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# A Simple Transient Method for Measurement of Thermal Conductivity of Rigid Polyurethane Foams

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**ABSTRACT:** Rigid polyurethane foams (PU) are widely used as thermal insulators in various applications. The thermal conductivity of the foam is the key parameter that governs the efficiency of thermal insulation provided by the foam. The usual technique employed to measure thermal conductivity is based on the rate of steady state heat transfer across a known thickness, induced by two different known temperatures at two opposite surfaces of the foam. We introduce a technique based on the transient measurement of heat transfer measured by an embedded needle probe. This technique is not only rapid but the instrumentation required for such a measurement is simple and the cost is only a fraction of the steady state counterpart. The values of thermal conductivity obtained by both methods are compared and found to agree within 4% over the range of 0.02–0.03 W/mK, which is the usual range of thermal conductivity for commercial rigid PU foams. The sensitivity of the needle probe technique is demonstrated by measuring the thermal conductivity values of foams made with

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Figure 1 appears in color online: <http://cel.sagepub.com>

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various concentrations of chemical blowing agent (water). The present technique is also shown to be effective for measuring the thermal conductivity of small samples, especially, free rise cup foams for which the steady state technique can not be used.

**KEY WORDS:** rigid polyurethane foam, insulation foam, needle probe, transient technique, thermal conductivity measurement, thermal resistance.

## INTRODUCTION

**R**igid polyurethane (PU) foams are a versatile class of polymeric materials with wide range of applications such as thermal insulating agents and construction materials. These foams are prepared by polymerization of a polyol with an isocyanate, in which the reacting mixture is foamed using one or more blowing agents, whose vapor phase thermal conductivity is lower than air. Insulating foams have closed cells which trap the blowing gas, decreasing their thermal conductivity.

The heat transfer through a foam board is governed by various mechanisms such as conduction through the cell walls and the structural elements of the foam called struts, conduction through the entrapped cell gases and thermal radiation [1]. The effect of convection through the cell gases being small, is generally neglected [1]. Detailed studies of various heat transfer mechanisms and effect of various foam properties on the thermal conductivity of board stock foam could be found in the above reference.

Thermal conductivity is the most important property of rigid PU foam. In order to have an accurate estimate of the thermal insulating capacity of the foam, a precise measurement of its thermal conductivity is essential. The conventional technique employed depends on the measurement of steady state heat transfer across a known thickness of the sample, confined between two plates kept at different temperatures. The details of the measurement are given in ASTM C 518. The  $k$ -value is measured from Fourier's law of heat conduction in one dimension. That is,

$$\frac{dQ}{dt} = -kA \frac{dT}{dx}, \text{ which at steady state becomes } q = -kA \frac{\Delta T}{\Delta x} \quad (1)$$

where,  $q$  is the heat flux (J/s),  $k$  is the thermal conductivity of the material (W/mK),  $A$  is the cross-sectional surface area,  $\Delta T$  is the temperature difference between the two sides of a semi-infinite slab, and  $\Delta x$  is the thickness of the slab through which heat transfers. The apparatus establishes steady state one dimensional heat flux through a test specimen between two parallel plates at two constant but different temperatures.

Although this method is widely used for thermal conductivity measurement of foam samples and is the industry standard, it has some inherent disadvantages. The equipments using this principle are expensive and can cost up to 20,000 dollars. Moreover, the sample size required for measurement of thermal conductivity is fairly large so that only slabstock or large molded samples can be tested. Measurement of thermal conductivity of small, lab scale free rise foams (cup foams) is not possible. The time required to get the heat flow to steady state is also high (20–30 min).

We introduced a fast, low cost, transient alternative to the conventional steady state technique. It can be used for varying size of foams, ranging from a small piece of nearly any shape to large molded or slabstock foams. Moreover, the time required to measure the thermal conductivity is considerably reduced. The technique is much cheaper than the presently employed steady state apparatus.

