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Determination of Effective Diffusion Coefficients in Calcium Alginate Gel Plates with Varying Yeast Cell Content

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ABSTRACT

Effective diffusion coefficients (D_e) have been determined for lactose, glucose, galactose, and ethanol in calcium alginate gel with varying yeast cell concentration. The measurements have been performed in a diffusion cell, and the results evaluated with the quasi-steady-state method.

An ultrasonic meter was used for gel thickness determination with an accuracy of 1.5% and a new method for the reproducible preparation of gel plates was developed.

It was found that D_e in pure alginate gel decreased to about 90% of the diffusivity in water and did not vary with alginate concentration. D_e decreased considerably with increasing yeast cell concentration. For the solutes studied, the effective diffusion coefficient can be estimated according to the equation $D_e = D_{eo} \cdot (1 - \phi)/[1 + (\phi/2)]$, where D_{eo} is the effective diffusivity in pure gel and ϕ is the volume fraction of yeast cells.

Index Entries: Gel; alginate; baker's yeast; diffusion; glucose; lactose; galactose; ethanol.

INTRODUCTION

The design of bioreactors with immobilized cells requires an understanding of the mass transfer characteristics (*1,2*). The external mass transfer influences the reaction velocity to a certain extent but this is often negligible compared with the internal mass transfer (*3–5*). A frequently used support material is calcium alginate with the cells immobi-

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lized in small gel beads. Therefore, a knowledge of the diffusion characteristics of the gel is essential, especially with a high cell content in the calcium alginate carrier. In a recent study it was shown that too high a cell content could lower the product yield (*6*). The model systems used were anaerobic fermentation of glucose to ethanol with baker's yeast immobilized in calcium alginate gel and anaerobic fermentation of lactose and whey to ethanol with coimmobilized yeast and β-galactosidase.

The purpose of this study was therefore to determine effective diffusion coefficients (D_e) for lactose, glucose, galactose, and ethanol in calcium alginate gel. The results were used to construct mathematical models for the prediction of reactor performance and to facilitate reactor choice (*7*).

Much work has been done to determine D_e for gels and polymers focusing on the dependence of the polymer content (*8*). In some studies carried out during recent years, diffusion coefficients (D) and effective diffusion coefficients (D_e) for gels containing cells have been reported (*5,9,10*). Unfortunately, there is considerable confusion in the literature whether D_e or D is determined. These differ by a factor k, which is defined as the partition coefficient for the solute between the gel and the surrounding solution.

For low alginate content in gel without cells it is often stated that the diffusion is only slightly (*11*) or not influenced at all (*5,12*) by the alginate matrix. This is a result of the low alginate content and the macroporous structure with pore diameters on the order of 10 μm (*11*).

When cells are introduced into the alginate matrix, D_e is reduced because of the exclusion effect of the cells (*5*). Hannoun et al. (*9*) recently reported that the diffusion coefficient (D) was unaffected by the cell content. If the partition coefficients had been determined, the diffusion coefficients could have been recalculated to give a decreasing D_e with increasing cell content. This would be consistent with the results reported by Furusaki et al. (*5*), where D_e decreased with increasing cell concentration in alginate gel.

Two of the most common methods of determining D and D_e are the nonsteady-state method (*12*) and the diaphragm diffusion cell method (*9,10,13*). With the nonsteady-state method the diffusion rate is measured while the solute diffuses into or out of gel beads in a well-stirred solution. In this way, D_e can be determined if the beads are assumed to be uniform and equal in size.

The diffusion cell method is reliable if gel plates can be made reproducibly with a uniform thickness, and if the thickness of the plates can be accurately measured. The results can be evaluated either according to the time-lag method, giving D, or the quasi-steady-state method, giving D_e.

In the present study, a diffusion cell was used to study the behavior of four solutes with various gel plates. The experiments have been evaluated with the quasi-steady-state method.

THEORY

Diffusion in Gels

The diffusion in the gel is assumed to be Fickian in nature according to Eq. (1):

$$\frac{\delta c^*}{\delta t} = D\frac{\delta^2 c^*}{\delta x^2} \tag{1}$$

where c*represents the concentration of the solute in the gel.

At steady state the molar flow, N mol/s, through the gel plate (Fig. 1) can be expressed as

$$N = -DA\frac{c_A^* - c_B^*}{l} \tag{2}$$

Since these concentrations cannot be easily measured, Eq. (2) is reformulated as

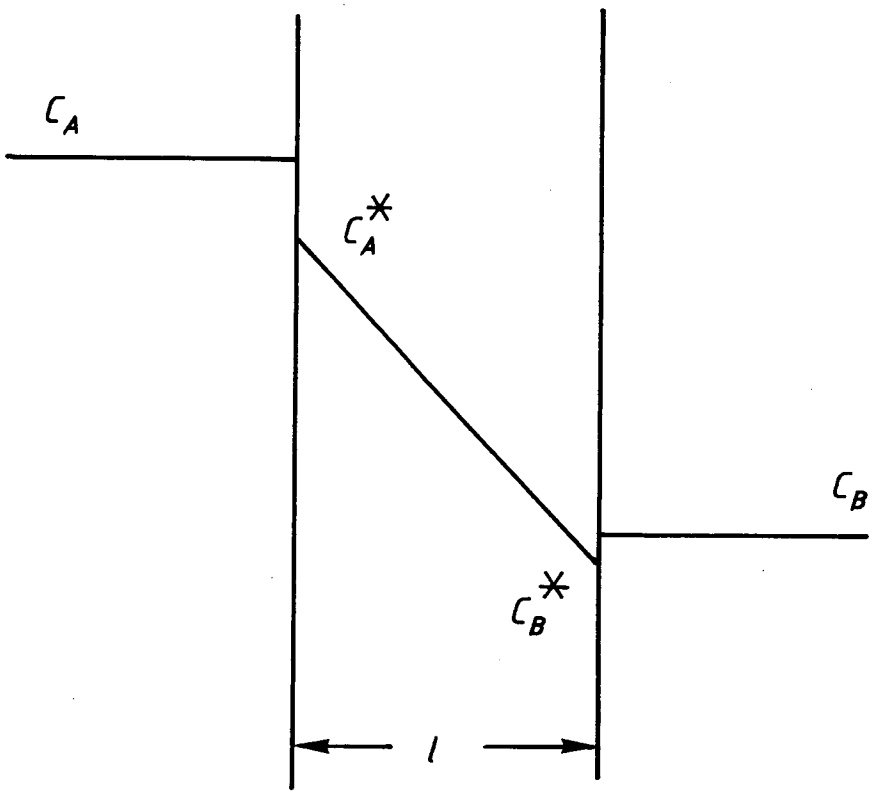


Fig. 1. Concentration profile across gel plate.

$$N = -D_e A \frac{c_A - c_B}{l} \tag{3}$$

where c_A and c_B represents the liquid concentrations. The partition coefficient is defined as $k = c_A^*/c_A = c_B^*/c_B$ and the effective diffusion coefficient $D_e = kD$.

