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The Properties of Effective Diffusion Coefficient of Sodium Chloride in Agar Gel Cubes

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ABSTRACT

Cubes of 2% agar gel with side lengths (2L) of 1–10 cm were soaked in a 0.1 M sodium chloride solution at 25°C for 0–75 hours (t) in order to investigate three-dimensional diffusion. After soaking, the sodium chloride and water content of the gels were measured. From these values, the mean concentration ($\overline{C}(t)$) of the sodium chloride in the gel cube was calculated. After substituting the experimental value (L, t and $\overline{C}(t)$), except in the last stage of the transport, into the solution of the diffusion equation, the effective diffusion coefficient (D_e) value of 0.99×10^{-5} cm^2/s was obtained. As for the last stage of the transport, the experimental value significantly deviated from the value calculated using the effective diffusion coefficient. Therefore, another diffusion coefficient, called the apparent diffusion coefficient (D_{app}), was calculated by partially substituting the values of t, L and $\overline{C}(t)$ into the solution and it was observed that D_{app} decreased sharply.

On the other hand, the value of D_e obtained in a one-dimensional diffusion experiment was 1.12×10^{-5} cm^2/s, which was found to be more than that obtained in the three-dimensional one.

The same properties of D_e and D_{app} were also observed in the experiments of sodium chloride in various concentrations of corn starch, potato starch, egg albumin and soy protein gels or sucrose in 2% agar gels.

The properties of the three-dimensional diffusion found in this study indicates that the total amount of diffusion substances in foodstuffs using one-dimensional diffusion coefficients so far reported has been over estimated. Particularly in the case of restricted diets for sick persons or large scale plants in the food industry, the strict amount of total uptake in foodstuffs must be known, therefore, it is very important and useful to obtain three-dimensional diffusion coefficients.

INTRODUCTION

Seasoning plays an important part in cooking and food processing. Various phenomena, such as diffusion, osmosis, adhesion and adsorption, occur during the seasoning process between the food materials and the seasoning substances. Since diffusion occurs mainly among these phenomena, it becomes possible to control the seasoning process by knowing the diffusion phenomena. In order to describe these phenomena, it is necessary to know the effective diffusion coefficient (D_e) of the diffusing substances.

The effective diffusion coefficient of sodium chloride in various kinds of foodstuffs such as swordfish flesh (Del Valle and Nickerson, 1967) and cheese (Geurts et al., 1974) have been already reported. Favetto et al. (1981) studied the more complicated system of a sodium chloride–glycerol solution and measured the D_e of sodium chloride. In addition, the D_e of sodium chloride in 1–4% agar gels was reported by Fujii and Thomas (1958) and Allen et al. (1963). Slade et al. (1966) and recently Djelveh et al. (1989) also reported the D_e of sodium chloride in 3% agar gel. The diffusion of different various substances (*n*-alcohols, polyhydric alcohols, oligosaccharides and alkali metal chlorides) in cellulose gels was reported by Brown and Chitumbo (1973). Hendrickx et al. (1986) measured the D_e of glucose in carrageenin gels. In all these reports, the diffusion flow was designed to occur in one direction, that is a one-dimensional diffusion, and it was assumed that the D_e was constant during the experiments. On the other hand, in most cooking or food processing, diffusion substances penetrate materials from all directions from the surface to the center. Consequently, the D_e in the three-dimensional diffusion must be different from that obtained in the one-dimensional diffusion system.

TABLE 1

Conditions for soaking the cubes in a 0.1 M sodium chloride solution

Example	Size of cube (cm)	Number of cubes	Solution volume (l)	Soaking time (h)
1	1	10	4	0–6
2	2	10	4	0–6
3	3	6	4	0–24
4	5	6	40	0–24
5	7	2	40	0–48
6	10	2	80	0–75

As for the three-dimensional diffusion, the D_e of various salts in tuna flesh was measured by Sakai and Miki (1982) and Sakai and Suzuki (1985). In these studies, the D_e was calculated by assuming it was constant during the diffusion process.

