Measurement of the Thermal Conductivity of Unfrozen and Frozen Food Materials by a Steady State Method with Coaxial Dual-cylinder Apparatus

Runghnaphar Pongsawatmanit,* Osato Miyawaki,** and Toshimasa Yang***
Department of Agricultural Chemistry, The University of Tokyo, 1-1-1 Yayoi, Bunkyo-ku, Tokyo 113, Japan
Received September 22, 1992

Coaxial dual-cylinder apparatus was used to measure the effective thermal conductivity of aqueous solutions of glucose, sucrose, gelatin and egg albumin over a temperature range from −20°C to 20°C by the steady state method. The accuracy of the apparatus was confirmed by testing with water and ice. The effective thermal conductivity decreased with an increase in the total solid content in both the frozen and unfrozen states. In the unfrozen state, the effective thermal conductivity was slightly dependent on temperature. In the frozen state, however, the effective thermal conductivity was strongly dependent on temperature; lower temperatures gave higher effective thermal conductivity, reflecting the increase in the ice fraction. For the unfrozen samples, the intrinsic thermal conductivity of each solid component was calculated by heat transfer models. All the models tested, series, parallel and Maxwell–Eucken, were equally applicable to describe the heat conduction in the unfrozen state. In the frozen state, however, the strong temperature dependency of the effective thermal conductivity suggests that the effect of the temperature dependency of the ice fraction should be incorporated into theoretical models.

The thermal conductivity of food materials in the frozen state, in spite of many investigations in the literature, is not fully understood yet because the thermal conductivity of a materials containing water is dependent on such factors as the composition, heterogeneous structure, intrinsic property of each component, and the ice fraction.

To measure the thermal conductivity, the steady state methods and transient methods are both applicable. For frozen samples, however, the steady state method is recommended because of the strong temperature dependency of the ice fractions, which strongly influences the apparent heat capacity with the transient method.

For a steady state analysis, the guarded hot plate method is well accepted and has been used for determining the thermal conductivity of meats, fats, and gelatin gel, although this method involves a complicated and expensive control system. In comparison, the coaxial dual-cylinder method is simpler and much more convenient, provided that the sample can be loaded into the annular space between the two cylinders. This method has been applied to measure the thermal conductivity of liquids, paddy grain, and dried skim milk.

The present work investigates the effective thermal conductivity of aqueous solutions of gelatin, egg albumin, glucose and sucrose by the steady state method, using coaxial dual-cylinder apparatus. The temperature dependency of the effective thermal conductivity is thoroughly analyzed in the unfrozen to frozen temperature range. For the unfrozen state, the intrinsic thermal conductivity of each solid component is analyzed by heat transfer models.

Theory

The equation for heat conduction, when expressed in cylindrical coordinates, is

\[
\frac{\partial T}{\partial t} = \alpha \left( \frac{\partial^2 T}{\partial r^2} + \frac{1}{r} \frac{\partial T}{\partial r} + \frac{1}{r^2} \frac{\partial^2 T}{\partial \theta^2} + \frac{\partial^2 T}{\partial z^2} \right)
\]

(1)

If a circular cylinder whose axis coincides with the z axis is heated, and the initial and boundary conditions are independent of coordinates \( \theta \) and \( z \), the temperature will be a function of \( r \) and \( t \) only, and the equation reduces to

\[
\frac{\partial T}{\partial t} = \frac{1}{r} \frac{\partial}{\partial r} \left( ra \frac{\partial T}{\partial r} \right)
\]

(2)

For a hollow cylindrical sample, whose inner and outer radii are \( r_1 \) and \( r_2 \), Eq. (2) in the steady state will be

\[
\frac{d}{dr} \left( r \frac{dT}{dr} \right) = 0, \quad r_1 < r < r_2
\]

(3)

The general solution of this with boundary condition of 
\( T(r_1) = T_1 \) and \( T(r_2) = T_2 \) is

\[
T = (B \ln r + C)
\]