The concentration profile can be seen in Fig. 1. The cells and the calcium alginate matrix exclude a fraction ϕ of the total gel volume for the diffusing solute. Because of this exclusion effect the fractional void volume will be $1 - \phi$.

A common approach is to predict k from the geometric exclusion effect, i.e., $k = 1 - \phi$ (*8*) if there are no additional adsorption effects. The relationship between D and D_e in this case will be

$$D_e = D(1 - \phi) \tag{4}$$

Inserting Eq. (4) into Eq. (3) results in Eq. (5):

$$N = -A(1 - \phi)D \frac{c_A - c_B}{l} \tag{5}$$

This equation illustrates four equivalent approaches to the exclusion effect.

1. Diffusion through an effective (reduced) area $A(1 - \phi)$.
2. Diffusion with an effective (reduced) concentration difference $c_A(1 - \phi) - c_B(1 - \phi) = c_A^* - c_B^*$
3. Diffusion with an effective (lengthened) diffusion path $l/(1 - \phi)$. It should be noted that the diffusion path is considered to be straight with no buoyancy at all.
4. Diffusion with an effective (reduced) diffusion coefficient. $D_e = D(1 - \phi)$

The last approach is most widely used.

Besides this exclusion effect the effective diffusion coefficient (D_e) will be further decreased by the obstruction effect. This means that there will be an increased diffusion path length caused by buoyancy around the obstructions creating a tortuous pathway. This is sometimes referred to as the tortuosity, which is the subject of many correlations in traditional heterogeneous catalysis.

The random-pore model (*14*) gives

$$\frac{D_e}{D_{aq}} = (1 - \phi)^2 \tag{6}$$

The cell model (*15*) or Maxwell's expression for spheres (*8*) gives

$$\frac{D_e}{D_{aq}} = \frac{(1 - \phi)}{[1 + (\phi/2)]} \tag{7}$$

In Eq. (7) the factor $1 - \phi$ emanates from the exclusion effect and $1 + (\phi/2)$ from the obstruction effect.

The Diffusion Cell—Steady-State Method

After an initial period of time (t_o) a quasi-steady state is attained resulting in a linear concentration gradient in the gel plate. If the volumes V_A and V_B of the two compartments, separated by the gel plate, are constant, with the concentrations c_A and c_B, respectively, the following equation (8) is obtained.

$$\ln \frac{c_A - c_B}{c_{Ao} - c_{Bo}} = - \frac{D_e}{l} A \left(\frac{1}{V_A} + \frac{1}{V_B} \right)(t - t_o) \tag{8}$$

MATERIALS AND METHODS

Materials

Sodium alginate was of the type Manugel SNF. All other chemicals were of reagent grade and purchased from local commercial sources. Deionized water was used.

Medium

For the preparation of gels, a solution of 0.1*M* $CaCl_2$ in acetate buffer (pH 4.9) was used. For the curing and storing of gels, a solution of 0.01*M* $CaCl_2$ in acetate buffer (pH 4.5) was used. This solution was also used as solvent with 20 mg/L merthiolate added (to prevent microbial activity).

Microorganisms

Fresh baker's yeast (Svenska Jästbolaget, Sweden) of commercial grade was deactivated. A suspension of 35 wt% yeast (wet basis) and 0.35 wt% iodoacetic acid was stored at 8°C overnight. The suspension was then vacuum-filtered, but not washed. The filter-cake, which was used for preparation of gels, had the same dry content (26 ± 0.5 wt%, 12 h at 95°C) and the same density (1.10 g/cm^3) as the original yeast.

No lysis of the cells could be seen in microscope studies. No fermentative activity could be detected with the yeast cells in a glucose solution.

Preparation of Gel Plates

A sodium alginate solution of 4 wt% was made, and mixed with a yeast–water suspension, to give the desired composition. When cell-free gels were made, the desired sodium alginate solution was made directly. The compositions of solutions and gels are given in wt% alginate (dry basis) and wt% yeast (wet basis).

A known weight of the solution was poured into a horizontal mold (diameter 120 mm, depth 8 mm), to a height of 3–6 mm, Fig. 2a. A metal ring (diameter 90 mm) with nylon threads (fishing line, 0.20 mm) stretched across it in a crisscross pattern, was placed in the mold. The arrangement is illustrated in Figs. 2a,2b. The purpose of the nylon threads was to strengthen the gel plate, as it otherwise bulged inside the diffusion cell, affecting the true area and thickness. The area and volume of the nylon threads were negligible, and the diffusion flow was presumed to be unaffected.

An aerosol of the 0.1*M* $CaCl_2$ solution was sprayed intermittently over the alginate surface. This process was continued for a period of 4–6 h, after which the $CaCl_2$ solution could be carefully poured into the mold to a depth of 2–3 mm. After an additional 3–6 h, the plate was transferred to a bath of $CaCl_2$ solution, where it remained for 2 h. It was then cured for 24–36 h, before weighing and experiments were performed.

The idea behind this method was to ensure the slow penetration of Ca^{2+} ions in a vertical direction only. In this way, the inevitable contraction would also be in a vertical direction only and a smooth and uniform gel was formed. The disadvantage was that the gel (at least at one side of the plate) remained in contact with the strong $CaCl_2$ solution for several hours, which is not the case when ordinary beads are produced.

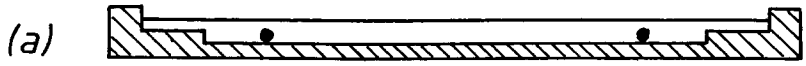
The resulting gel plates had thicknesses varying from 2.5–5.0 mm.

Gels were made specifically for the determination of shrinkage and partition coefficients. The demand for smoothness and horizontal alignment was less stringent, and no ring was used. Otherwise, the methods were the same.

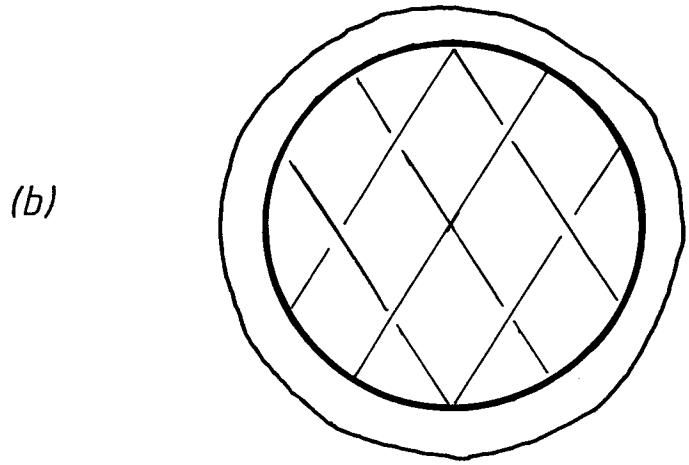
Determination of Gel Plate Thickness

An ultrasonic gage (Panametrics, MA, USA) with a piezoelectric probe was used to determine the plate thickness. With such a device, the thickness can be obtained if the velocity of sound in the medium is known. The velocity of sound in the gel medium was determined by using a large number of pieces of gel of well-known thickness (20–30 mm), and adjusting the velocity until the apparatus showed the correct thickness. The mean sound velocity in the gel was found to be 0.1545 cm/μs ($s < 1.5\%$), close to that of seawater, and was independent of yeast cell and alginate content.