This investigation was undertaken to reveal the properties of D_e when three-dimensional diffusion occurs in media. The D_e value for sodium chloride when it diffuses into 2% agar gel cubes from the surface to the center was studied using the diffusion equation for the mean concentration of sodium chloride in cubes.

MATERIALS AND METHODS

Materials

The agar was purchased from Kanto Chemical Co. Inc. The mercury (II) thiocyanate solution was prepared when 5 g of mercury (II) nitrate dissolved in 200 ml of 0.5 M nitric acid and 3 ml of saturated ammonium iron (III) sulfate were dissolved in 1 M nitric acid. As an aqueous potassium thiocyanate solution (4% w/v) was added drop-wise to the mixture, precipitate was produced until the supernatant turned slightly red. The obtained precipitate was then filtered through a glass filter (G3), washed with deionized water and dried *in vacuo*. A solution consisting of 0.3 g of the product dissolved in 95% ethanol is referred to as the mercury (II) thiocyanate solution. The ammonium iron (III) sulfate solution was prepared by dissolving 12 g of ammonium iron (III) sulfate in 200 ml of 6 M nitric acid. Deionized water was used throughout this study.

Preparation of the agar gel cubes

Agar powder was soaked in deionized water for 1 h, dissolved at 70°C for 30 min, then dissolved completely at 90°C for 30 min using a reflux condenser, degassed under reduced pressure and finally solidified in a stainless steel vat at 25°C for 12 h. The solid content of the gel was 2% (wt./wt.). Just before soaking the gel in a sodium chloride solution, the gel was cut into cubes with a knife, the side lengths of which were 1, 2, 3, 5, 7 and 10 cm. The surface of each cube was made as smooth as possible.

Preparation of agar gel as an infinite slab

Agar gel was similarly dissolved as gel cubes and solidified in a cylindrical stainless steel container (diameter, 10 cm; height, 10 cm). After leaving at 25°C for 12 h, the gel was used in a diffusion experiment.

Measurement of the mean concentration in gel cubes

The agar gel cubes were soaked in a 0.1 M sodium chloride solution at 25°C (±0.5°C) under the conditions shown in Table 1. Since the cubes were stood in the solutions on a network of suspended stainless steel or plastic, sodium chloride could penetrate the cubes from every surface to the center. The solution was stirred with a magnetic stirrer.

After soaking for a prescribed period, the gel cubes were taken out of the sodium chloride solution, the solution on the surfaces was removed with filter paper and weighed. The water content was measured by drying the gel cubes at 70°C *in vacuo*. With the other half, the total amount of sodium chloride, which had penetrated the gel cubes, was measured in the following way. Each gel cube was homogenized at 18,000 rpm for 2 min with a DX-8 homogenizer (Nihonseiki Kaisha Ltd.), then the homogenate was filled to an appropriate volume with deionized water and centrifuged at 5,000 × g for 10 min. To the supernatant (2.5 ml) the following reagents were successively added: mercury (II) thiocyanate (1 ml), iron (III) ammonium sulfate (2 ml) and deionized water (7 ml). After the mixed solution was allowed to stand for 10 min, the absorbency of the solution was determined at 460 nm. Since the concentration of Cl^- could be determined in this measurement, the concentration of sodium chloride was then obtained from the ratio of NaCl/Cl = 58.44/35.45. The mean concentration of sodium chloride in the cube was then calculated from the following equation:

$$\overline{C}(t) = M(t)/\{W \times (P/100)/d\} \qquad (1)$$

$\overline{C}(t)$ (mol/ml) is the mean concentration of sodium chloride in the cube at time t, M(t) (mol) is the total amount of sodium chloride which had penetrated the cube at time t, W (g) is the weight of the cube, P (%) is the percentage of water content in the cube, and d (g/ml) is the density of water. The soaking experiment was done in triplicated.

