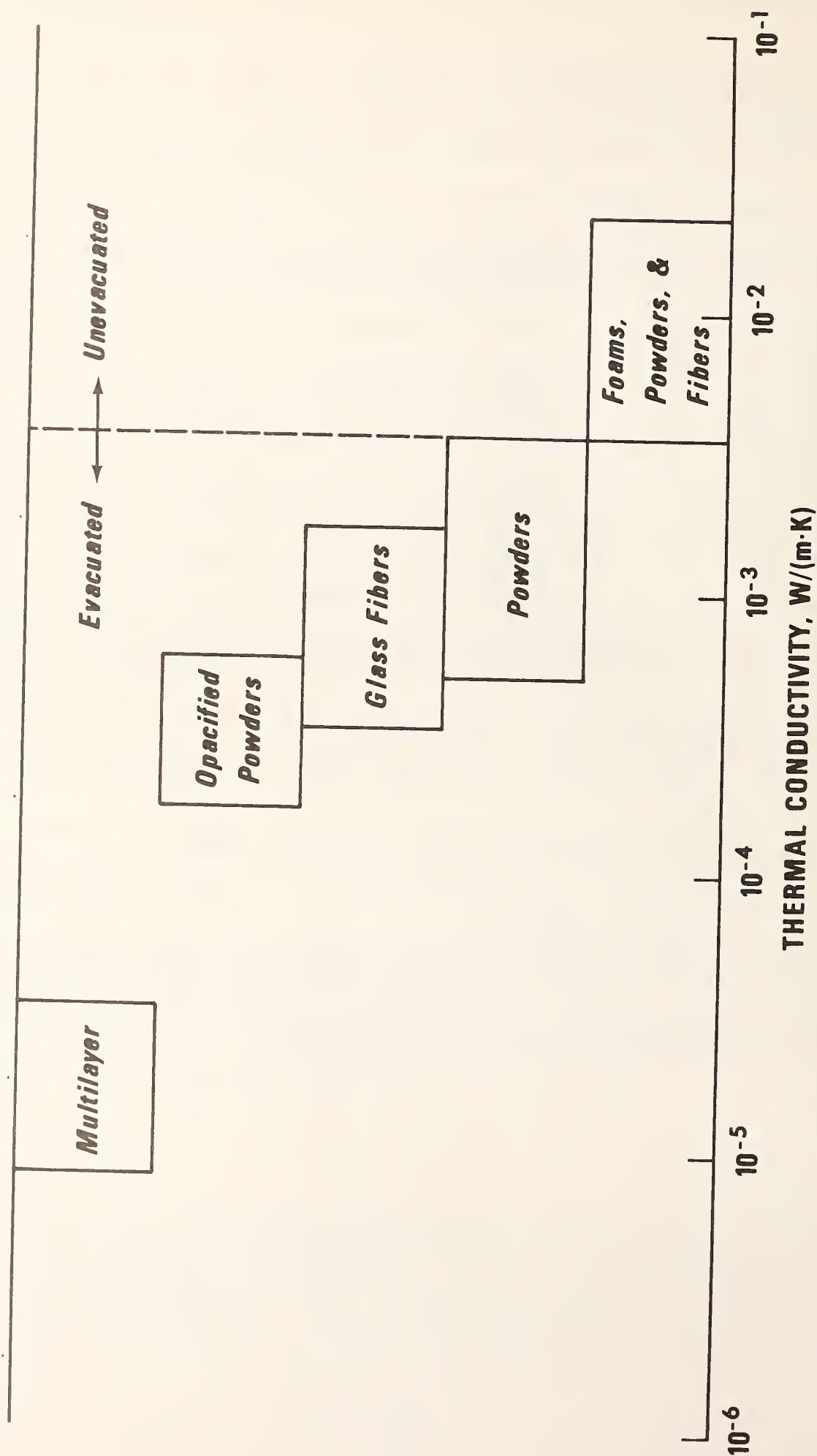


Figure 1. Thermal conductivity of generic thermal insulations (Glaser, Black, Linstrom, Ruccia, and Wechsler, 1967) in the temperature range  $77 \lesssim T \lesssim 238$  K ( $-320 \lesssim T \lesssim -30^\circ\text{F}$ ) and densities in the range  $30 \lesssim \rho \lesssim 80$  kg/m<sup>3</sup> ( $2 \lesssim \rho \lesssim 5$  lb/ft<sup>3</sup>).