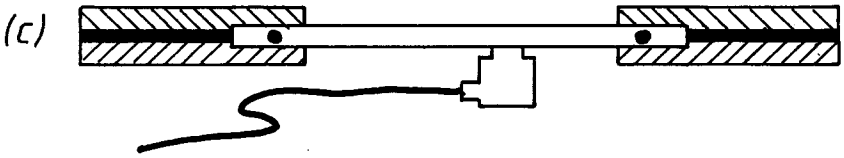
The thickness of the pieces of gel used for calibrating the ultrasonic gage were measured with a micrometer. This is a time-consuming and



Mold, with solution and ring



Gel plate, with ring



Gel plate in holder, and ultrasonic probe

Fig. 2. (a) Mold with solution and ring, (b) gel plate with ring, (c) gel plate in holder and ultrasonic probe.

laborious method. If care is not taken, the micrometer tip may compress the gel. Once the sound velocity in the gel had been determined, the convenient and precise ultrasonic method could be used. The display showed the thicknesses directly, with a resolution of 0.01 mm.

The wet gel plate was first fitted into the holder, see Fig. 2c. One side was dried with soft paper tissue, while the other was kept wet by a thin liquid layer (acting as a "coupling liquid" between gel and probe).

The thickness of the gel plate was calculated as the mean value of 26 measurements, half of these from each side at 13 different points over the whole area. The values for the same point, measured from different sides, never differed by more than 1.5%. After the diffusion experiment the thickness was measured once more and the mean value was calculated. The variation of gel plate thickness was 6–7% around the mean value.

Diffusion Cell

The plexiglass diffusion cell is shown in Fig. 3. The inner diameter of each chamber was 100 mm and the length was 52 mm. The open area, through which diffusion occurred, had a diameter of 70 mm. Rubber seals and plugs are shown in black. The cell was kept at 30 ± 0.5°C, by a thermostatically controlled jacket.

The pretempered solutions were poured simultaneously into the chambers (left chamber A, the solutes; right chamber B, solvent only). Care was taken to ensure that the final levels were equal, and the exact volumes (515–550 mL) were noted. Agitation (800 rpm) was started, and the zero-time was noted.

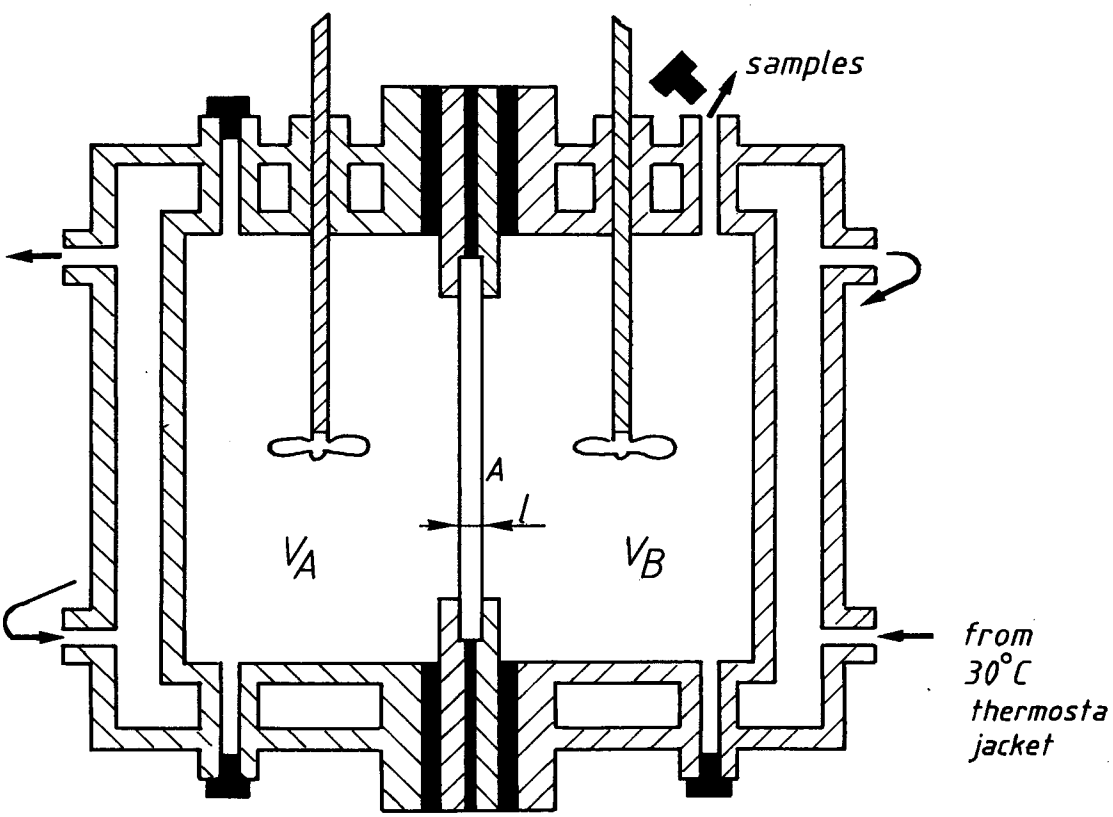


Fig. 3. Diffusion cell

An experiment lasted for 10–12 h, and 11 samples were collected with a hypodermic needle from chamber B. The samples were immediately filtered (0.45 μm) into an HPLC sample bottle. The sampling intervals were 30–100 min. The volume of every sample (1.5 mL) taken from chamber B was replaced by the same volume of pure solvent. When the experiment was finished samples were also taken from chamber A, in order to examine the mass balance.

Shrinkage

The shrinkage was simply calculated from the initial weight of the alginate-solution and the final weight of the alginate gel, since the densities of alginate solution and alginate gel were about the same.

Partition Coefficients

The gels used for the determination of shrinkage were cut into 6 mm cubes. 20 g of the gel were then placed in a graduated glass containing 30 mL of solution, which was placed on a tared balance, so that both weight and volume of gel and solution was known. Gel and solution were then poured into a beaker that was sealed with a rubber plug and stored at 8°C for 4 d and shaken twice a day. After an additional 4 h at 18–22°C, two samples of the solution were collected with a hypodermic needle, diluted, and filtered into a HPLC sample bottle. The final weight and volume of the gel were determined.