The cation exchanger properties in agar mentioned by Fujii and Thomas (1958) will influence the transport of Na^+. According to our preliminary investigation, however, there was only a negligible difference between the concentration of Na^+ measured by atomic absorbency flame photometry (Shimadzu AA-660) and Cl^- measured as already described after soaking the agar gel in the sodium chloride solution.

Measurement of the concentration gradient in a semi-infinite gel slab

The agar gel slab in the stainless steel container was soaked horizontally in 5 l of 0.1 M sodium chloride solution at 25°C for 1, 3, 6, 9 or 12 h with the solution being stirred with a magnetic stirrer. After soaking, the gel was removed from the stainless steel container and a small cylindrical section of 2.83 cm diameter was cut out from the center of the gel (Fig. 1a). In this section, the diffusion occurred one-dimensionally without the influence of diffusion flow from the side of the gel. The cut-out cylindrical gel sample was sliced into 6 disks of 0.5 cm thickness, in the direction away from the surface in contact with the sodium chloride solution (Fig. 1b). Each disk was cut in half, one half being measured for water content and the other half for the mean concentration of sodium chloride in the same manner as that used for the cubes.

Solution of the diffusion equation for cubical media

A solution of the diffusion equation concerning the mean concentration in a cube has been expressed as the product of three one-variable problems by Bird et al. (1960) as follows:

boundary conditions:
$C(x,y,z,t) = C_1,\ x = \pm L,\ y = \pm L,\ z = \pm L,\ t>0$ (2)

initial conditions:
$C(x,y,z,t) = C_o, -L<x<L,\ -L<y<L,\ -L<z<L,\ t=0$ (3)

$$\frac{C_1-\overline{C}(t)}{C_1-C_o} = 8\sum_{m=0}^{\infty}\sum_{n=0}^{\infty}\sum_{p=0}^{\infty}\frac{1}{(m+1/2)^2(n+1/2)^2(p+1/2)^2\pi^6}$$
$$\exp\left[-\left\{(m+1/2)^2+(n+1/2)^2+(p+1/2)^2\right\}\pi^2Dt/L^2\right] \quad (4)$$

where $\overline{C}(t)$ (mol/ml) is the mean concentration in the cube at time t (sec) described in Equation (1) and L is the half side length of the cubes (0.5–5 cm) employed in the experiment. D is the diffusion coefficient and C_o and C_1 are the initial and boundary concentrations, respectively. The initial condition C_o was zero and the boundary condition C_1 was 1×10^{-4} mol/ml in this study.

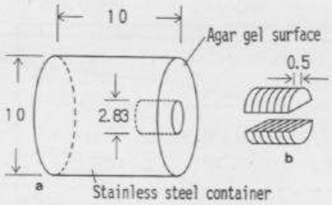

Fig. 1. Schematic diagram for the measurement of concentration gradient in a semi-infinite gel slab. The unit in the figure is cm. a: Stainless steel container in which the agar gel slab was contained. b: Cut-out cylindrical sample which was used for measuring the concentration gradient of sodium chloride.

Solution of the diffusion equation for a semi-infinite slab

The solution of the diffusion equation for a semi-infinite slab under

boundary conditions: $C = C_1 \quad x = 0,\ t > 0$ (5)

and initial conditions: $C = C_o \quad x > 0,\ t = 0$ (6)

was given by J. Crank (1975) as the error function:

$$\{C_1 - C(x,t)\}/(C_1 - C_o) = \mathrm{erf}\{x/2(Dt)^{(1/2)}\} \quad (7)$$

where C(x,t) (mol/ml) is the concentration of the diffusion substance at distance x (cm) and time t (s). The error function is usually written as erf (z), where

$$\mathrm{erf}(z) = \{2/\pi^{(1/2)}\}\int_0^z \exp(-w^2)dw \quad (8)$$

For an approximate solution of Equation (7), C(x,t) was assumed to be the mean concentration of sodium chloride at the center of the sliced disks (Fig. 1b); i.e., 0.25, 0.75, 1.25, 1.75, 2.25 and 2.75 cm from the surface.