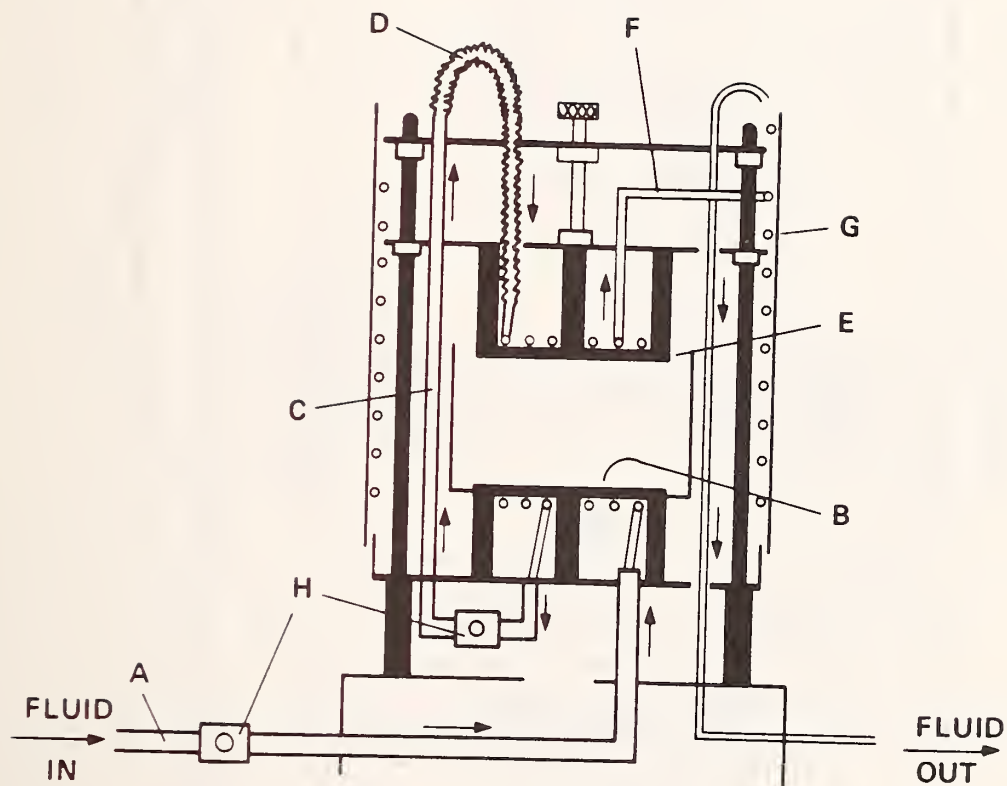


Figure 2. Schematic of the guarded-hot-plate thermal conductivity apparatus. A, vacuum insulated transfer tube; B, bottom cold plate; C, vacuum insulated transfer tube; D, flex tubing; E, top cold plate; F, transfer tube; G, shroud cooling tubes; H, throttle valve.