Analytical Methods

Lactose, glucose, galactose, and ethanol were analyzed using high performance liquid chromatography. A Shimadzu chromatograph was used, with a column of 7.8 mm inner diameter and length of 300 mm (Biorad Aminex HPX-87-H) placed in an oven at 60°C. Diluted H_2SO_4 (5 m*M*) was used as the diluent, at a flow rate of 0.5 mL/min. A refractive index detector (ERMA ERC-7510) was used.

If necessary, samples were diluted to the 0–3 g/L range, which gave the best accuracy. Very good linearity of the standard series, between 0.015 g/L (0.030 g/L for ethanol) and 3 g/L was obtained.

EXPERIMENTAL

A comprehensive study has been carried out to apply the classical diffusion cell to diffusion measurements of gel plates (*16*). This work includes the preparation of reproducible gel plates, the measurement of gel plate thickness, and the choice of a sufficiently high mixing speed to ensure that there is no external mass transfer hindrance outside the gel plate. The choice of adequate diameter and thickness of the gel plate have been subject to experimental investigations as well as mathematical

modeling. These calculations show that a gel plate, with a small diameter (≤50 mm) and, at the same time, a large thickness (≥ 5 mm), results in experimental errors because of the additional diffusion flux through the edges of the gel plate, which are clamped into the gel plate holder. This work will be reported elsewhere (*17*).

Effective diffusion coefficients were measured for gel plates with different alginate and yeast cell concentrations. As the gel shrank during preparation and curing, and to some extent during the experiment, the shrinkage as a function of alginate- and yeast cell concentration was determined.

Finally, the partition coefficients for the solutes between the gel and solution were determined as these are required in the diffusion models simulating the mass transfer behavior.

RESULTS AND DISCUSSION

Effective Diffusion Coefficients (D_e)

The effective diffusion coefficients D_e, were calculated from Eq. (8), represented graphically in Fig. 4. To ensure that the quasi-steady-state condition was fulfilled the first points were excluded. In the experiment shown in Fig. 4, quasi-steady-state was obtained at t_o = 170 min.

D_e for glucose (initial concentration 100 g/L) was determined for 11 gel plates without yeast cells and an alginate concentration of 3 wt% in the gel. The value obtained, $D_e = 6.23 \times 10^{-10}$ m^2/s (s = 7%), was 87% of the value for glucose diffusivity in water, $D_{aq} = 7.0 \cdot 10^{-10}$ m^2/s (*18*). This indicates that the effective diffusivity is only slightly lowered by the

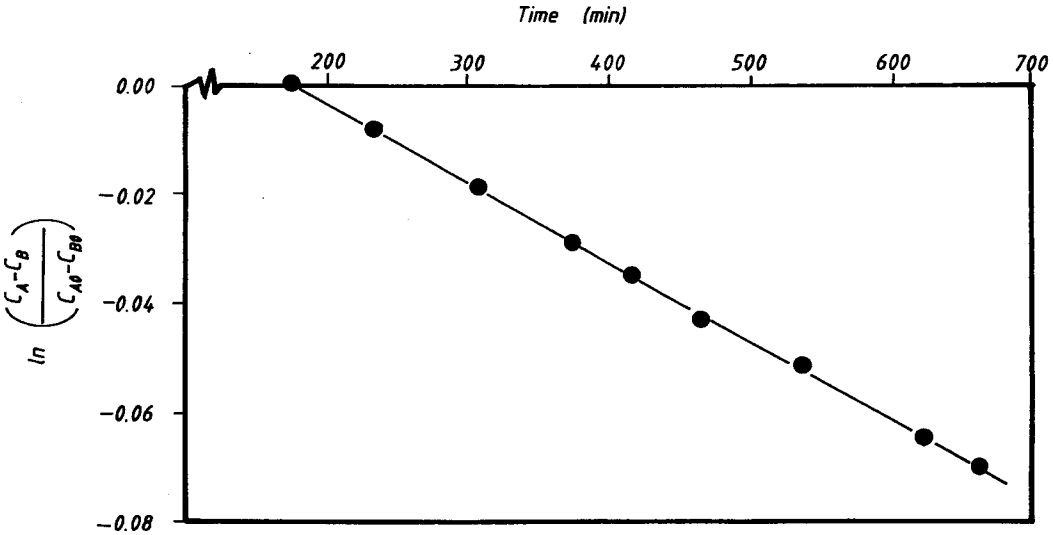


Fig. 4. Evaluation of D_e from a quasi-steady-state plot for ethanol (V_A = V_B = 530 mL; l = 3.51 mm; 38 wt% yeast cells in the gel; $D_e = 5.8 \cdot 10^{-10}$ m^2/s).

calcium alginate matrix, which is consistent with data reported in the literature (*5,11,12*).

To examine the dependence of D_e on varying alginate concentrations in the gel (1.4–3.8 wt%), 6 different gel plates were prepared. The effective diffusivities of lactose, glucose, galactose, and ethanol were determined simultaneously. To do this the solution in chamber A contained from the start all the solutes, each at a concentration of 30 g/L. The dependence of D_e on alginate concentration for glucose was negligible as can be seen in Fig. 5. Neither did lactose, galactose, nor ethanol show any dependence of D_e on alginate content. At the alginate concentrations (< 3.8%) used in our experiments, the exclusion effect, as well as the obstruction effect have been shown to be of minor importance. This is in contrast to results reported by Hannoun et al. (*9*), who found a decreased diffusivity when the alginate content was increased from 1–2%. From our experience, gel plates containing very low alginate concentrations are very soft, which can easily result in an additional convective flow through the gel, explaining their results. Furthermore, the shrinkage varies with alginate content which can introduce an apparent dependence of alginate concentration if the gel thickness is not accurately measured.

The effective diffusion coefficients are compared with the aqueous diffusivities for each solute in Table 1. Literature values of aqueous diffusivities have been recalculated to 30°C according to the Wilke-Chang equation (*20*). For all the solutes D_e was about 90% of the corresponding

