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ORIGINAL PAPER

Diffusion of Cesium Ions in Agar Gel Containing Alkali Metal Bromides

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Abstract The present paper gives an account of different aspects of the tracer diffusion of Cs^+ ions in alkali metal bromides. We have measured the diffusion coefficients, D, of cesium ions in 1% agar gel medium at 25 °C using a zone-diffusion technique over a concentration range of 5×10^{-5} to $0.1\,\text{mol}\cdot\text{dm}^{-3}$. The values of the diffusion coefficients were found to deviate from theory, which are explained on the basis of different types of interactions occurring in the ion-gel-water system. The study is also focused on the effect of alkali metal bromides on the obstruction effect and activation energy for the tracer-diffusion of cesium ions in agar gel medium. It is observed that both parameters, extent of obstruction, α, and activation energy, E, decrease with increasing charge density of the cation of the supporting electrolyte. The influence of these trends is explained on the basis of competitive hydration between the ions and agar molecules, and the relative distortion in the water structure that is brought about by these different ions and agar molecules.

Keywords Diffusion · Cesium ions · Supporting electrolytes · Obstruction effect · Activation energy · Tracer diffusion

1 Introduction

Study of the diffusion of cesium ions provides important information in the biomagnifications in the aquatic food chains. It is generally one of the less mobile radioactive metals in the environment and preferentially adheres quite well to soil. It enters into the body through food, water and air. Hence studies on diffusion of cesium ions are important.

Onsager's theory [1] of tracer diffusion predicts that the diffusion of an ion is affected by the nature of different ions present in the supporting medium. This has been pointed out experimentally by several workers in different systems [2–4]. There are only a few reports on the effect of the nature of the ion and its concentration on the activation energy [5–8] for the diffusion process. This is mainly due to the experimental difficulties in measuring the

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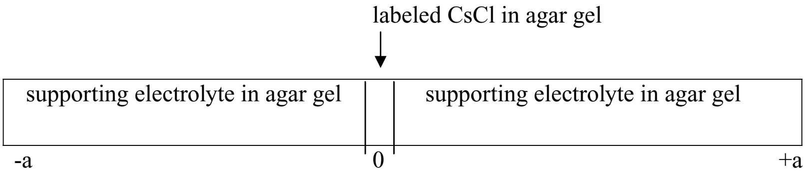
diffusion coefficient at higher temperatures. The present paper deals with measurements of the diffusion coefficient, D, at various electrolyte concentrations, c, as well as the obstruction effect and activation energy, E, for the tracer diffusion of cesium ions in alkali metal bromides using the zone-diffusion technique.

2 Experimental procedure

In the first set of experiments, the tracer-diffusion of Cs^+ ions, labeled with ^{134}Cs ($t_{1/2} = 2.07y$) was studied using a 1% agar gel medium containing LiBr, KBr and NaBr electrolytes. The concentration of the electrolyte was varied between 5×10^{-5} and 0.1 mol·dm^{-3} at 25 °C. In a second set of experiments, tracer diffusion coefficients were measured by varying the gel concentration between 1 and 2.5%, keeping the temperature at 25 °C as well as a constant electrolyte concentration of 0.1 mol·dm^{-3}. From these results the extent of obstruction in the presence of different electrolytes was determined. In the third set of experiments, the value of the activation energy, E, for the tracer-diffusion of Cs^+ ions in 1% agar gel containing different alkali metal bromides 0.1 mol·dm^{-3} each, was estimated by varying the temperature of the system between 25 and 45 °C. The diffusion coefficients, D, in all the sets of experiments were determined using the zone-diffusion technique.

The diffusion coefficients in these experiments were determined using the zone-diffusion technique [4]. The details of this technique are described below. The central zone of the column contains labeled Cs^+ ions in the form of CsCl, whereas the columns on either side of it contained the supporting electrolyte of interest (LiBr, NaBr or KBr) at the desired concentration. It is to be noted that the agar gel has a uniform concentration throughout the column being 1% in case of concentration dependence of the diffusion coefficient and activation energy studies, and was varied between 1–2.5% in the case of the obstruction effect studies. The corresponding column is shown below.



