TEMPERATURE DEPENDENCE OF THE EFFECTIVE THERMAL CONDUCTIVITY OF FOOD GELS IMPREGNATED WITH AIR BUBBLES

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Air-impregnated alginate and ovalbumin gels were prepared as model systems for porous foods, and their effective thermal conductivity was measured at 30-70°C by a steady-state method. The effective thermal conductivity of the air-impregnated gels increased with temperature more greatly than that of gels containing no air bubbles. For effective thermal conductivity of air-impregnated gels, a theoretical model which included latent heat transport accompanied by water vapor diffusion in addition to heat conduction was presented and was compared with experimental data. The model gave a good approximation to the experimental data up to 50°C. At higher temperature, however, the model overestimated the effective thermal conductivity, especially for gels of high porosity.

Introduction

Knowledge of the effective thermal conductivity of food materials is important not only for process design but also for the prediction and control of various changes occurring in food materials during thermal processing. For such porous foods as baked products, however, the information regarding the effective thermal conductivity is scarce⁷⁾.

The effects of bulk density and moisture content on the effective thermal conductivity of dough and baked products have been investigated by a few researchers. Unklesbay *et al.*¹⁰⁾ measured the effective thermal conductivity of bread after baking, and discussed the effects of the bulk density and moisture loss on the effective thermal conductivity. Bakshi and Yoon¹⁾ also correlated the effective thermal conductivity of bread after baking to its density and moisture content. Griffith³⁾ reported that the effective thermal conductivity of Mexican corn-based tortilla dough was affected mostly by temperature, processing time, and moisture content. However, the temperature dependence of the effective thermal conductivity has not yet been clearly understood.

In our previous work⁹⁾, gelatin gels impregnated with air bubbles were used as model systems to analyse the effective thermal conductivity of such porous foods. The results suggested that, in air-impregnated gels of high water content, contribution of latent heat transport accompanied by water vapor diffusion to the effective thermal conductivity was not negligible even at 20°C. At

high temperature, such latent heat transport will affect the effective thermal conductivity more greatly because of the increase in vapor pressure of water.

The objective of this paper is to study the temperature dependence of the effective thermal conductivity of gels impregnated with air bubbles. A model for predicting the effective thermal conductivity of air-impregnated gels is developed and compared with the experimental results.

1. Theoretical Model

If heat is transferred only by conduction through a dispersed system of spheres of various sizes, and if $0.01 < \lambda_c / \lambda_d < 100$, the effective thermal conductivity of the system can be predicted by the following Maxwell-Eucken model⁸⁾.

$$\frac{\lambda_e}{\lambda_c} = \frac{\lambda_d + 2\lambda_c - 2\phi(\lambda_c - \lambda_d)}{\lambda_d + 2\lambda_c + \phi(\lambda_c - \lambda_d)} \quad (1)$$

In porous food materials of high water content, however, the contribution of a heat-transfer mechanism other than conduction is not negligible⁹⁾. In such foods, water vapor diffuses in a pore space according to the gradient of water vapor pressure caused by the temperature gradient. At the same time, evaporation and condensation of water take place at the interface of the pore as illustrated in **Fig. 1**. The latent heat of evaporation of water will be thus conveyed in the pore.

The latent heat transport accompanied by the water vapor diffusion can be evaluated as shown in the following equivalent thermal conductivity⁶⁾.

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Fig. 1 Latent heat transport accompanied by water vapor diffusion in gels impregnated with air bubbles

$$\lambda_v = \frac{D}{RT} \frac{P}{P - P_v} L \frac{dP_v}{dT} \quad (2)$$

The partial pressure of water vapor, P_v , is related to the saturated vapor pressure of water, P_s , and the water activity of the material, a_w , as follows.

$$P_v = a_w P_s \quad (3)$$

If the temperature dependence of the water activity is neglected for simplicity, Eq. (2) results in the following equation.

$$\lambda_v = \frac{D}{RT} \frac{P}{P - a_w P_s} L a_w \frac{dP_s}{dT} \quad (4)$$

Figure 2 shows the values of λ_v calculated from Eq. (4). In the calculation, the partial pressure of air, $P - a_w P_s$, was assumed to be 0.10 MPa. The value of λ_v increased with the increase in temperature and/or water activity, and exceeded the thermal conductivity of water, λ_{water} , at high temperature. This suggests that the contribution of the latent heat transport will be of great importance for overall heat transfer in porous foods, especially at high temperature.