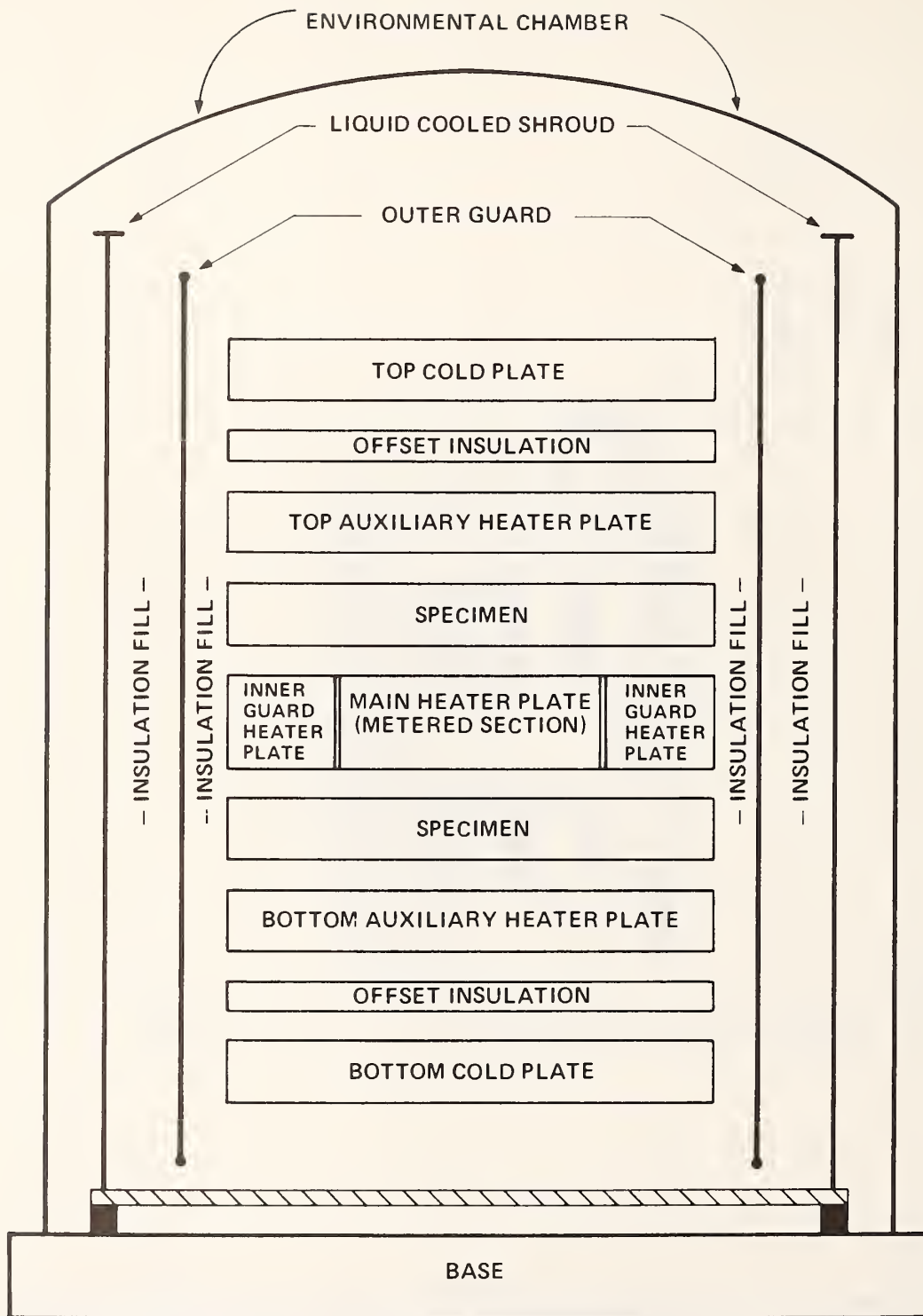


Figure 3. Schematic of guarded-hot-plate specimen configuration and environmental chamber.



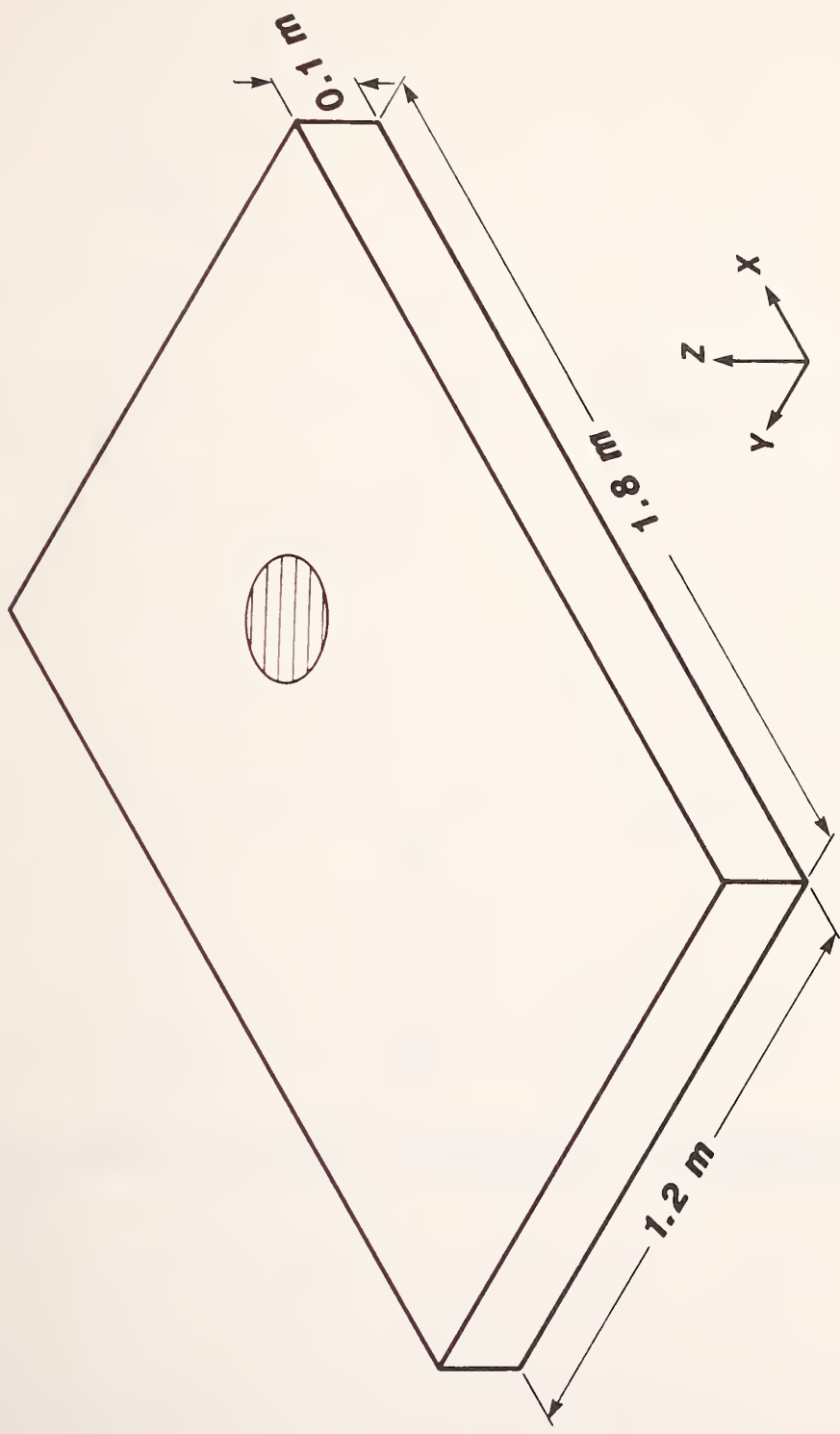


Figure 4. Bulk supply of  $32 \text{ kg/m}^3$ , PU foam designated GM30. The thermal conductivity specimens were taken from the circular-shaded area.