RESULTS

Diffusion coefficient in semi-infinite media

The values for C_r in a semi-infinite agar gel slab are shown in Fig. 2 by symbolic marks. The value for diffusion coefficient D obtained by best-fitting Equation (7) to the experimental value was 1.12×10^{-5} cm^2/s (D_{e1}). The solid curves in Fig. 2 were drawn by using the obtained D_{e1} value. This D_{e1} value is between the value 0.97×10^{-5} cm^2/s and 1.27×10^{-5} cm^2/s, reported formerly by Fujii et al. (1958) and

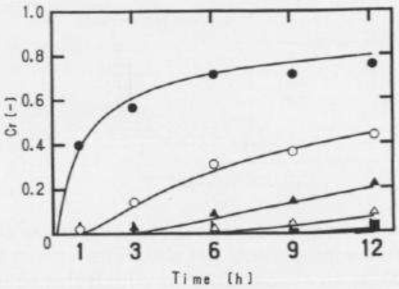

Fig. 2. Changes in C_r for a semi-infinite agar slab after soaking in a 0.1 M sodium chloride solution. Experimental values are shown by: filled circles, 0.25 cm; hollow circles, 0.75 cm; filled triangles, 1.25 cm; hollow triangles, 1.75 cm; filled squares, 2.25 cm; hollow squares, 2.75 cm, which represent the distance from the surface in contact with the sodium chloride solution to the center of each sliced sample. The solid curves represent the calculated value from Equation (7) using D = 1.12×10^{-5} cm^2/s.

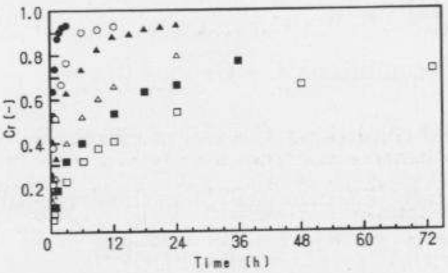

Fig. 3. Changes in the C_r of various agar gel cubes after soaking in a 0.1 M sodium chloride solution. Filled circles, 1 cm; hollow circles, 2 cm; filled triangles, 3 cm; hollow triangles, 5 cm; filled squares, 7 cm; hollow squares, 10 cm in side length of the cubes.

Slade et al. (1966), respectively. The difference between these values is caused by the difference in agar used.

Diffusion coefficient in agar gel cubes

Change of the mean concentration in the gel cubes

The change of C_r for the various agar gel cubes after soaking in a 0.1 M sodium chloride solution are shown in Fig. 3. C_r represents the ratio of the mean concentration of sodium chloride in the gel cubes to its boundary concentration; i.e., $C_r = \overline{C}(t)/C_1$. The smaller cube took a shorter time to attain equilibrium; i.e., $C_r = 1$.

Relationship between C_r and t/L^2

The relationship between C_r and t/L^2 is shown in Fig. 4. It can be recognized that the value of $1 - C_r$ is a function of the value of t/L^2 regardless of the size of the cube.

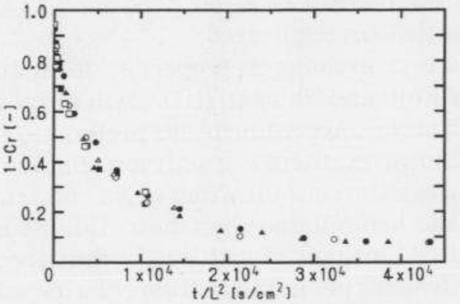

Fig. 4. Relationship between $1-C_r$ and t/L^2. Symbols are the same as those in Fig. 3. The value for $1-C_r$ is a function of the value for t/L^2 regardless of the size of the cube.