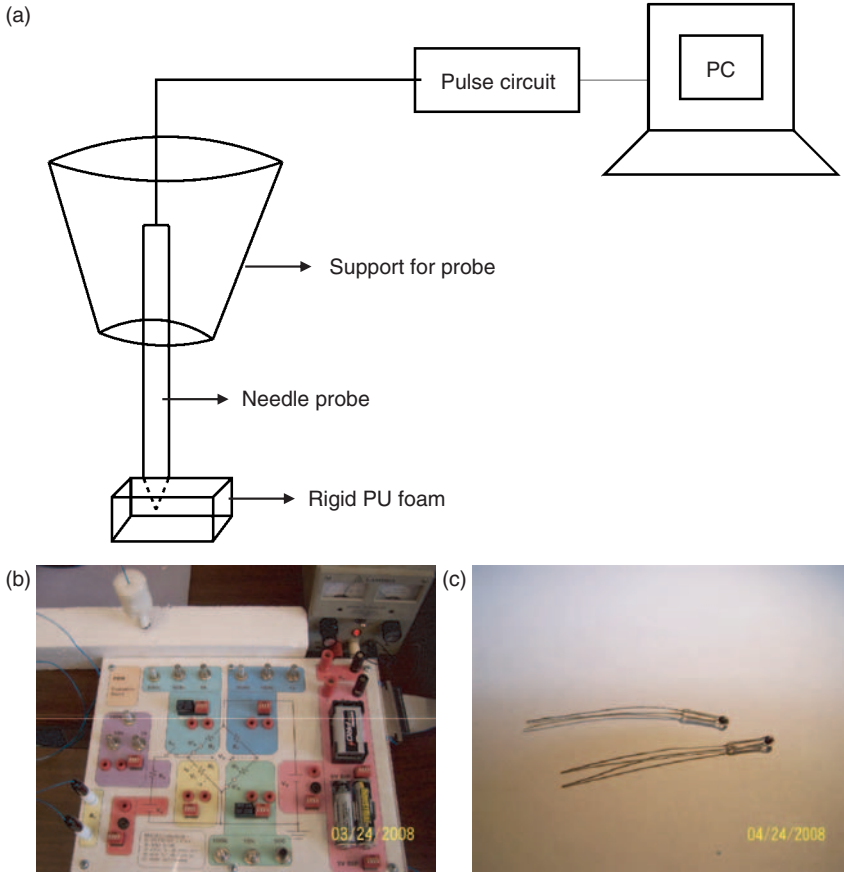
### TRANSIENT MEASUREMENT USING NEEDLE PROBE

The needle probe is a commonly used technique to measure the thermal conductivity and thermal diffusivity of solids and liquids [2]. The biggest advantage is that the time required is generally of the order of seconds to a few minutes for liquids and solids. Several researchers have employed it to measure the thermal conductivity of various materials such as rock core samples [3], soils [4], deep sea sediments [5], laboratory specimens of saturated solids [6], living tissues [7], and food items [8]. In almost all these cases, the thermal conductivity has been obtained to an accuracy of 5% [2]. Several variations of the technique are available for solids and liquids [2,7,9]. We used a technique in which the temperature decay after discharging a short pulse of energy into a probe embedded in the sample is measured [7]. In this technique, the heated volume increases with time and the measurement of thermal conductivity is independent of shape and properties of the probe [7]. The pulse penetration rate depends on the heat diffusivity of the material whose thermal conductivity is to be measured. The theory and solution to the governing equations with various assumptions are given in detail elsewhere [7]. The decay of the temperature at the probe is measured as a function of time and thermal conductivity is calculated using Equation (2) [10].

$$T_0(t) - T_i = \frac{P(\rho C_p)^{0.5}}{4(\pi k)^{1.5}} [(t - t_p)^{-0.5} - t^{-0.5}] \quad (2)$$

where,  $T_0$  is the temperature ( $^{\circ}\text{C}$ ) at the probe center,  $T_1$  is the initial temperature at the probe ( $^{\circ}\text{C}$ ),  $P$  is the electrical power input (W),  $\rho$  is the density of the material ( $\text{kg}/\text{m}^3$ ),  $C_p$  is the specific heat capacity ( $\text{J}/\text{kg}^{\circ}\text{K}$ ),  $k$  is the thermal conductivity ( $\text{W}/\text{m}^{\circ}\text{K}$ ),  $t$  is the elapsed time (s), and  $t_p$  is the duration of pulsed energy (s). The above equation assumes that the pulse is generated from a point source. Since the thermistor used has a finite volume, the initial  $k$ -value measured from the pulse decay will be that of the thermistor itself. However, as time proceeds the pulse penetrates the entire material volume and errors due to assumption of an ideal point source become negligible.