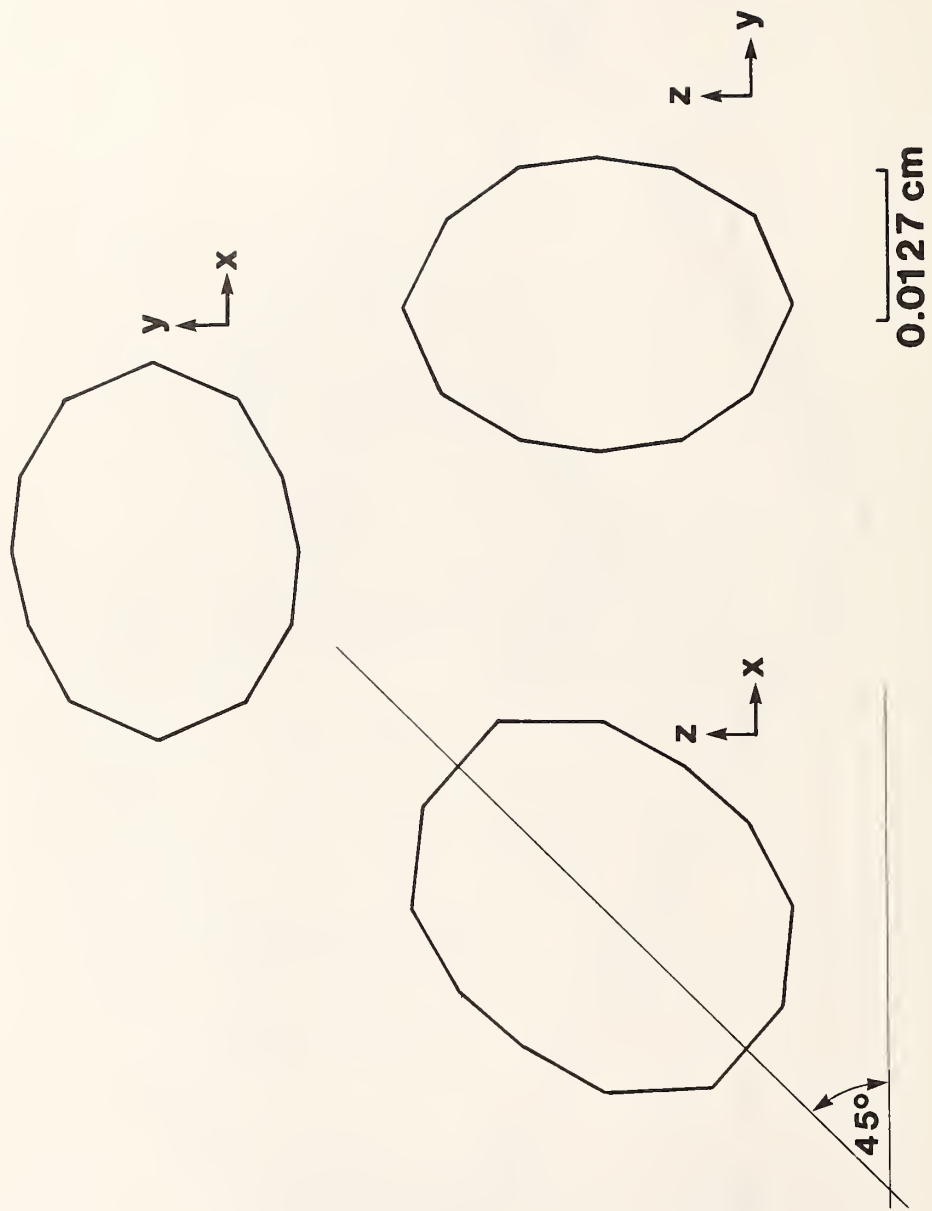


Figure 5. Cell shape and orientation relative to the principal-orthogonal axes of the bulk supply of GM30. The connected short straight lines do not represent actual cell boundaries but are rather artifacts of the method.