The latent heat transport will occur simultaneously with heat conduction through the gas phase. Assuming that these two mechanisms are independent, the overall thermal conductivity of the pore will be represented as the sum of λ_d and λ_v . The effective thermal conductivity of porous foods may be evaluated using the Maxwell-Eucken model with this overall thermal conductivity of the pore. The replacement of λ_d in Eq. (1) by $(\lambda_d + \lambda_v)$ results in the following equation.

$$\frac{\lambda_e}{\lambda_c} = \frac{(\lambda_d + \lambda_v) + 2\lambda_c - 2\phi(\lambda_c - (\lambda_d + \lambda_v))}{(\lambda_d + \lambda_v) + 2\lambda_c + \phi(\lambda_c - (\lambda_d + \lambda_v))} \quad (5)$$

2. Experimental

2.1 Materials

Sodium alginate was purchased from Kanto Chemical Co., Inc. Ovalbumin (crude) and gelatin were pur-

Fig. 2 Equivalent thermal conductivity for latent heat transport, λ_v , calculated from Eq. (4)

chased from Nacalai Tesque, Inc. These were used without further purification. Other materials were of analytical grade.

2.2 Preparation of air-impregnated alginate gels

Five grams of sodium alginate and 2.5 g of gelatin were dissolved in 375 ml of distilled water and incubated at 90°C. The presence of gelatin made it possible to prepare gels of high porosity. Gelation of the mixture was initiated with the addition of an aqueous solution containing Ca^{2+} and citrate, which was prepared by mixing 35 ml of 5% $\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$ solution and 90 ml of 5% sodium citrate ($\text{Na}_3\text{C}_6\text{H}_5\text{O}_7 \cdot 2\text{H}_2\text{O}$) solution for 1 min at 90°C. Citrate acts as a chelating agent for Ca^{2+} and prevents rapid partial gelation of alginate⁴⁾. Into the mixture, fine air bubbles were dispersed with a mixer (Mixmaster, Sunbeam) at room temperature. As gelation of the mixture proceeded, bubble migration was retarded. The air-impregnated mixture was then packed in a sample container (160 mm in inside diameter, 20 mm in height) and stored overnight at 5°C.

2.3 Preparation of air-impregnated ovalbumin gels

Fine air bubbles were dispersed into a 30% ovalbumin solution with a rotating impeller, and the mixture was heated up to 60°C with continuous stirring. As the thermal denaturation of ovalbumin proceeded, bubble migration was retarded. The air-impregnated mixture was then packed in the sample container, solidified at 80°C for 1 h, and stored overnight at 5°C.

2.4 Measurement of effective thermal conductivity

Effective thermal conductivity of each gel was measured by a steady heat flow method with a Shotherm RTM-G5 (Showa Denko Co., Ltd.). The method was the same as described in our previous paper⁸⁾. In this measurement, the temperature difference between the upper and lower surfaces of the sample container was controlled to be constant. When a steady state was reached, heat fluxes at the upper and lower surfaces of the container were measured with two heat-flow meters and

Fig. 3 Effective thermal conductivity of air-impregnated alginate gel at 40°C in comparison with Eqs. (1) and (5)

averaged. Temperatures at the upper and lower surfaces were also measured with chromel-armel thermocouples to obtain the temperature gradient applied to the sample. The thermal conductivity was calculated from the values of the average heat flux and the temperature gradient. The mean temperature of the sample was varied in the range of 30–70°C, and the temperature gradient applied to the sample was 240–300 K/m. Under these experimental conditions, convective and radiative heat transfer in air-impregnated gels was regarded as negligible.