A schematic diagram of the experimental setup for measurement of thermal conductivity is shown in Figure 1(a) and photographs of the entire setup and thermistor bead are shown in Figure 1(b) and 1(c), respectively. The probe consists of a thermistor embedded inside a glass bead (P60BA102K, Thermometrics Corporation, NJ). The glass encasing setup has an outer diameter of 1.5 mm. The thermistor was calibrated at room temperature. A cylindrical tail extends from the thermistor. Figure 2 shows the details of the pulsing and measuring circuit. It is essentially a Wheatstone bridge circuit with heating components. The resistance of the probe ( $R_P$ ) is determined by a multimeter and  $R_1$  is made equal to  $R_P$  by means of a series of potentiometers. Then the value of  $R_2$  and  $R_3$  are set equal and the ratio  $R_2/R_1$  is set at a value between 10 and 30. The reason for setting  $R_2=R_3$  and  $R_1=R_P$  is to obtain a zero voltage across the bridge ( $V_0=0$ ). Then, a change in  $V_0$  becomes a direct indication of a change in  $R_P$  induced due to a temperature drop. The values of  $R_2$  and  $R_3$  should be significantly higher than  $R_1$  in order to prevent a large current flow, which would heat up the thermistor. However, extremely large values of  $R_2$  and  $R_3$  should be avoided, because it will result in a poor signal to noise ratio. The bridge voltage  $V_B$ , was maintained by a dry cell battery (9 V) and the thermistor heating voltage  $V_h$  was maintained by a regulated power supply at 24 V.  $V_B$ ,  $V_h$ , and pulse duration  $t_p$  were measured before each experiment. Power was calculated by measuring the voltage across the thermistor during the pulsing period. The temperature sensing resistance  $R_P$  was  $1\text{ k}\Omega$  ( $\pm 10\%$ ) at room temperature. The pulse timer is represented by a switch ( $S_1$  in Figure 2). After a 2 s delay to measure the initial probe temperature, a pulse with a duration of 2 s was given to thermistor during which the bridge switches remain open. Upon completion of the pulse, the pulsing switch is opened while the bridge switches ( $S_2$  and  $S_3$ ) remain closed and the acquisition of the bridge voltage and the voltage across the thermistor begins. The analog signals from the circuit were converted to digital signals by an A/D



**Figure 1.** (a) Needle probe setup for thermal conductivity measurements, (b) Photograph of needle probe setup and (c) Thermistor bead.

(National Instruments, TX converter) and were analyzed by a data acquisition software (Lab View). Figure 3 shows the temperature decay with time as measured by the circuit. Figure 4 shows a typical thermal conductivity – time curve obtained by fitting the pulse decay to Equation (2). It is seen from Figure 3 that there is a rest time of 2 s and the pulse duration, which is also of 2 s before the decay starts. The emitted pulse penetrates to lesser distances and the higher value of  $k$  shown initially in Figure 4 is for the thermistor – glass bead assembly. As time proceeds, the pulse penetrates to the entire foam volume and the  $k$ -value decreases (because the thermal conductivity of foam is less than

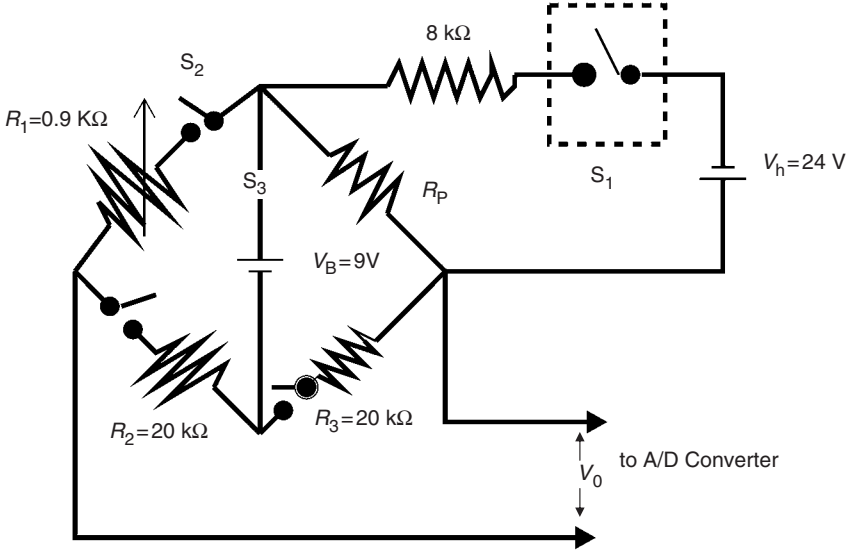


Figure 2. Pulse/temperature measuring circuit.