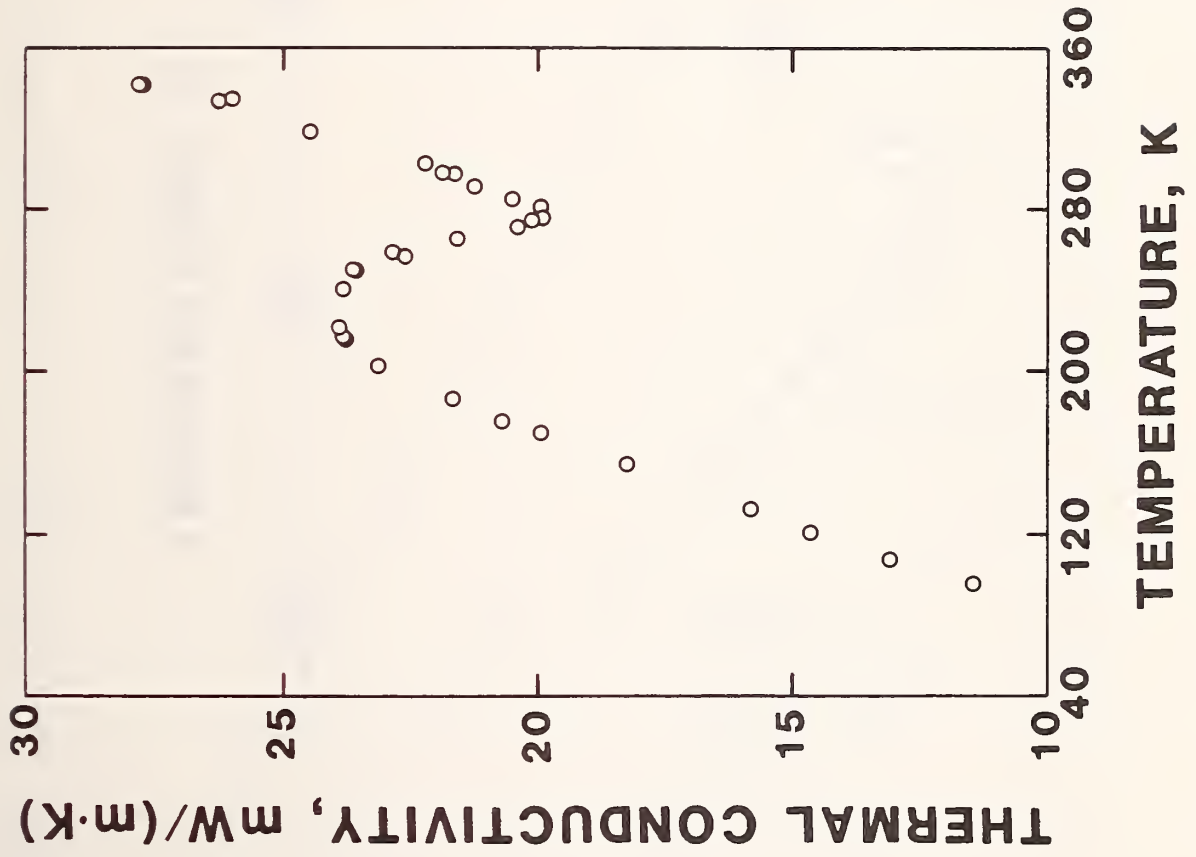


Figure 6. Thermal conductivity,  $mW/(m \cdot K)$ , versus temperature,  $K$ , for the 32  $kg/m^3$ , PU foam studied at NBS. The cellular gas content at the time of measurement was approximately 71 mole %  $CCl_3F$ , 6 mole %  $N_2$ , and 22 mole %  $O_2$ .