2.1 The Zone-diffusion Technique

A uniform and thin layer of tracer with the total initial activity, A_0, is allowed to diffuse into a semi-infinitely long diffusion column of length a from the medium corresponding to the following boundary conditions for the activity, $A_{(x,t)}$, at distance x from the origin and time t. The boundary conditions for activity at distance x and time t, $A_{(x,t)}$, are:

$$A_{(0,0)} = A_0 \quad \text{for } x = 0 \text{ and } t = 0$$

$$A_{(x,0)} = 0 \quad \text{for } x > 0 \text{ or } x < 0 \text{ at } t = 0$$

$$A_{(x,t)} = 0 \quad \text{for } x = \pm a$$

Under the above boundary conditions, Fick's second law takes the form:

$$A_{(x,t)} = \frac{A_0}{\sqrt{4\pi}\ Dt} e^{-x^2/4Dt}$$

where A_0 is the constant representing the total activity in the initial zone at $t = 0$ and $A_{(x,t)}$ is the radioactivity at distance x from the origin at time t. The values of the diffusion coefficients are obtained by plotting $\log_{10}A$ against x^2 which gives a straight line with slope given by

$$Slope = -\frac{1}{2.303 \times 4 \times D \times t}$$

Each value of D presented in Tables 2–4 is an average of at least four independent measurements. The reproducibility of the D values as evidenced by multiple runs of the experiment was within ±0.5%. The experimental details for determining the diffusion coefficients are described below.

A clean Pyrex tube with plane edges and uniform diameter of about 12 mm and a length of 30 cm was used. With one end closed, this tube was filled to half the length with the viscous gel solution containing the desired concentration of unlabeled electrolyte. This column was then allowed to cool to yield a solid form. After the gel cooled, the cork was gradually removed and one end of the gel was pushed out. A small piece of this gel was cut off with a clean, sharp blade along the edge of the tube, in order to yield a plane boundary. The gel column was then reinserted in the tube. An additional 1 mL of the gel was prepared with the electrolyte solution of interest that also contained labeled ions; this was added to the tube and rapidly cooled to form a solid gel. Leaving a 0.5 cm band of this gel in the column, the remaining part was cut off to form a plane surface as before. The whole gel column was then moved to one side of the tube so that a 0.5 cm labeled band remained at the middle of the tube. The remaining part of the tube was then filled with the same (unlabeled) gel, which was used for the first column under the labeled zone, and rapidly cooled. Once the column was ready, both ends of the tube were tightly corked and suspended horizontally in an automatically regulated temperature thermostat maintained to within ±0.1 °C of the desired temperature. The gel column was then extruded carefully after 24 h and the region of 5 cm on either side of the central zone was sliced into 0.5 cm samples. These were transferred into aluminum planchettes and dried under an infrared lamp. The radioactivity in each sample was measured using a well-type gamma-ray single-channel scintillation counter coupled to a NaI (TI) detector.

3 Results and discussion

3.1 Concentration dependence of *D*

The theoretical values of the tracer-diffusion coefficient of cesium ions in different supporting electrolytes have been calculated using the following Onsager–Gosting–Harned expression [9].

$$D_J^* = \frac{RT\lambda_J^o}{|z_J|\,F^2} - \frac{\lambda_J^o\,|z_J|\,F}{3N\varepsilon} 2.694 \times 10^{16}\sqrt{\frac{4\pi}{\varepsilon RT}}\left[1 - \sqrt{d\,(\bar{\omega}_J)}\right]\sqrt{\sum C_J z_J^2} \tag{1}$$

where R = gas constant (8.31451 $\text{J}\cdot\text{K}^{-1}\cdot\text{mol}^{-1}$); F = Faraday constant (96500 $\text{C}\cdot\text{mol}^{-1}$); N = Avagadro number (6.023×10^{23} mol^{-1}); ε = dielectric constant (78.5); T = temperature (298.15 K); C = Concentration ($\text{mol}\cdot\text{dm}^{-3}$); λ_l^{o} = limiting equivalent conductance (10^{-4} $\text{S}\cdot\text{m}^2\cdot\text{mol}^{-1}$); $d(\varpi_j)$ is the mobility function involving charges and limiting equivalent conductance of different ions in a given system given by the following equations, and other symbols have their usual meaning.