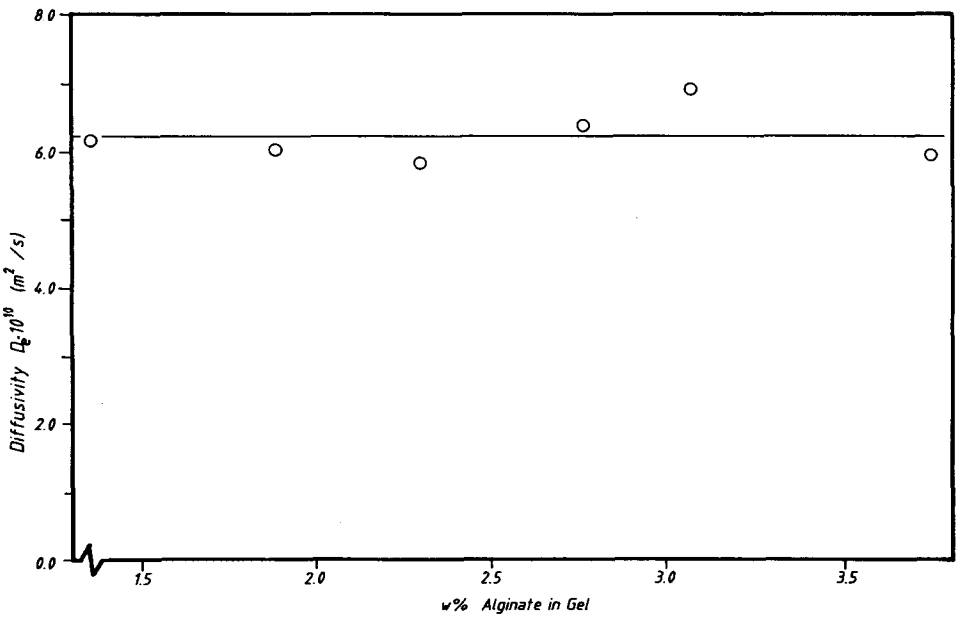


Fig. 5. Effective diffusion coefficient for glucose in calcium alginate without cells.

Table 1
Effective Diffusion Coefficients in Alginate Gel without Cells (D_{eo}) Compared with Aqueous Diffusivities (D_{aq}) at 30°C

	$D_{eo} \times 10^{10}$, m^2/s			
	Mean	S, %	$D_{aq} \times 10^{10}$, m^2/s	D_{eo}/D_{aq}
Lactose	4.36	4	5.0^{19}	0.87
Glucose	6.23	7	7.0^{18}	0.89
Galactose	6.38	7	7.0^{18}	0.91
Ethanol	12.48	8	13.50^{19}	0.92

D_{aq} values. Although diffusivity in general is dependent on concentration, this effect is less pronounced in this concentration range. At least the effect of 100 g/L glucose is similar to 90 g/L mixed sugars + 30 g/L ethanol. Furthermore, as the values obtained are to be used for calculations of the internal mass transfer of a system containing these solutes simultaneously, the effective diffusion coefficients determined in this way will give a more realistic picture of their diffusional behavior (7).

The dependence of the yeast cell content in the gel was determined for the four solutes simultaneously. The cell concentration varied between 10–45 wt% while the alginate concentration was kept constant (2.4–2.8 wt%). The concentration of the solutes was, as before, 30 g/L. D_e as a function of % yeast cells in the gel for glucose and lactose is shown in Fig. 6, and for galactose and ethanol in Fig. 7. The relative decrease in D_e for all the solutes is similar. At a cell concentration of 50 wt% yeast cells in the gel, the D_e values were decreased by 30–40% of the D_e value for the gel without cells. This is a somewhat stronger dependence than that reported for xylose (5). The present results indicate that the internal mass transfer has a more pronounced effect at higher cell contents.

To ensure that the D_e values obtained simultaneously for the four solutes were not influenced by each other, another experiment was performed. In this glucose was the single solute at a concentration of 100 g/L. The cell content was varied from 5–60 wt% and the alginate concentration from 0.5–1.2 wt%. The dependence was similar to that shown in Fig. 6 for glucose. This confirms the conclusions that simultaneous measurements give adequate results and that the alginate concentration is not important.

Shrinkage

The shrinkage for gels without cells was studied by preparing calcium alginate solutions with concentrations ranging from 0.6–4 wt%. The shrinkage was determined by simply weighing, and the results are shown in Fig. 8. The results are not very surprising if sodium alginate is

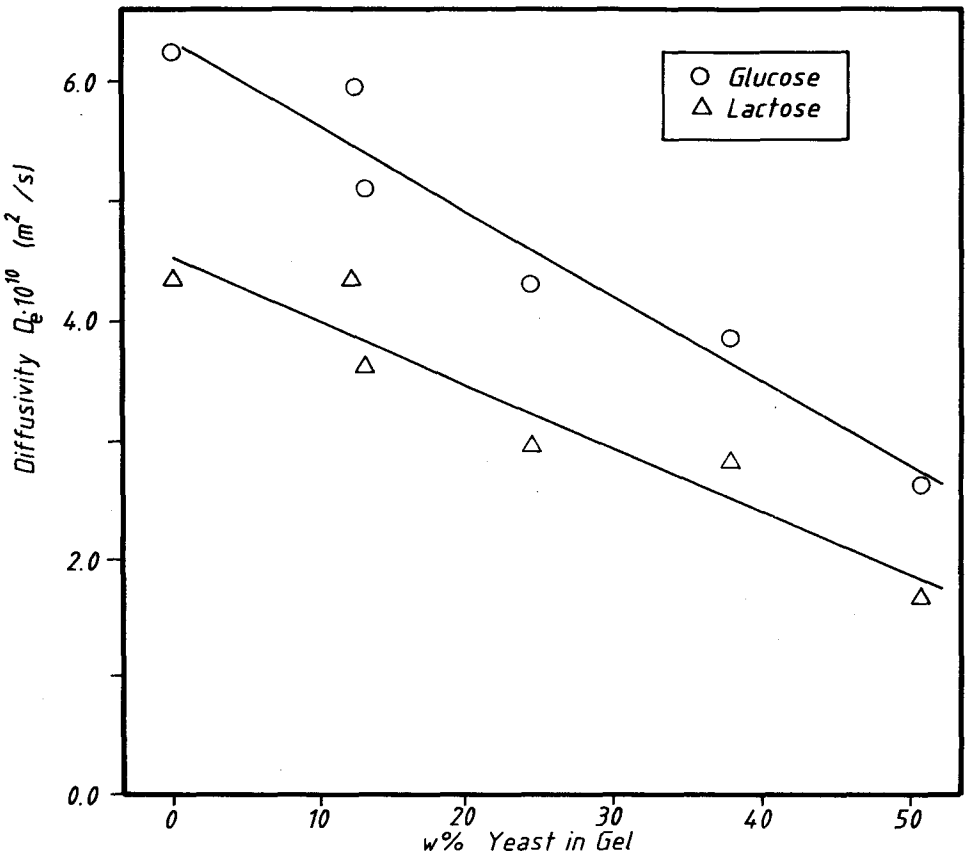


Fig. 6. Effective diffusion coefficients for glucose and lactose in alginate gel with varying yeast cell concentration.

regarded as a water-soluble "salt," which precipitates as an unsoluble "salt" when calcium ions are added.

When yeast cells are introduced into the gel the shrinkage will be less, as can be seen in Fig. 9. In these experiments the cell concentration was varied from 0–45 wt% in the solution, while the alginate concentration was kept constant at 2.1 wt% in the solution.