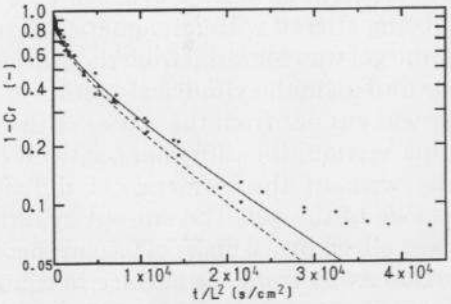

Fig. 5. Comparison between the calculated and the experimental value of $1-C_r$ and t/L^2 using one and three-dimensional diffusion coefficient. Dots represent experiment values (data number is 60). Solid line represents the calculated value employing three-dimensional diffusion coefficient 0.99×10^{-5} cm^2/s, which was obtained by best correlation to the experimental 52 data. Dashed line represents the calculated value employing one-dimensional diffusion coefficient 1.12×10^{-5} cm^2/s which was obtained in semi-infinite slab.

Three-dimensional diffusion coefficient

The total number of individual experimental data points were 60. The dashed line in Fig. 5 represents the calculated value when the one-dimensional diffusion coefficient, 1.12×10^{-5} cm^2/s (D_{e1}) obtained in semi-infinite agar gel slab, was used in Equation (4). The dashed line gave lower values for $1-C_r$ than the experimental ones. The solid line represents the case using the three-dimensional diffusion coefficient, 0.99×10^{-5} cm^2/s (D_{e3}) which was obtained by the best correlation of 52 data points to the solution of the diffusion equation except in the final stage of transport where it obviously deviated from the calculated value (8 data points in the range $1-C_r$ is less than 0.1).

Statistical study of the difference between one- and three- dimensional diffusion coefficients

The following values were introduced.

$$X_1 = (C_{r1Cal} - C_{rExp})/C_{r1Cal} \qquad (9)$$

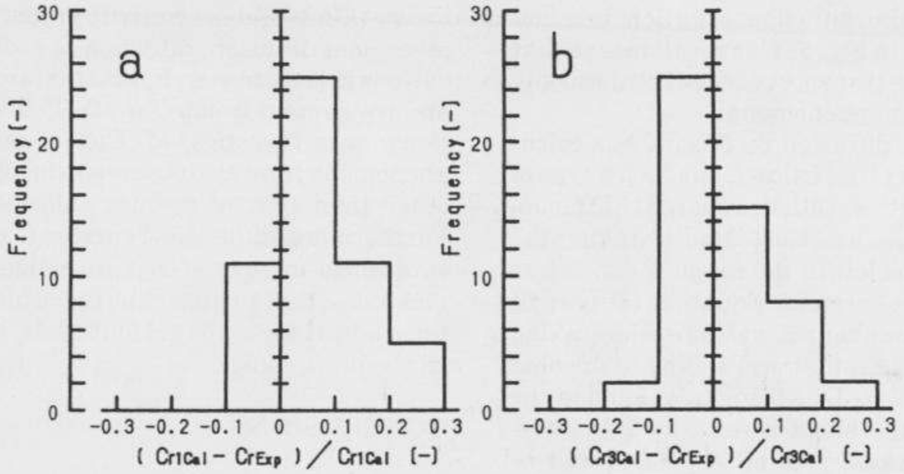

Fig. 6. Distribution histogram for the balance of experimental and calculated C_r ((experimental C_r – calculated C_r)/experimental C_r). a: The case using one-dimensional diffusion coefficient. b: The case using three-dimensional diffusion coefficient.