$$d\left(\bar{\omega}_J\right) = \frac{|z_1|}{|z_2| + |z_3|}\left[\frac{|z_2|\,\lambda_2^o}{|z_1|\,\lambda_2^o + |z_2|\,\lambda_1^o} + \frac{|z_3|\,\lambda_3^o}{|z_1|\,\lambda_3^o + |z_3|\,\lambda_1^o}\right] \tag{2}$$

In deriving Eq. (1) the ions were treated as point charges, justifiable at very low concentrations only. However, at higher concentrations one has to take into consideration the sizes of the ions present in the solution as they are reasonably close to each other. Taking this into account, Stokes *et al.* [10] made an empirical correction to the equation given by Onsager–Gosting–Harned [9] by replacing $\sqrt{C}$ by $\sqrt{C}/\{(1 + A'\sqrt{C})(1 + A'\sqrt{C})/\sqrt{2}\}$ where $A'\sqrt{C} = \kappa a$, in which a is the closest distance of approach of opposite ions and κ is the reciprocal radius of the ionic atmosphere.

The values of the various parameters are listed in Table 1. Substitution of these values in Eq. (1) leads to following limiting law expressions.

For LiBr system:

$$D^*_{\text{Cs}^+}/10^{-9}\ \text{m}^2\cdot\text{s}^{-1} = 2.055 - 0.374\sqrt{C} \tag{3}$$

For KBr system:

$$D^*_{\text{Cs}^+}/10^{-9}\ \text{m}^2\cdot\text{s}^{-1} = 2.055 - 0.458\sqrt{C} \tag{4}$$

For NaBr system:

$$D^*_{\text{Cs}^+}/10^{-9}\ \text{m}^2\cdot\text{s}^{-1} = 2.055 - 0.407\sqrt{C} \tag{5}$$

Whereas the extended-law expressions [10] take the following forms:

For LiBr system:

$$D^*_{\text{Cs}^+}/10^{-9}\ \text{m}^2\cdot\text{s}^{-1} = 2.055 - 0.374\left[\frac{\sqrt{C}}{(1 + 7.942\sqrt{C})(1 + 7.942\sqrt{C})/\sqrt{2}}\right] \tag{6}$$

Table 1 Values of various parameters for the calculation of the theoretical diffusion coefficient for diffusion of Cs^+ ions in alkali metal bromides

	Supporting Electrolyte		
Parameter	LiBr	KBr	NaBr
$\lvert z_1\rvert$	1	1	1
$\lvert z_2\rvert$	1	1	1
$\lvert z_3\rvert$	1	1	1
$\lambda_1^{\text{o}}/10^{-4}\ \text{S}\cdot\text{m}^2\cdot\text{mol}^{-1}$	77.26	77.26	77.26
$\lambda_2^{\text{o}}/10^{-4}\ \text{S}\cdot\text{m}^2\cdot\text{mol}^{-1}$	38.64	73.5	50.15
$\lambda_3^{\text{o}}/10^{-4}\ \text{S}\cdot\text{m}^2\cdot\text{mol}^{-1}$	78.17	78.17	78.17
$d(\varpi_J)$	0.584	0.508	0.554

Table 2 Variation of the tracer-diffusion coefficient of Cs^+ ions with the concentration of LiBr in 1% agar gel at 25 °C

	D_{Cs^+} /10^{-9} $m^2 \cdot s^{-1}$		
	Theoretical		
Conc./ $mol \cdot dm^{-3}$	Extended limiting law	Limiting law	Experimental
0	2.055	2.055	-----
5×10^{-5}	2.051	2.052	2.284
1×10^{-4}	2.050	2.051	2.194
5×10^{-4}	2.046	2.032	2.103
1×10^{-3}	2.045	2.023	2.084
5×10^{-3}	2.040	2.028	1.994
1×10^{-2}	2.039	2.017	2.018
0.05	2.038	1.971	2.166
0.1	2.032	1.936	2.416