(4)

\[
B = \frac{(T_1 - T_2)}{\ln (r_1/r_2)}
\]

(5)

Fourier's Law becomes

\[
Q = -\lambda_e (2\pi r L) \frac{dT}{dr} = -\lambda_e (2\pi L) T = \frac{2\pi \lambda_e L (T_1 - T_2)}{\ln (r_2/r_1)}
\]

(6)

\[
\lambda_e = \frac{Q L (r_2/r_1)}{2\pi L (T_1 - T_2)}
\]

(7)

where \( \lambda_e \) = effective thermal conductivity (W/m·K), \( Q/L \) = heat flux per unit length (W/m), \( r_1 \) = radius of the inner...
cylinder (m), $r_2 =$ radius of the outer cylinder (m), $T_1 =$ temperature at the surface of the inner cylinder (m), and $T_2 =$ temperature at the surface of the outer cylinder (m).

Therefore, by measuring heat flux ($Q$), $T_1$ and $T_2$, the effective thermal conductivity ($\lambda_e$) can be obtained from Eq. (7).

**Materials and Methods**

**Materials.** Gelatin was obtained Nacalai Tesque, sucrose and glucose being supplied by Kanto Chemical, and egg albumin by Difco. The materials were used as supplied. For the unfrozen samples, 0.3% (w/w) agar (Nacalai Tesque) was added to prevent the convection effect during measurements.

**Apparatus.** Figure 1 schematically shows the system for measuring the thermal conductivity by the coaxial dual-cylinder apparatus. The heater had a uniformly distributed resistance of 1 Ohm/cm and was inserted along the axis of the inner cylinder (110 mm long and 15 mm in diameter) to provide a heat flux in the radial direction. The sample being tested was loaded into the annular space between the inner cylinder and the outer cylinder (37 mm in inner diameter and 122 mm long), both ends of which were fitted with Teflon stoppers to provide thermal insulation. The coaxial dual-cylinder apparatus was placed in a coolant (ethylene glycol), the temperature of which was controlled by a thermostat. DC powder was supplied by Hewlett Packard 6034A equipment, and the voltages across the standard resistance and the heater were measured with a digital voltmeter (Hewlett Packard 3456A). One and three copper-constantan thermocouples were embedded at the surfaces of the inner and outer cylinders, respectively, to measure temperature. The power transmitted to the heater was calculated from the product of the electric current and the voltage drop over the heater. A four-wire system was used, two wires for supplying the measuring current to the heater, and the other two wires for measuring the voltage drop across the heater.

**Measurement of the effective thermal conductivity.** A sample was dissolved in distilled water by heating up to boiling in a flask. The sample was then degassed under vacuum to remove air bubbles for 10 min and poured into the annular space of the apparatus, ca. 92% of which, in volume, was used when allowing for the volume expansion during freezing. The sample was frozen by rapidly immersing the apparatus in the coolant. The same samples of gelatin and egg albumin were used in both the frozen and unfrozen states. For the carbohydrate samples, 0.3% agar (w/w) was added to prevent the convection effect during measurement in the unfrozen state.

**Intrinsic thermal conductivity of the solids.** The intrinsic thermal conductivity of each solid component was determined by using the series, parallel and Maxwell–Eucken$^{16}$ models for heat conduction.

**Results and Discussion**

**Coaxial dual-cylinder method for measuring thermal conductivity.** The coaxial dual-cylinder apparatus was tested for accuracy with pure water containing 0.5% agar to prevent the convection effect during the experiments in both the frozen and unfrozen states from $-20^\circ$ to $20^\circ$C. The temperature difference across the sample was changed by varying the current applied to the heater at about 0.2, 0.3, 0.4, and 0.5 ampere to determine the optimum temperature difference.

The effect of this temperature difference throughout the sample on the observed thermal conductivity of ice was tested by using pure water with a coolant temperature of $-15^\circ$ and $-10^\circ$C. The observed thermal conductivity of ice and the temperature difference as a function of heat flux are shown in Fig. 2. It was found that, for ice, a temperature difference between 0.6$^\circ$ and 2$^\circ$C did not have a significant effect on the observed thermal conductivity. Lentz$^{21}$ has recommended a temperature difference of 2–3$^\circ$C with the guarded hot-plate methods.