The yeast cells can be considered as a noncompressible volume which explains the fact that the apparent shrinking is less the more yeast cells the gel contains. It can be seen (in Figs. 8 and 9) that 2.1 wt% alginate appears in both experiments. Although the experimental conditions were similar, the results differed slightly—a shrinkage of 65% and 75%, respectively. It can be concluded that the composition of the gel, rather than the composition of the solution, should be used to compare data from different sources. This was actually the purpose of these experiments: To show that the gel composition will not be the same as the composition of the solution. The information given in Figs. 8 and 9 can be

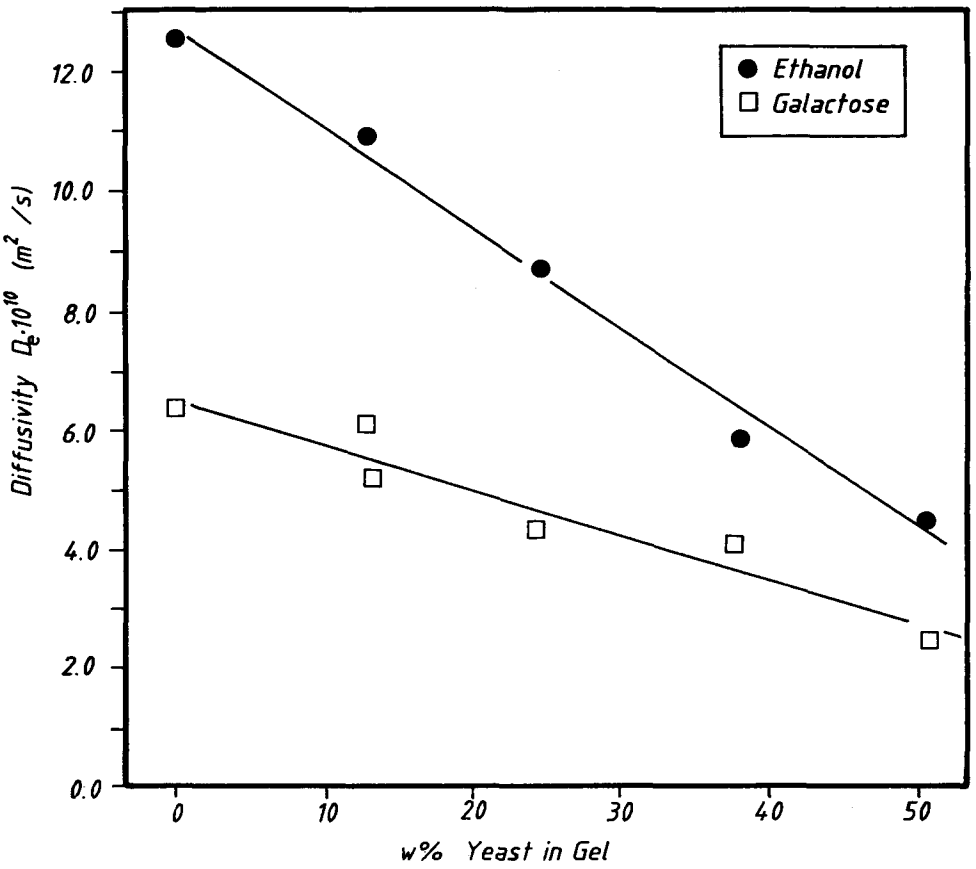


Fig. 7. Effective diffusion coefficients for galactose and ethanol in alginate gel with varying cell concentration.

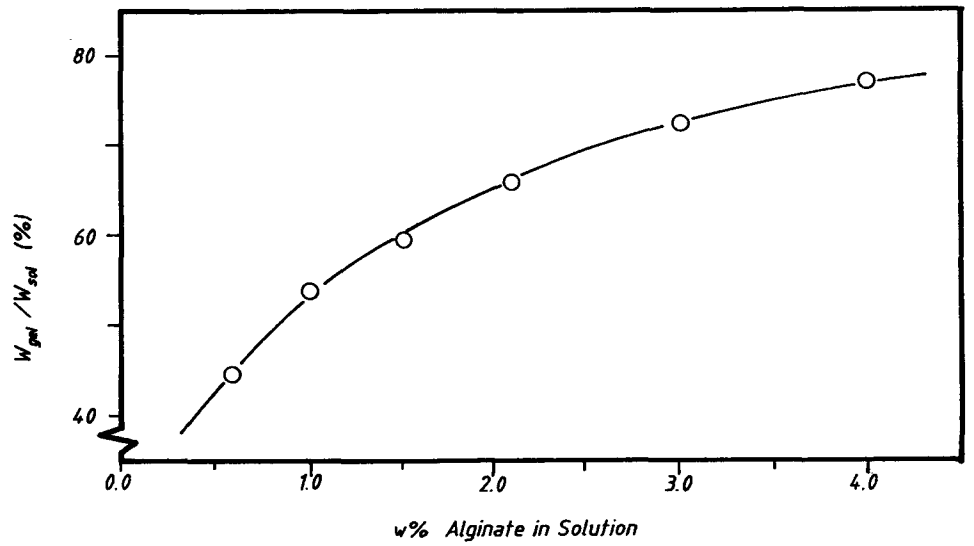


Fig. 8. Shrinkage for alginate gel as a function of wt% alginate in solution.

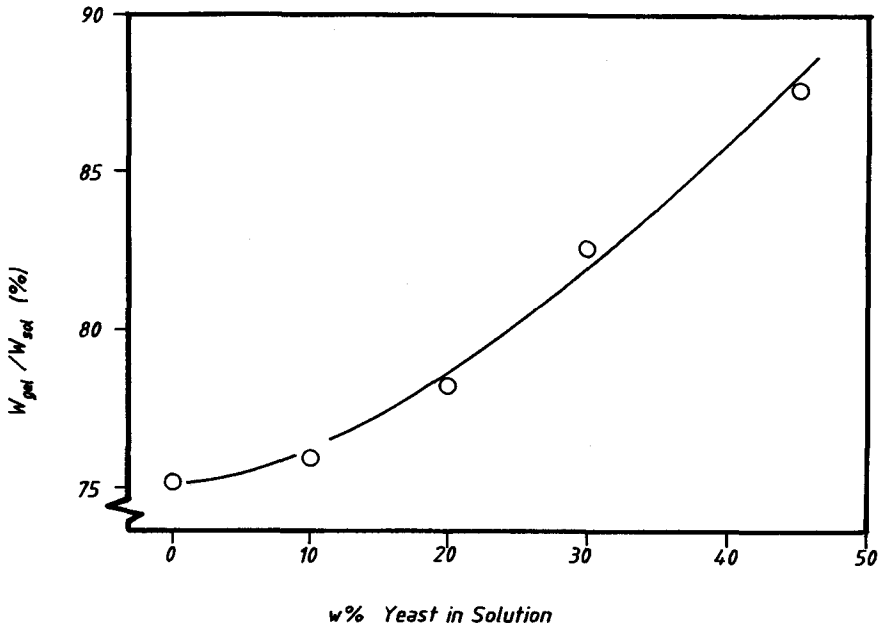


Fig. 9. Shrinkage for alginate gel as a function of yeast cell concentration in solution.

used to make an approximate comparison of diffusion coefficients from different sources.