2.5 Volume fraction of gas phase

The volume fraction of gas phase dispersed in each gel was determined from the density of the gel.

3. Results and Discussion

Figure 3 shows the effective thermal conductivity of air-impregnated alginate gels at 40°C as a function of the volume fraction of gas phase. The effective thermal conductivity gradually decreased with the increase in volume fraction of gas phase. For comparison, values calculated from Eqs. (1) and (5) are also shown in Fig. 3. In the calculation with Eq. (5), the value of λ_v at the mean temperature of the sample was used, and the water activity of the alginate gel was assumed to be 1.0 since the solid content of the gel was very low. As shown in Fig. 3, the values calculated from Eq. (1) were smaller than the experimental data. A much better approximation was obtained from Eq. (5). Thus, the prediction of effective thermal conductivity of air-impregnated gels was greatly improved by incorporating the equivalent thermal conductivity for latent heat transport into the Maxwell-Eucken model.

Kumazawa *et al.*⁵⁾ used the following equation to estimate the effective thermal conductivity of an aqueous maltodextrin solution containing gas bubbles.

Fig. 4 Temperature dependence of the effective thermal conductivity of air-impregnated alginate gels

$$1 - \phi = \frac{(\lambda_d + \lambda_v) - \lambda_e \left(\frac{\lambda_c}{\lambda_e} \right)^{1/3}}{(\lambda_d + \lambda_v) - \lambda_c \left(\frac{\lambda_e}{\lambda_c} \right)} \quad (6)$$

Equation (6) was derived by incorporating the equivalent thermal conductivity for latent heat transport into the Bruggeman equation. However, they did not experimentally confirm the applicability of Eq. (6). When evaluated at 40°C, Eq. (6) gave an effective thermal conductivity a little bit smaller than Eq. (5). Though the values calculated from Eqs. (5) and (6) differed by no more than 2 % from each other, Eq. (5) gave a better approximation to the experimental results shown in Fig. 3.

Figure 4 shows the temperature dependence of the effective thermal conductivity for air-impregnated alginate gels. When the gel contained no air bubbles, the effective thermal conductivity increased slightly with temperature, and was practically equal to the thermal conductivity of water, λ_{water} , at each temperature. When the gel was impregnated with air bubbles, the effective thermal conductivity increased with temperature more greatly than that of water or of the gel containing no air bubbles. According to Eqs. (1) and (5), the effective thermal conductivity was calculated for each volume fraction of gas phase. As shown in Fig. 4, Eq. (1) gave

Fig. 5 Temperature dependence of the effective thermal conductivity of air-impregnated ovalbumin gels

the effective thermal conductivity with a weak temperature dependence. The values calculated from Eq. (1) were far from the experimental data, especially at high temperature. On the other hand, Eq. (5) gave a fairly good approximation of the experimental data at 30–60°C when $\phi = 0.38$, indicating that Eq. (5) is valuable for predicting the temperature dependence of the effective thermal conductivity of porous gels. When $\phi = 0.59$, however, Eq. (5) gave a much larger value of the effective thermal conductivity at 60°C than the experimental data.

Figure 5 shows the temperature dependence of the effective thermal conductivity of ovalbumin gels impregnated with air bubbles. The effective thermal conductivity of the gel containing no air bubbles increased almost linearly with temperature from 20°C to 70°C. Similarly to the results shown in Fig. 4, the temperature dependence of the effective thermal conductivity of the air-impregnated gels was greater than that of the gel containing no air bubbles. Moreover, the effective thermal conductivity of the air-impregnated gel ($\phi = 0.40$) was larger than that of the gel containing no air bubbles at 70°C, suggesting a substantial contribution of latent heat transport at high temperature. Values of the effective thermal conductivity calculated from Eq. (5) are shown in Fig. 5. In the calculation, the water activity of the ovalbumin gel was assumed to be 1.0, since the water sorption data²⁾ suggested that the water activity of coagulated ovalbumin at 40°C was 0.95 when the water content was 0.243 g/g-dry mater, which was much smaller than that of the gel used in this study. As shown in Fig. 5, the effective thermal conductivity was well approximated by Eq. (5) in the temperature range of 30–70°C when $\phi = 0.40$. When $\phi = 0.67$, however, Eq. (5) overestimated the effective thermal conductivity at 70°C.