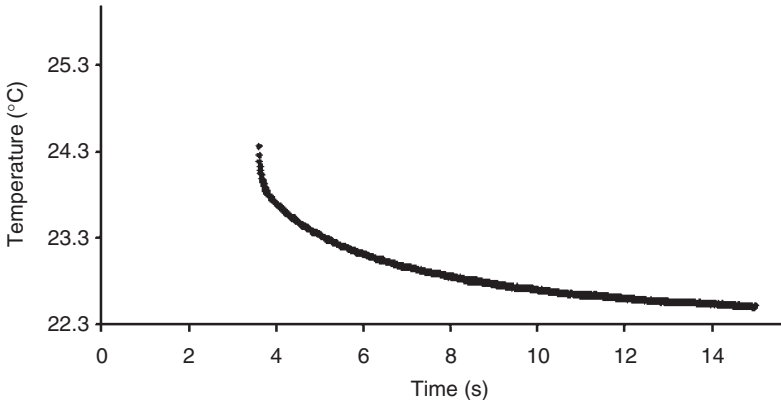
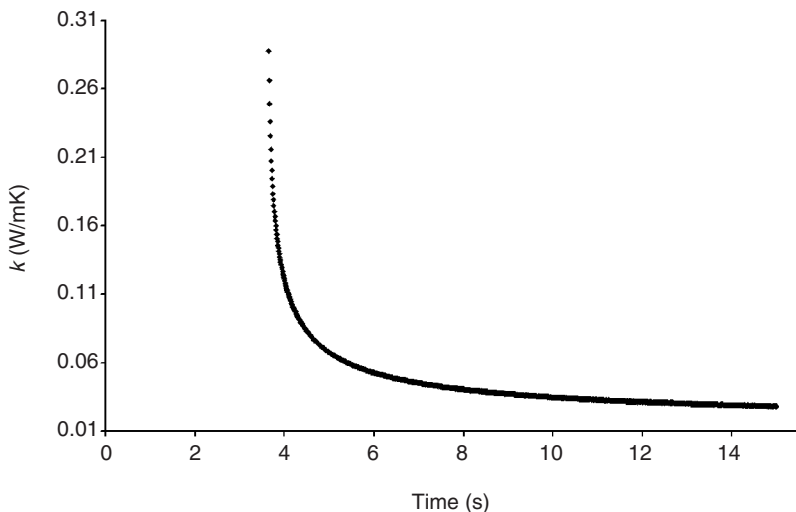


Figure 3. Temperature decay as a function of time after discharge of pulse.

that of the thermistor). Once the pulse penetrates to a sufficient volume of the foam, the curve stabilizes to a plateau for which the  $k$ -value represents the thermal conductivity of the foam. The time at which the thermal conductivity curve reaches a plateau depends on the  $k$ -value of the sample. The lower the  $k$ -value, the more the time required to stabilize. In the current case, the curve did reach a plateau at around 14–15 s.



**Figure 4.** Typical thermal conductivity versus time curve for needle probe method.

## EXPERIMENTAL

### Foaming Process

Rigid PU foams, both molded and free rise were made as follows. A poly ether-based polyol (Jeffol<sup>®</sup> SD361, Huntsman) of hydroxyl value 360 mg KOH/g of polyol and functionality 4.2 was used. The polyol was mixed with catalysts (Polycat<sup>®</sup> 5 and Polycat<sup>®</sup> 8, Air Products), surfactant (Tegostab<sup>®</sup> 8404, Goldschmidt), physical blowing agent (cyclopentane, Sigma Aldrich), and distilled water. The formulation of the polyol blend is shown in Table 1. All these components were stirred at a speed of 2500 rpm for 20 s. Then, polymeric methylenediphenyl diisocyanate, PMDI (Rubinate<sup>®</sup> M, Huntsman) was added to give an isocyanate index of 110. This mixture was stirred at a speed of 2500 rpm for 8 s.

To prepare molded foams, the reacting mixture (~350 g) was poured into a paper-lined wooden mold of size  $32 \times 32 \times 7 \text{ cm}^3$ . The foams were removed from the mold after 30 min. Some molded foams were also made by mixing the components using high pressure impingement mixing equipment i.e., via, reaction injection molding (RIM) process at Huntsman Advanced Technology Centre in The Woodlands, Texas. The molded foams were made at just fill using a  $90 \times 35 \times 5 \text{ cm}^3$  steel mold.

Table 1. Formulation of polyol blend.