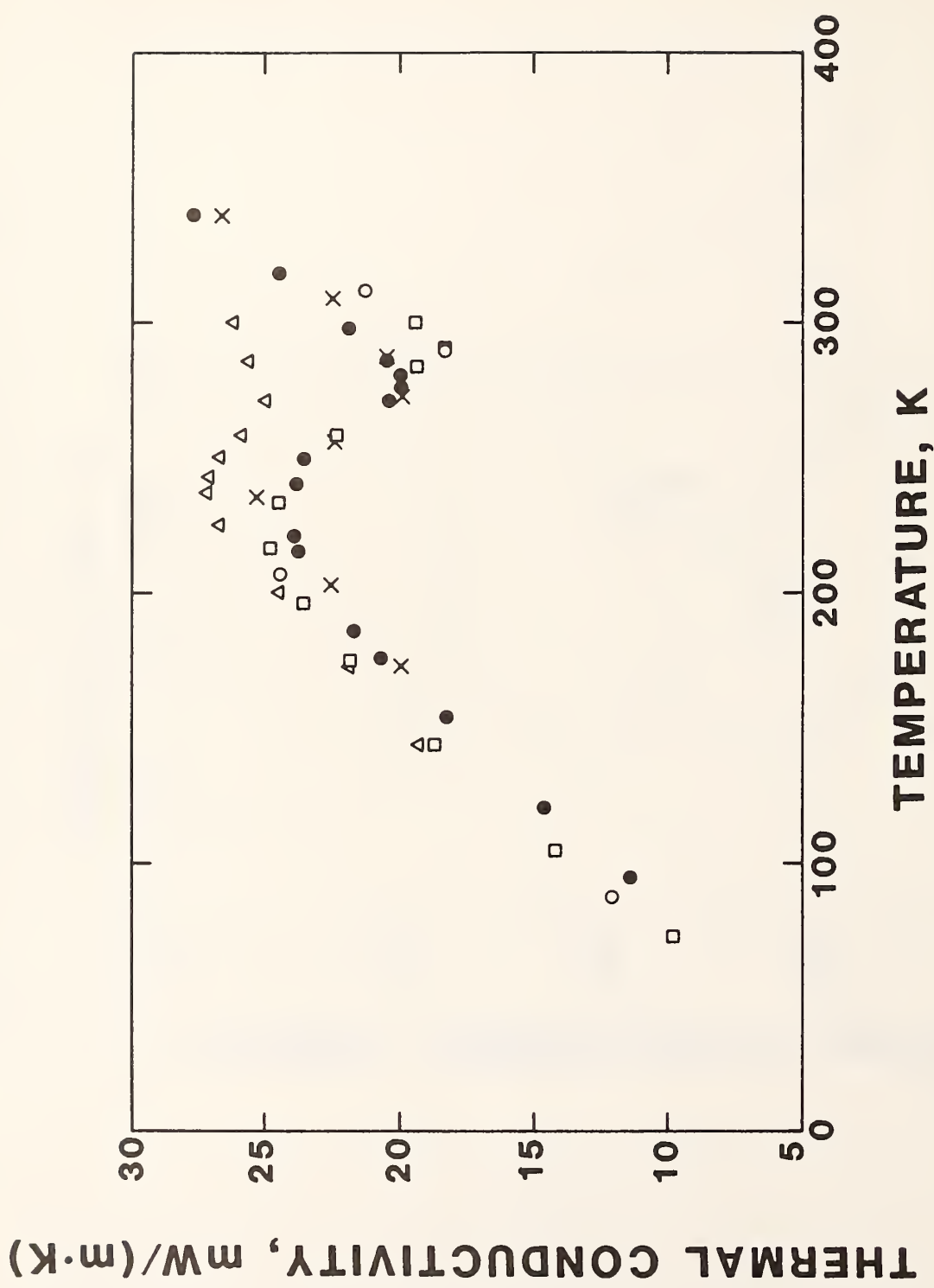


Figure 7. Thermal conductivity,  $mW/(m \cdot K)$ , versus temperature,  $K$ , for five nominal  $32 \text{ kg}/m^3$ , PU foams:  $\circ$ , Stephan Chemical Co. (1978);  $\times$ , Tye (1968);  $\Delta$ , Tye (1971);  $\square$ , Schroeder (1973);  $\bullet$ , NBS (this report).