The experimental error in thermal conductivity with water containing 0.3% (w/w) agar was less than 2%, while it was about 10% for ice. This might have been due to the “film” or “contact” resistance at the interface between the sample and the cylinder, especially in the ice phase.$^{17,18}$

The effect of this film resistance would be more significant for ice than for water because the thermal conductivity of ice is four times higher than that of water. Therefore, the

---

**Fig. 1.** Apparatus for Measuring the Effective Thermal Conductivity.

**Fig. 2.** Effect of Heat Flux on the Apparent Thermal Conductivity of Ice and the Temperature Difference across a Sample.

**Fig. 3.** Comparison of the Experimental Thermal Conductivity Values for Ice and Water with Those from the Literature.$^{19}$
apparatus was calibrated with the film resistance \( (R) \) for this apparatus based on the literature value for ice\(^{19}\) by using the following equation:

\[
R = \frac{2\pi L(r_2 - r_1)}{\ln \left( \frac{r_2}{r_1} \right)} \frac{\Delta T}{Q} \frac{(r_2 - r_1)}{\lambda_c}
\]

(8)

The thermal conductivity values for ice and water after calibrating the skin resistance by Eq. (8) are shown in Fig. 3, being in good agreement with those in the literature.\(^{19}\) This shows that the apparatus was reliable for determining the thermal conductivity of ice and water over a temperature range of -20° to 20°C.

The present apparatus assumes a uniform heat flux in the radial direction of the cylinder from the axis to the sample. To prove this assumption, the overall heat flux in both the radial and axial directions was estimated from the size and from the thermal properties of the sample and stoppers at each end. The values differed by at least 20-fold. In addition, the temperature difference across the sample was measured at the midpoint of the apparatus, where the effect of the axial heat flux is minimal. Therefore, the effect of heat flux in the axial direction is considered to have been negligible.

For the steady-state measurement of thermal conductivity, the guarded hot-plate method is well accepted. However, it involves a complicated control system to provide a uniform heat flux from one side of the plate to the other side. Compared with this, the coaxial dual-cylinder apparatus is much simpler and more convenient. For thermal conductivity measurement, the transient method is also frequently employed for its simplicity and convenience. The method, however, is of limited use for frozen samples because of the effect of latent heat. In food materials, the ice fraction is strongly dependent on the temperature, so that the effect of latent heat cannot be neglected with the unsteady state method when used in the frozen state.

**Effective thermal conductivity values**

Figures 4, 5, 6, and 7 show the effective thermal conductivity values for gelatin gel, egg albumin gel, sucrose and glucose solutions, respectively, at various concentrations in the temperature range from -20° to 20°C. These figures show that the effective thermal conductivity above the freezing point slightly decreases with increasing concentration due to the lower intrinsic thermal conductivity of the solute compared with that of water. The temperature dependency of the effective thermal conductivity is weak, reflecting the weak temperature dependency of the intrinsic thermal conductivity of both water and the solutes.

In the frozen state, however, the effective thermal conductivity is strongly dependent on both the concentration and temperature. The thermal conductivity decreases with increasing concentration and also decreases with increasing temperature. This is because the ice fraction
Table: Intrinsic Thermal Conductivity (W/m·K) of Solute Components According to Different Heat Conduction Models for Samples in the Unfrozen State

<table>
<thead>
<tr>
<th>Model</th>
<th>Gelatin</th>
<th>Egg albumin</th>
<th>Glucose</th>
<th>Sucrose</th>
</tr>
</thead>
<tbody>
<tr>
<td>Series</td>
<td>0.340</td>
<td>0.377</td>
<td>0.351</td>
<td>0.345</td>
</tr>
<tr>
<td>Parallel</td>
<td>0.237</td>
<td>0.299</td>
<td>0.257</td>
<td>0.257</td>
</tr>
<tr>
<td>Maxwell–Eucken</td>
<td>0.280</td>
<td>0.331</td>
<td>0.295</td>
<td>0.293</td>
</tr>
</tbody>
</table>

decreases with increasing concentration and temperature, the thermal conductivity of ice being about four times greater than that of water. Therefore, an easy-to-freeze sample gives a higher effective thermal conductivity in the frozen state.