Component	Ratio (parts by weight)
Polyol	100
Catalyst 1 (Polycat <sup>®</sup> 8)	2.0
Catalyst 2 (Polycat <sup>®</sup> 5)	0.3
Surfactant (Tegostab <sup>®</sup> 8404)	1.5
Cyclopentane	9.0
Distilled water	1.8

A  $30 \times 30 \times 5 \text{ cm}^3$  rectangular slab was cut from the center of both types of molded sample for thermal conductivity measurement.

Free rise foams were prepared from 40 g of the same formulation used for molded foams. The polyol-foaming agent blend was added to MDI and stirred in a 16 oz. paper cup, at a speed of 2500 rpm for 8 s. The foam was allowed to rise freely.

All the foams were cured at room temperature for at least 24 h before the thermal conductivity tests were performed. Different rigid foams were made by changing the chemical blowing agent concentration (water) and keeping the weight ratio of physical blowing agent concentration and other foaming agents, constant in the polyol blend.

### Thermal Conductivity Measurements

The specific heat capacity ( $C_p$ ) of the foam sample was measured by differential scanning calorimetry. Density was measured by weighing a sample of known dimensions after the surface skin was removed. In case of molded foams, three needle probe measurements were made for each foam, along the diagonal across the top surface (the rise direction being parallel to thickness). The probe was inserted at the center and at approximately  $\pm 3 \text{ cm}$  away. The average of the three thermal conductivity values is considered the effective thermal conductivity of the foam.

In case of free rise foams, measurements were done at top and bottom of the foam. Measurement at the top was done at a distance of 0.5 cm beneath the point where the foam rose above the top of the cup. Measurements at the bottom were done 0.5 cm from the base of the foam after removing the foam from the paper cup. Two measurements were made at random points at both top and bottom of the foam.

For comparing the performance of needle probe with conventional technique, a steady state thermal conductivity analyzer (Fox 150,

LaserComp) was used to measure  $k$  on several of the molded samples. Only one measurement per sample was made.

**RESULTS AND DISCUSSION**

Table 2 shows the values of thermal conductivity obtained for molded foams by conventional steady state and needle probe techniques. Samples 1 and 2 were made by hand mixing and tested first by needle probe followed by the steady state method. Samples 3–5 are RIM-made foams, which were tested initially by the steady state (conventional) method followed by needle probe. From Table 2, it is clear that the thermal conductivity values obtained by the needle probe technique are very close to those obtained by the steady state method. The average values by the needle probe are 2–4% higher and the difference from the steady state is <5%, which is the uncertainty given in ASTM C518.

Table 3 shows the thermal conductivity values obtained for molded foams made with varying water concentration. It is seen from the table

*Table 2. Thermal conductivity values of molded foam by steady state and transient methods.*

Foam sample	$k$ -steady state (W/mK)*	$k$ -needle probe (W/mK)			
		$K_1$	$K_2$	$K_3$	$k$
Sample 1	0.0270	0.0279	0.0279	0.0277	0.0278 ± 0.0002
Sample 2	0.0245	0.0253	0.0255	0.0255	0.0254 ± 0.0002
Sample 3	0.0233	0.0240	0.0240	0.0238	0.0239 ± 0.0001
Sample 4	0.0231	0.0235	0.0235	0.0235	0.0235 ± 0.0000
Sample 5	0.0207	0.0215	0.0211	0.0217	0.0214 ± 0.0003

\*The uncertainty of this method is 5% (ASTM C518).

*Table 3. Thermal conductivity values of molded foam with varying water concentration.*

Core density (kg/m <sup>3</sup> )	Water/ Cyclopentane	$k_1$ (W/mK)	$k_2$ (W/mK)	$k_3$ (W/mK)	$k$ (W/mK)
		38.2	0.200	0.0242	0.0242
38.1	0.211	0.0246	0.0246	0.0246	0.0246 ± 0.0000
37.9	0.233	0.0250	0.0257	0.0254	0.0254 ± 0.0003
37.9	0.255	0.0262	0.0266	0.0262	0.0264 ± 0.0002
37.5	0.278	0.0279	0.0279	0.0284	0.0281 ± 0.0003

Table 4. Thermal conductivity values of free rise cup foams measured at top and bottom.

Sample	$k_{top1}$ (W/mK)	$k_{top2}$ (W/mK)	$k_{top}$ (W/mK)	$k_{bottom1}$ (W/mK)	$k_{bottom2}$ (W/mK)	$k_{bottom}$ (W/mK)
Foam 1	0.0272	0.0272	0.0272	0.0264	0.0266	0.0265
Foam 2	0.0270	0.0271	0.0271	0.0262	0.0262	0.0262

that the thermal conductivity values increase with water concentration. With more water in the polyol blend, the amount of CO<sub>2</sub> generated by blowing reaction is higher and consequently the concentration (and partial pressure) of CO<sub>2</sub>, which has a higher thermal conductivity than the physical blowing agent (cyclopentane) is higher, contributing to the increase in thermal conductivity of foam. This shows that the needle probe method is efficient in probing small variations in the thermal conductivity of molded foam.