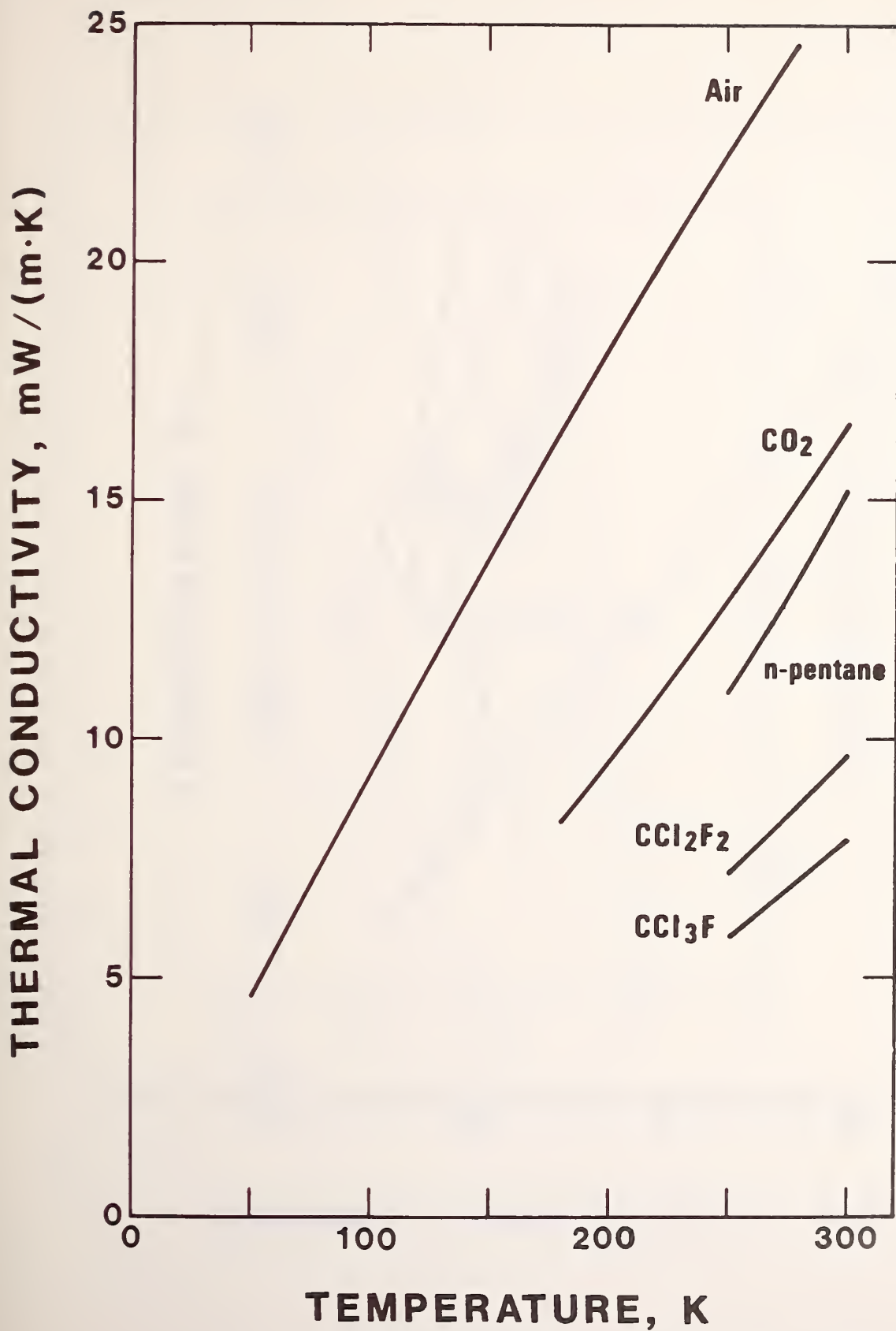


Figure 8. Thermal conductivity, mW/(m·K), as a function of temperature, K, for commonly used fill gases (Touloukian, Liley, and Saxena, 1970).

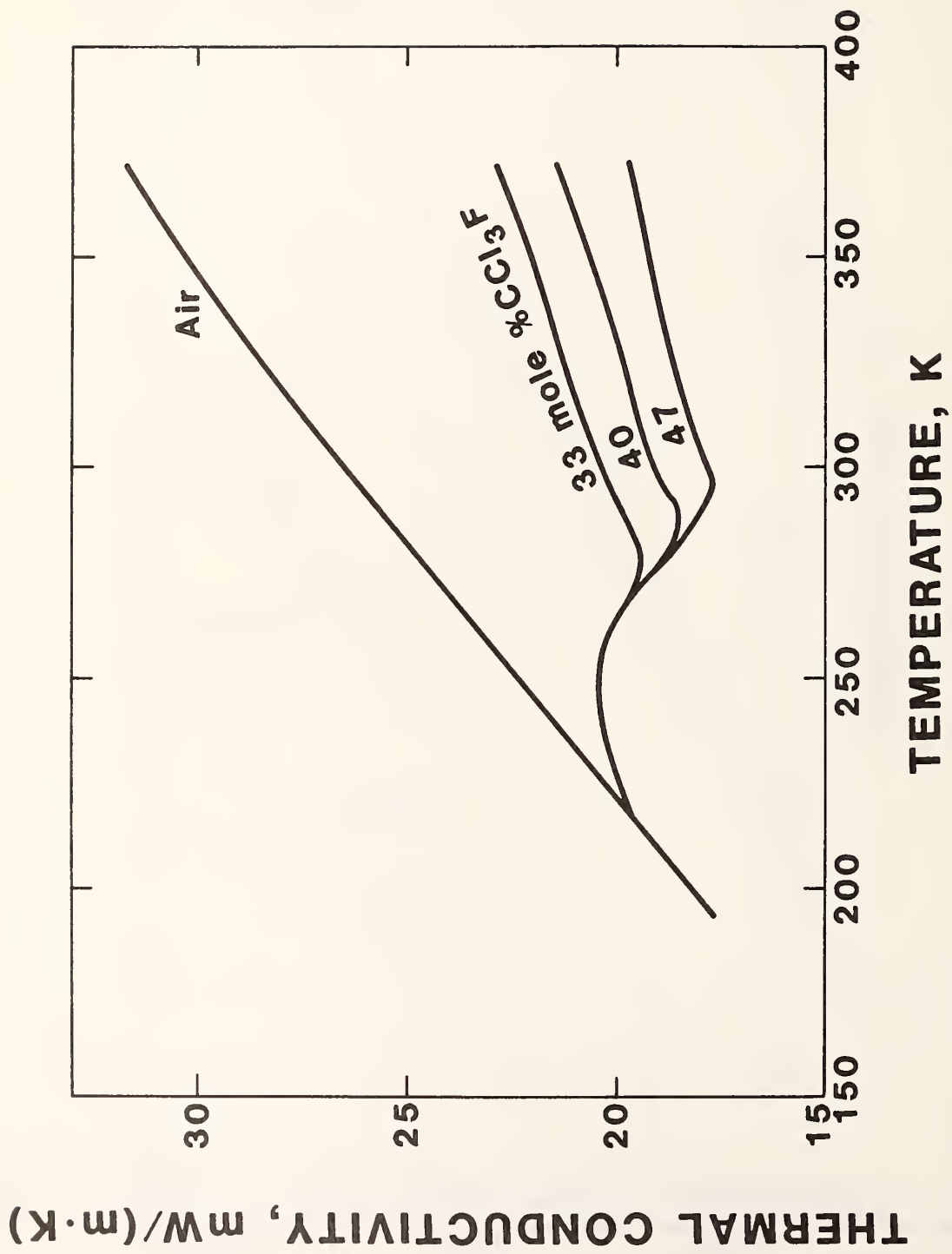


Figure 9. Computed thermal conductivity,  $mW/(m \cdot K)$ , as a function of temperature, K, for air and mixtures of air and  $CCl_3F$  at 293 K (Patten and Skochdopole, 1962).