For KBr system:

$$D^*_{Cs^+}/10^{-9}\ m^2 \cdot s^{-1} = 2.055 - 0.458\left[\frac{\sqrt{C}}{(1+1.400\sqrt{C})(1+1.400\sqrt{C})/\sqrt{2}}\right] \tag{7}$$

For NaBr system

$$D^*_{Cs^+}/10^{-9}\ m^2 \cdot s^{-1} = 2.055 - 0.407\left[\frac{\sqrt{C}}{(1+1.397\sqrt{C})(1+1.397\sqrt{C})/\sqrt{2}}\right] \tag{8}$$

The theoretical values of the diffusion coefficients calculated using Eqs. (3–8) are given in Tables 2–4 along with the experimental values and are presented graphically in Figs. 1–3.

An examination of these tables and figures reveals that the experimental values of the diffusion coefficients are greater than the theoretical ones over the entire concentration range, except at 5×10^{-3} $mol \cdot dm^{-3}$ for the LiBr and KBr systems. Further, a minimum in the plot

Table 3 Variation of the tracer-diffusion coefficient of Cs^+ ions with the concentration of NaBr in 1% agar gel at 25 °C

	D_{Cs^+} /10^{-9} $m^2 \cdot s^{-1}$		
	Theoretical		
Conc./ $mol \cdot dm^{-3}$	Extended limiting law	Limiting law	Experimental
0	2.055	2.055	-----
5×10^{-5}	2.051	2.052	2.512
1×10^{-4}	2.049	2.050	2.363
5×10^{-4}	2.044	2.046	2.166
1×10^{-3}	2.039	2.042	2.143
5×10^{-3}	2.022	2.016	2.094
1×10^{-2}	2.011	2.014	2.243
0.05	1.982	1.963	2.792
0.1	1.970	1.926	2.991

Table 4 Variation of the tracer-diffusion coefficient of Cs^+ ions with the concentration of KBr in 1% agar gel at 25 °C

	D_{Cs^+} /10^{-9} $m^2 \cdot s^{-1}$		
	Theoretical		
Conc./ mol·dm^{-3}	Extended limiting law	Limiting law	Experimental
0	2.055	2.055	-----
5×10^{-5}	2.051	2.055	2.370
1×10^{-4}	2.049	2.050	2.284
5×10^{-4}	2.042	2.044	2.166
1×10^{-3}	2.037	2.040	2.096
5×10^{-3}	2.018	2.022	1.994
1×10^{-2}	2.006	2.009	2.016
0.05	1.973	1.952	2.243
0.1	1.959	1.910	2.732

of D^*_{expt} *versus* $\sqrt{C}$ at 5×10^{-3} mol·dm^{-3} is a characteristic of all the curves compared to its absence in the theoretical curve. These observations over the entire concentration range can be accounted for on the basis of the different types of interactions occurring in the ion-gel-water system, *viz.*, ion-ion, ion-water and gel-water interactions as well as adsorption and obstruction effects, the details of which are discussed in our previous papers [11–13].

The higher value of the experimental diffusion coefficient compared to the theoretical one is attributed to the dominance of water-gel interactions along with: ion-water and ion-ion interactions over the obstruction and adsorption effects. Further, as can be seen from Tables 2–4 the deviations from the theoretical value vary with concentration for a particular system and also vary from system to system indicating that the degree of interaction of all these effects varies with the nature of the electrolyte as well as its concentration.