Partition Coefficients

The partition coefficients (k) for lactose, glucose, galactose, and ethanol were determined for the gels containing yeast cells used for the shrinkage experiment. The partition coefficients were determined simultaneously for all the solutes, each with an initial concentration of 30 g/L. The alginate concentration in the gels was an almost constant 2.4–2.8 wt%. The results can be seen in Figs. 10 and 11. For lactose, glucose, and galactose the behavior of k as a function of yeast content is similar. The higher the yeast cell concentration, the lower the partition coefficient. Up to a concentration of about 25% yeast cells in the gel $k \approx 1 - \phi$, if the solid fraction ϕ is roughly estimated as wt% yeast cells in the gel. This indicates that the usual method of predicting k from the geometric exclusion effect is valid up to this concentration.

At higher cell concentrations the partition coefficient will be somewhat higher than predicted by $k = 1 - \phi$. The reason for this is not quite clear. If an additional volume is excluded because of steric hindrances at high cell concentrations, the partition coefficient should be lower than $1 - \phi$. As the yeast cells were inactivated, the glucose, galactose, and lactose were expected not to enter into the cells since this is an active transport. No lysis of the cells could be seen in microscope studies.

The partition coefficients for ethanol are higher. One reason for this could be that ethanol diffuses into the cells. It also indicates that ethanol

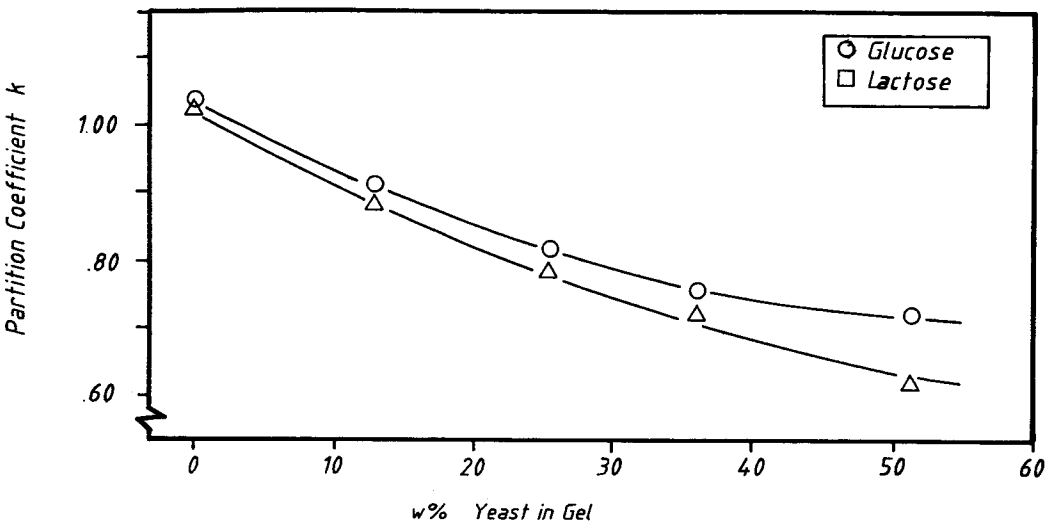


Fig. 10. Partition coefficients for glucose and lactose as a function of yeast cell concentration in the gel.

is adsorbed on to the pure alginate-gel. From the diffusion experiments it can, however, be concluded that ethanol did not penetrate the cells, as the lowering of D_e with increased cell content did not differ from that of the other solutes. The diffusion experiments, however, lasted for only 6

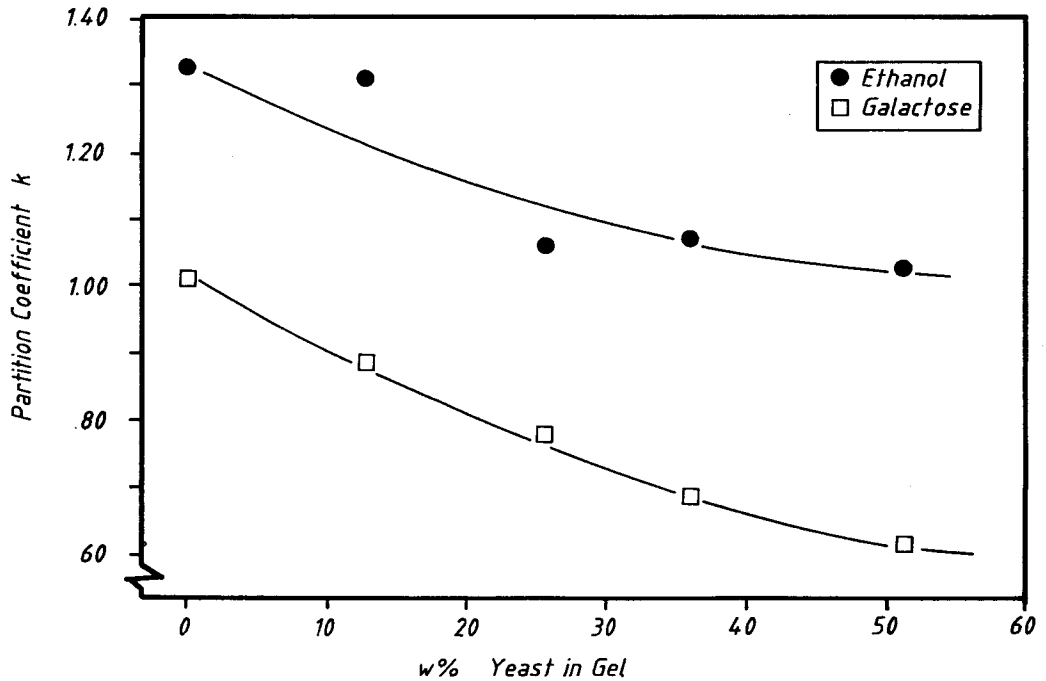


Fig. 11. Partition coefficients for galactose and ethanol as a function of yeast cell concentration in the gel.

h while the partition coefficients were measured after 4 d. Further studies on this matter are required.

General Expression for the Variation of D_e with Cell Concentration

If D_e for the four solutes in Figs. 6 and 7 is interpreted in the form of linear relationships with a least-square line fit this results in the following 4 equations.

$$D_e = 4.52 - 0.0532C_c \quad \text{for lactose} \tag{9}$$

$$D_e = 6.33 - 0.0717C_c \quad \text{for glucose} \tag{10}$$

$$D_e = 6.43 - 0.0742C_c \quad \text{for galactose} \tag{11}$$

$$D_e = 12.69 - 0.167C_c \quad \text{for ethanol} \tag{12}$$

where C_c is the weight fraction of yeast cells in the gel.