To discuss the applicability of Eq. (5) in further detail, the effective thermal conductivity of air-impregnated ovalbumin gels of various porosities was measured

Fig. 6 Effective thermal conductivity of air-impregnated ovalbumin gels as a function of the volume fraction of gas phase

at 50°C and 70°C. The results are shown in Fig. 6 in comparison with Eqs. (1) and (5). At 50°C, the experimental data were approximated by Eq. (5) with relative deviations less than 10 %. At 70°C, however, Eq. (5) overestimated the effective thermal conductivity of air-impregnated gels, which lay between the values calculated from Eqs. (1) and (5). Thus, the applicability of Eq. (5) for the effective thermal conductivity of air-impregnated gels was limited to temperatures up to 50°C.

Equation (5) overestimated the effective thermal conductivity at high temperature, especially for gels of high porosity. In the calculation of effective thermal conductivity with Eq. (5), the value of λ_v was evaluated at the mean temperature, though there was a temperature gradient across the gels. As shown in Fig. 2, the effect of temperature on the value of λ_v was greater at higher temperature. Thus, the higher the temperature the greater was the variation of the local thermal conductivity along the direction of heat flow, even though the temperature gradient applied to the gels was equal. This may be one of the reasons for the discrepancy between the experimental data and Eq. (5). Another reason may be the moisture distribution in the air-impregnated gels. If water migrates through the air-impregnated gel under a

temperature gradient as shown in Fig. 1, the water content, and hence the water activity, will decrease on the high-temperature side. Although the water content may increase on the low-temperature side, the water activity will be hardly affected since the water activity of the gel at the initial state is approximately 1. Thus, the water migration will result in a decrease in water activity on the high-temperature side. Since the value of λ_v decreases with decreasing water activity as shown in Fig. 2, there will be a decrease in the local thermal conductivity on the high-temperature side of the gel. For more accurate prediction of the effective thermal conductivity, the distribution of water activity in the air-impregnated gel should be considered.

Conclusions

The effective thermal conductivity of air-impregnated alginate or ovalbumin gels was measured at 30-70°C, and compared with the theoretical model proposed, which included latent heat transport accompanied by water vapor diffusion in addition to heat conduction. The effective thermal conductivity of the air-impregnated gels increased with temperature more greatly than that of gels containing no air bubbles. The experimental data were approximated by the model up to 50°C, though the model overestimated the effective thermal conductivity at higher temperature, especially for gels of high porosity. The discrepancy between the experimental data and the theoretical model at high temperature is possibly ascribable to the moisture distribution in gels. For more accurate prediction of the effective thermal

conductivity of porous foods, moisture distribution should be considered in the model.

Nomenclature

a_w	=	Water activity	[-]
D	=	Diffusivity of water vapor in air	[m ² ·s ⁻¹]
L	=	Latent heat of evaporation of water	[J·mol ⁻¹]
P	=	Total pressure	[Pa]
P_s	=	Saturated vapor pressure of water	[Pa]
P_v	=	Partial pressure of water vapor	[Pa]
R	=	Gas constant	[J·mol ⁻¹ ·K ⁻¹]
T	=	Absolute temperature	[K]
λ_c	=	Thermal conductivity of continuous phase	[W·m ⁻¹ ·K ⁻¹]
λ_d	=	Thermal conductivity of dispersed phase	[W·m ⁻¹ ·K ⁻¹]
λ_e	=	Effective thermal conductivity	[W·m ⁻¹ ·K ⁻¹]
λ_v	=	Equivalent thermal conductivity for latent heat transport	[W·m ⁻¹ ·K ⁻¹]
λ_{water}	=	Thermal conductivity of water	[W·m ⁻¹ ·K ⁻¹]
ϕ	=	Volume fraction of dispersed phase	[-]

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