TABLE 2

The mean and the variance of distributions

Diffusion coefficient	Distribution	
	a One dimensional ($D_{e1} = 1.12\times10^{-5}$ cm^2/s)	b Three-dimensional ($D_{e3} = 0.99\times10^{-5}$ cm^2/s)
Mean ($\overline{X}_i$)	6.65×10^{-2}	2.22×10^{-2}
Variance (s_i^2)	7.58×10^{-3}	8.14×10^{-3}

$$X_3 = (C_{r3Cal} - C_{rExp})/C_{r3Cal} \tag{10}$$

where C_{r1Cal} and C_{r3Cal} were C_r values calculated with D_{e1} and D_{e3}, respectively and C_{eExp} was the experimental value. Since the value D_{e3} was obtained by the best correlation of experimental values, the histograms of the distribution for X_3 is thought to be a normalized one and the mean value for X_3 ($\overline{X}_3$) must become very small. Both histograms of X_1 and X_3 are shown in Figs. 6a and b, respectively. The number of samples was 52. The mean ($\overline{X}_i$) and the variance (s_i^2) of each distribution is shown in Table 2. Since these distributions correspond to the samples extracted from each population, the means (μ_i) of the populations are able to be presumed using the Student *t* test from these distributions.

When the hypothesis, $\mu_1 = \mu_3$, is not able to be rejected, it means that the difference between X_1 and X_3 is not significant. This result also leads to the fact that the difference between C_{r1Cal} and C_{r3Cal} is not significant and consequently, there is no significant difference between the values D_{e1} and D_{e3}. In the case of rejection, we will be able to derive the significant difference between D_{e1} and D_{e3}.

Since the F-value (s_1^2/s_3^2) was smaller than the value of $F_{51,51}$ as shown in Equation (11),

$$F = 1.07 < F_{51,51} \fallingdotseq 2.14 \quad (P<0.01) \tag{11}$$

the hypothesis that $\sigma_1^2 = \sigma_3^2$ was acceptable. Consequently, the Student *t* test is able to be applied to these distribution data. The result of the test for a T-value was the following.

$$|T| = 2.551 > t_{102} \fallingdotseq 1.980 \quad (P<0.05) \tag{12}$$

From this result, the hypothesis that $\mu_1 = \mu_3$ was rejected. The result of the Student *t* test indicated that there exists a significant difference between the one-and three-dimensional diffusion coefficients. Consequently, the fact that the three-dimensional diffusion coefficient is less than the one-dimensional one was not caused by the experimental error.

Apparent diffusion coefficient in the last stage of the transport on three-dimensional diffusion system

In the final stage of the diffusion, the experimental values deviated from the calculated one and the approximation by the diffusion equation was not successful as shown in Fig. 5. This result means that in the final stage, the transport does not conform any longer to the diffusion phenomena.

As for this stage, diffusion coefficient was calculated from Equation (4) as follows, but such a type of diffusion coefficient is called apparent diffusion coefficient (D_{app}). D_{app} was calculated by taking the first 4 data, from the left in the range $1 - C_r$ is less than 0.1, as one set to make Equation (4) best fit these 4 data, then another set was formed by taking the last 3 data of the first set and adding to the next datum moving right, its D_{app} being calculated in the same way, and so on. Thus, the value for D_{app} corresponding to these small sets of t/L^2 or C_r could be approximated. When the number of data in a set was smaller, the value for D_{app} was so widely scattered that the dependency of D_{app} on t/L^2 or C_r could not be recognized. The calculated D_{app} versus C_r is shown by circles in Fig. 7. The value for C_r in Fig. 7 represents the mean value of the 4 data in each set. From the result of the apparent diffusion coefficient, transport in the final stage was considered to be small.

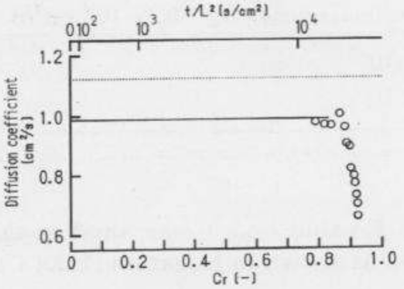

Fig. 7. Change of D_{app} for three-dimensional diffusion. Circles represent the D_{app} value in the final stage of the diffusion. Solid line represents the three-dimensional diffusion coefficient, 0.99×10^{-5} cm²/s. Dashed line represents the one-dimensional diffusion coefficient 1.12×10^{-5} cm²/s.