**Intrinsic thermal conductivity of the solutes**

From the effective thermal conductivity in the unfrozen state, the intrinsic thermal conductivity of each solid component was determined by using the series, parallel and Maxwell–Eucken models. The results are shown in Table. As the intrinsic thermal conductivity values are dependent on the model, the values cannot be 'intrinsic' in the strict sense. The true intrinsic value, however, can be found among these values obtained. These values are all useful in practice, and are therefore referred to as the intrinsic thermal conductivity in this paper. In calculating the intrinsic thermal conductivity, the values of density used were 997.2, 1330, and 1599 kg/m³ for water, proteins and carbohydrates, respectively.¹⁹ The intrinsic thermal conductivity from the parallel model had the lowest value while that from the series model was the highest, the Maxwell–Eucken model giving an intermediate value. All the intrinsic thermal conductivity values obtained are lower than those of water (0.588 W/m·K at 10°C) and ice (2.385 W/m·K at −20°C), which explains the concentration dependency of the effective thermal conductivity both in the unfrozen and frozen states. Sakiyama et al.²⁰ have obtained a value for the intrinsic thermal conductivity of gelatin of about 0.3 W/m·K from both the parallel and series models, based on steady state data. Kong et al.,²¹ based on unsteady state measurements, obtained a value for the intrinsic thermal conductivity of an unfrozen gelatin sample of about 0.3 W/m·K from the series model. Andrieu et al.²² also determined the intrinsic thermal conductivity of 0.31 W/m·K for gelatin by the hot-wire probe method.

Figure 8 shows a correlation between the effective thermal conductivity values observed and calculated from each model. Good correlation was obtained between the experimental and theoretical results with no significant difference among the three models employed, showing that any heat conduction model can be used for the unfrozen sample to predict the effective thermal conductivity, as long as it is combined with the inherent intrinsic thermal conductivity. This is probably due to the relatively small difference in the absolute value for the intrinsic thermal conductivity of solutes (0.23–0.36 W/m·K) from that of water (0.588 W/m·K). In the frozen state, however, the effective thermal conductivity is dependent not only on the model and the intrinsic thermal conductivity, but also on the temperature-dependent ice fraction. Figure 9 shows the effective thermal conductivity value for solutions at

-15°C as a function of the concentration. The figure shows greatly different values for sucrose and glucose at the same concentration. This was not expected from the analytical models employed here, because the intrinsic thermal conductivity values for these materials are not very different from each other (Table), but probably results from the difference in the amount of the ice fraction. Figure 9 shows that the ice fraction for sucrose was higher than that for glucose at the same concentration, which agrees with the results reported in the literature.²³

The ice fraction seems to have been lower for the solutions with lower molecular weight. For a more precise analysis of the thermal conductivity in the frozen state by a model, the ice fraction and its temperature dependency needs to be determined. This is presently under investigation by our group.

In conclusion, the coaxial dual-cylinder apparatus was conveniently applied to measuring the effective thermal conductivity of food solutions with a reasonable accuracy. The temperature dependency of the effective thermal conductivity was greatly different between the unfrozen and frozen states. In the unfrozen state, the effect of temperature was not great for the gelatin gel, egg albumin gel, and glucose and sucrose solutions, and the intrinsic
thermal conductivity for each component was calculated by using the series, parallel and Maxwell-Eucken models for heat conduction. In the frozen state, however, the effect of temperature on the effective thermal conductivity was considerable, suggesting a strong effect of the temperature dependency of the ice fraction.

References