Table 4 shows the thermal conductivity of two free rise foams made using the same formulation, using the needle probe technique. The good reproducibility is another confirmation of our hand mixing method. Measurement of thermal conductivity of free rise foams (cup foams) by the conventional steady state method is impossible because of its minimum size requirement of about  $20 \times 20 \times 5 \text{ cm}^3$ . From the table, it is seen that thermal conductivity at the top is higher than that at bottom for both the samples. This is due to larger size of the cells at the top of free rise foam in comparison to those at the bottom caused by bubble coalescence [11]. Smaller cells will induce lower thermal conductivity since the same amount of gas is entrapped in more number of cells so that the concentration difference of the gas between adjacent cells is reduced, thus the rate of diffusion is also reduced [12,13].

## CONCLUSIONS

A transient method has been introduced for measurement of thermal conductivity of rigid foams. The method is much faster and of lower cost than the conventional steady state technique, yet the two methods gave excellent agreement, within the accuracy of the steady state technique. Measurements made on thermal conductivity of rigid molded foams with varying water concentrations show that the needle probe technique is suitable for measuring small changes in thermal conductivity induced due to subtle changes in formulation. The needle probe technique is also suitable for measuring the thermal conductivity of free rise cup foams,

which is impossible by the conventional technique due to the small size of the test specimen.

### ACKNOWLEDGMENTS

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### REFERENCES

1. Zhu, Z. (2007). Modeling of Expansion and Collapse of Extruded Filamentary Foam through Cell-to-Cell Diffusion, PhD Thesis, Chapter 5, University of Toronto.
2. Asher, G.B., Sloan, E.D. and Graboski, M.S. (1986). A Computer Controlled Transient Needle – Probe Thermal Conductivity Instrument for Liquids, *International Journal of Thermo physics*, **7**(2): 285–294.
3. Bloomer, J.R. and Ward, J. (1979). Semi Automatic Field Apparatus for the Measurement of Thermal Conductivities of Sedimentary Rocks, *Journal of Physics E – Scientific Instruments*, **12**(11): 1033–1035.
4. Mitchell, J.K., Asce, F. and Kao, T.C. (1978). Measurement of Soil Thermal Resistivity, *Journal of Geotechnical Engineering*, **104**: 1307–1320.
5. Stoll, R.D. and Bryan, G.M. (1979). Physical Properties of Sediments Containing Gas Hydrates, *Journal of Geophysics Research*, **84**: 1629–1634.
6. Woodside, W. and Messmer, J.H. (1961). Molecular Effects in Heat Conduction Through Porous Rocks, *Journal of Applied Physics*, **65**: 3481–3485.
7. Chen, M.M., Holmes, K.R. and Rupinskas, V. (1981). Pulse-decay Method for Measuring the Thermal Conductivity of Measuring Tissues, *Journal of Biomechanical Engineering*, **103**: 253–260.
8. Cogne, C., Andrieu, J., Laurent, P., Besson, A. and Nocquet, J. (2003). Experimental Data and Modelling of Thermal Properties of Ice Creams, *Journal of Food Engineering*, **58**(4): 331–341.
9. Waite, W.F., Gilbert, L.Y., Winters, W.J. and Mason, D.H. (2006). Estimating Thermal Diffusivity and Specific Heat from Needle Probe Thermal Conductivity Data, *Review of Scientific Instruments*, **77**: 044904-1-5.
10. Carslaw, H. and Jaeger, J. (1959). *Conduction of Heat in Solids*, Clarendon Press, Oxford.
11. Yasunaga, K., Neff, R.A., Zhang, X.D. and Macosko, C.W. (1996). Study of Cell Opening in Flexible Polyurethane Foams, *Journal of Cellular Plastics*, **32**(5): 427–448.
12. Widya, T. and Macosko, C.W. (2005). Nanoclay-modified Rigid Polyurethane Foam, *Journal of Macromolecular Science; Physics*, **844**(6): 897–908.
13. Wilson, F.G., Ulrich, H. and Riese, W. (eds) (1992). *Reaction Polymers*, Hanser Publishers, Munich.