Characteristic properties in the three-dimensional diffusion system

In order to reveal the actual transport profile of seasonings or additives in foods, the transport of sodium chloride in 2% agar gel cubes was investigated. From this investigation, two characteristic properties in a three-dimensional diffusion system was confirmed. These are: (1) the diffusion coefficient obtained three-dimensionally is less than that obtained one-dimensionally and indicates that a substance moves more slowly in the three-dimensional diffusion than in the one-dimensional diffusion; (2) the apparent diffusion coefficient in the final stage decreases sharply and it means that there is minimal transport in the final stage. In order to confirm the two kinds of characteristic properties in three-dimensional diffusion, diffusion of sodium chloride in various gels (corn starch, potato starch, egg albumin and soy protein isolate for 10–25.5% gel concentrations) was investigated. Consequently, the same phenomena have also observed among these samples other than that of sodium chloride in agar gels. Furthermore, diffusion of sucrose (as non electrolyte substance) in agar gels is investigated in order to make sure that sodium chloride, which has a character of adsorbing to the gel materials, has no influence on the properties.

CONCLUSIONS

According to this study, the actual transport profile of seasoning or additives in foodstuffs was revealed and the properties of three-dimensional diffusion found in this study indicates that the total amount of diffusion substances in foodstuffs using one-dimensional diffusion coefficients so far reported was overestimated. In other words, in the case of cooking for the agar gel cube with a side length of 5 cm, 8.6 h must be added to the predicted time with one-dimensional diffusion coefficient. As for not only the simple gel samples, used in this investigation, but also various food materials, three-dimensional diffusion coefficient must be determined. It is important and useful to know the three-dimensional diffusion coefficient and to realize the actual total uptake into foodstuffs especially in the case of restricted diets for the sick and the large-scale plants of the food industry.

ACKNOWLEDGMENT

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NOMENCLATURE

C_r	ratio of the mean concentration of sodium chloride in the gel cube to the boundary concentration ($= \overline{C}(t)/C_1$) [–]
C_{r1Cal}	calculated C_r value when one-dimensional diffusion coefficient (D_1) is used [–]
C_{r3Cal}	calculated C_r value when three-dimensional diffusion coefficient (D_3) is used [–]
C_{rExp}	experimental C_r value [–]
$\overline{C}(t)$	mean concentration [mol/ml]
C_1	boundary concentration [mol/ml]
C_o	initial concentration [mol/ml]
D_{app}	apparent diffusion coefficient [cm²/s]
D_e	effective diffusion coefficient [cm²/s]

D_{e1}	one-dimensional diffusion coefficient obtained in semi-infinite agar gel slab [cm^2/s]
D_{e3}	three-dimensional diffusion coefficient obtained by best correlation [cm^2/s]
d	density of water [g/ml]
L	half side length of a cube [cm]
$M(t)$	total amount of solute that has penetrated into the gel cube at time t [mol]
m	number of summations
n	number of summations
P	percentage of water content [%]
p	number of summations
s	deviation [–]
s_1^2	variance of distribution-a [–]
s_3^2	variance of distribution-b [–]
t	time [s]
W	weight [g]
x	distance [cm]
X_1	difference between the calculated and experimental C_r values when one-dimensional diffusion coefficient is used $(=(C_{r1Cal} - C_{rExp})/C_{r3Cal})$ [–]
$\bar{x}_1$	mean of distribution for Fig. 6a [–]
X_3	difference between the calculated and experimental C_r values when three-dimensional diffusion coefficient is used $(=(C_{r3Cal} - C_{rExp})/C_{r1Cal})$ [–]
$\bar{x}_3$	mean of distribution for Fig. 6b [–]
μ_1	mean of the population for sample distribution-a [–]
μ_3	mean of the population for sample distribution-b [–]
σ_1^2	variance of the population for sample distribution-a [–]
σ_3^2	variance of the population for sample distribution-b [–]

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