To study the influence of the cell content on D_e and to make a comparison with the models given in Eqs. (4), (6), and (7), these are normalized together with Eq. (10). C_c is replaced by the volume fraction of cells, ϕ, by multiplying by the gel density (~1030 kg/m^3) and dividing by the yeast cell density (~1100 kg/m^3). In this way D_{eo} will be equal to the effective diffusivity of an alginate gel without cells at $\phi = 0$. This comparison is shown in Fig. 12. For the case of simplicity, the experimental correlation and the data points for glucose only are shown, but as can be seen from Eqs. (9)–(12) the other solutes behave in a similar way. From the curves in Fig. 12 it can be concluded that the decrease in D_e is greater than that indicated by the exclusion effect (curve 1). An additional obstruction effect gives curve 2, according to the cell model (*15*) or Maxwell's model for spheres (*8*), which agrees very well with the experimental correlation. The random-pore model (*14*) results in an underestimation of D_e (curve 3).

As the effective diffusivity for pure alginate gel, D_{eo}, was shown earlier to be about 90% of the diffusivity in water (D_{aq}) the following procedure is proposed to estimate the effective diffusivity.

First, D_{eo} is estimated from D_{aq}, in this case, $D_{eo} = 0.90\ D_{aq}$; and then D_e is estimated from the relationship $D_e/D_{eo} = (1 - \phi)/[1 + (\phi/2)]$ where ϕ is the volume fraction of yeast cells.

CONCLUSIONS

Diffusion measurements have been carried out with a diffusion cell and the results have been evaluated using the quasi-steady-state method. Much effort has been put into preparing reproducible gel plates and measuring the gel plate thickness accurately.

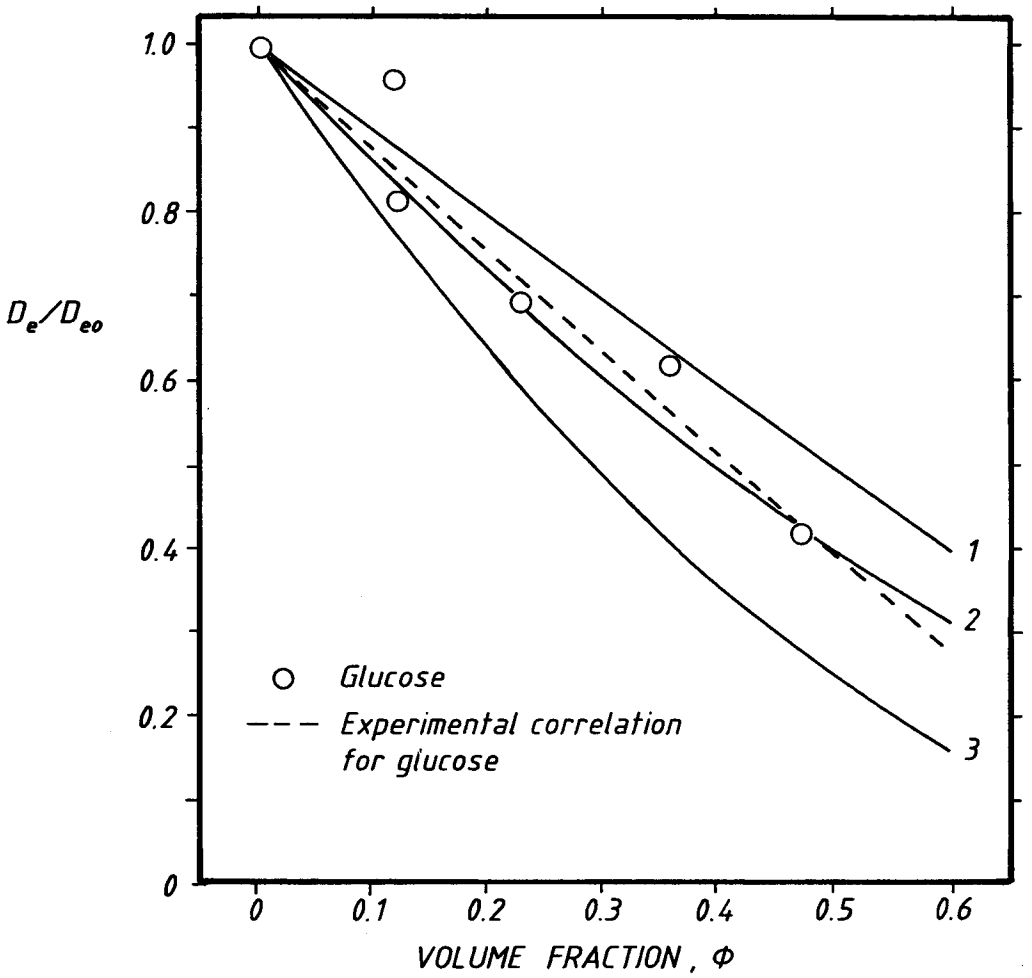


Fig. 12. D_e/D_{eo} as a function of ϕ. Comparison between experimental correlation for glucose and theoretical models. Exclusion effect $1 - \phi$; cell model $(1 - \phi)/[1 + (\phi/2)]$; random-pore model $(1 - \phi)^2$.

Partition coefficients for four solutes (lactose, glucose, galactose, and ethanol) and syneresis effects of the calcium alginate gels with and without yeast cells have been studied. The following results were obtained.

D_e was found to be independent of alginate concentration in the gel.

D_e decreased with increasing yeast cell concentration. An estimate of the decrease can be made using $D_e/D_{eo} = (1 - \phi)/[1 + (\phi/2)]$ where ϕ is the volume fraction of cells in the gel. The $(1 - \phi)$ factor lowers D_e because of the exclusive effect, and the $(1 + \phi/2)$ factor causes an additional decrease because of the obstruction effect.

D_{eo} in pure alginate gel was about 90% of the diffusivity in water for all the solutes.

NOMENCLATURE

A	area (m^2)
ic	concentration in solution (mol/m^3 or g/L)
c^*	concentration in gel (mol/m^3 gel)
c_A	concentration of solute in upstream compartment A (g/L)
c_B	concentration of solute in downstream compartment B (g/L)
c_{Ao}	concentration of solute in compartment A at t_o (g/L)
c_{Bo}	concentration of solute in compartment B at t_o (g/L)
C_c	concentration of yeast cells in the gel (wt%)
D	diffusion coefficient (m^2/s)
D_e	effective diffusion coefficient (m^2/s)
D_{eo}	effective diffusion coefficient in pure alginate gel (m^2/s)
D_{aq}	diffusion coefficient in water (m^2/s)
k	partition coefficient for the solute between gel and solution (m^3 solution/m^3 gel)
l	gel plate thickness (m)
N	molar flow (mol/s)
s	standard deviation (% of mean value)
t	time (min or s)
t_o	time at which quasi-steady-state was obtained (min or s)
V_A	volume of compartment A (m^3)
V_B	volume of compartment B (m^3)
wt%	weight percentage
w_{gel}	weight of gel (g)
w_{sol}	weight of alginate solution (g)
x	length in direction of diffusion (m)
ϕ	volume fraction of solids

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