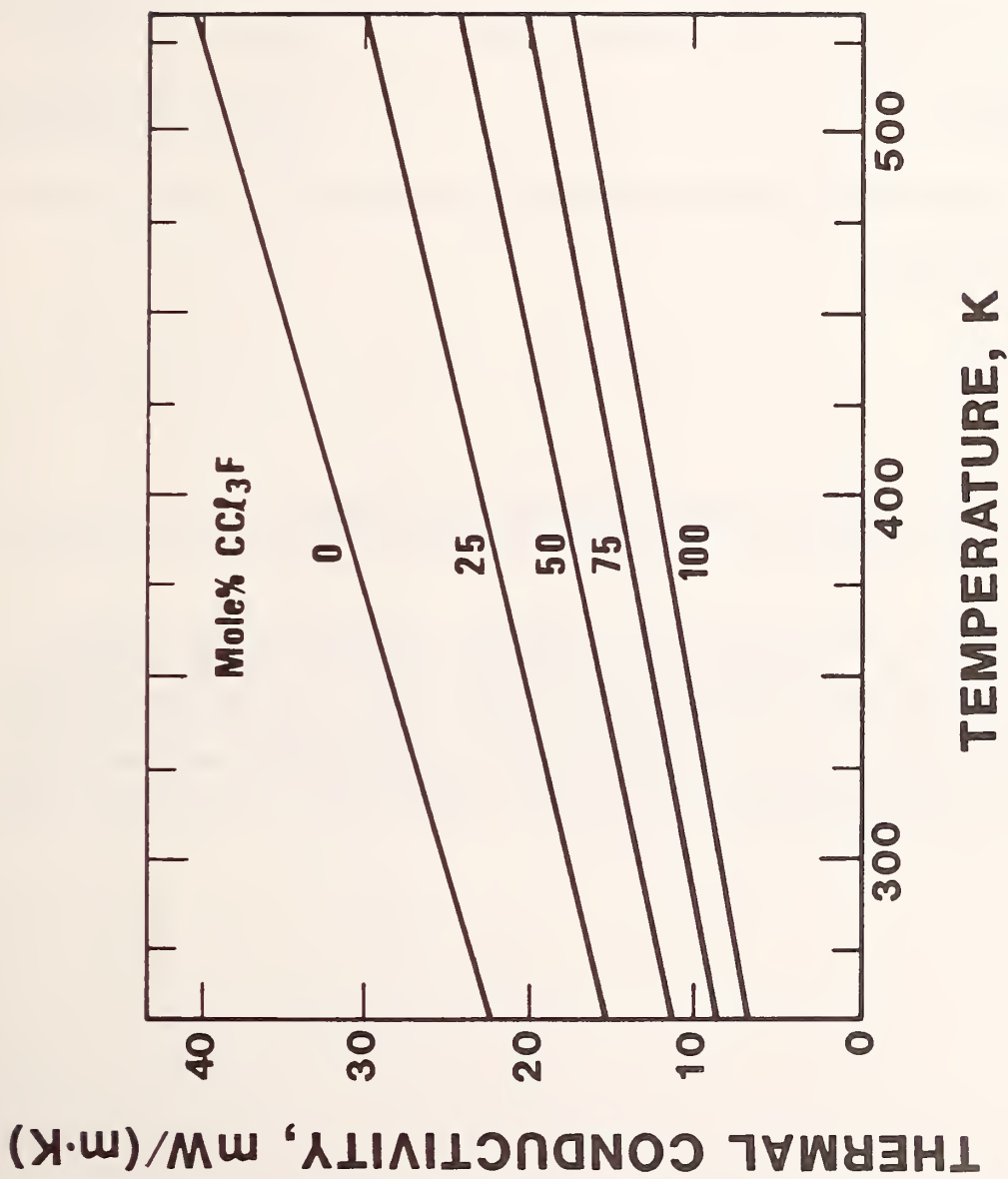


Figure 10. Thermal conductivity,  $mW/(m \cdot K)$ , as a function of temperature, K, for mixtures of  $CCl_3F$  and air (Hammond, 1968).

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11. ABSTRACT <i>(A 200-word or less factual summary of most significant information. If document includes a significant bibliography or literature survey, mention it here)</i>  Values of thermal conductivity for a 32 kg/m <sup>3</sup> , CCl <sub>2</sub> F blown polyurethane foam are presented in the temperature range from 95 to 340 K. These data were obtained using a guarded-hot-plate apparatus (ASTM C 177). The apparent conductivity is discussed in terms of contributions from radiation and solid and gas conduction. Heat transfer via gas convection is not significant when the cell diameters are less than approximately 3 mm; cell dimensions of the thermal conductivity specimen are on the order of 0.5 mm.			
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