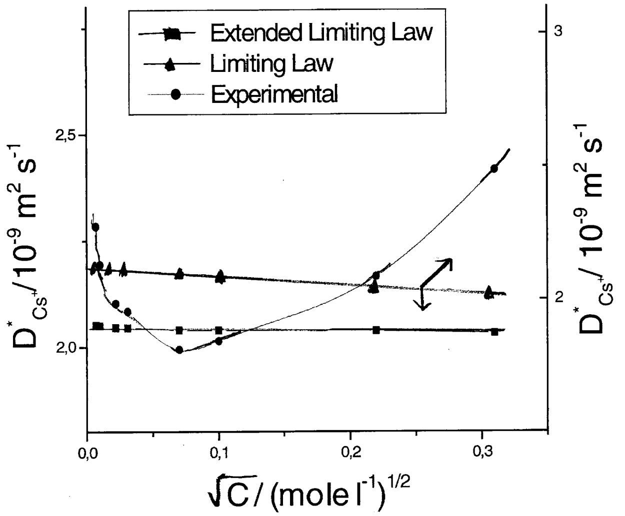


Fig. 1 Variation of tracer-diffusion coefficient of Cs^+ ions with square root of concentration of LiBr in 1% agar gel medium at 25 °C

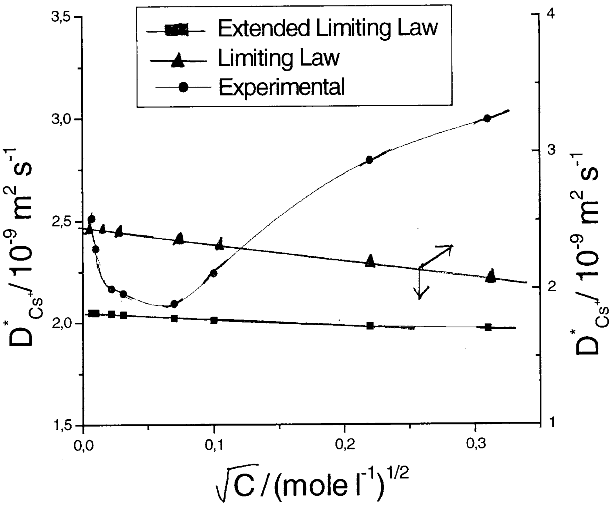


Fig. 2 Variation of tracer-diffusion coefficient of Cs^+ ions with square root of concentration of NaBr in 1% agar gel medium at 25 °C

The occurrence of a minimum in the D^*_{expt} versus $\sqrt{C}$ plot can be explained qualitatively on the basis of Wang's model [14] which considers the changes in the physical properties taking place at the microscopic level with electrolyte concentration.

The ions are fully hydrated at lower concentrations suggesting that any distortion present in the solvent water immediately surrounding each hydrated ion is the same as that existing at infinite dilution [21]. Therefore, the only effect of increasing salt concentration in this

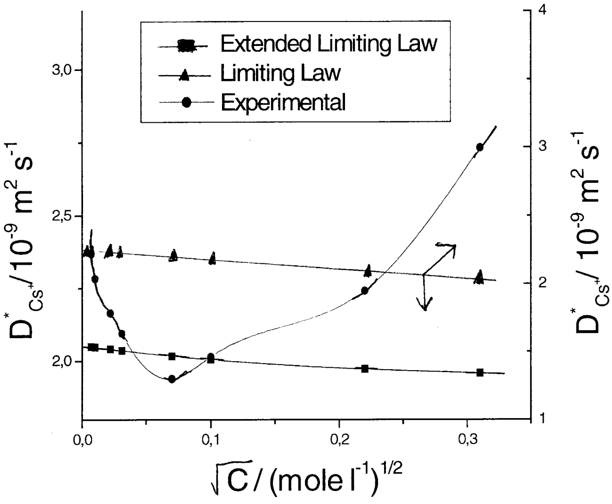


Fig. 3 Variation of tracer-diffusion coefficient of Cs^+ ions with square root of concentration of KBr in 1% agar gel medium at 25 °C

range is to increase the long-range ionic attraction (*i.e.*, the relaxation effect) which gives a decreasing trend in the D^*_{expt} with the concentration of electrolyte.

When the concentration of electrolyte increases, the distance as well as the number of water molecules between the two ions is reduced and it becomes increasingly difficult for water molecules to orient themselves and maintain the stable semi-crystalline structure of water that exists at infinite dilution. Hence, the semi-crystalline structure of water gets distorted giving rise to a decrease in the local dielectric constant of the solvent water. This decrease in dielectric constant increases the self-energy of the ions in the normal state while the energy of the activated state remains unaffected. Thus, the total energy barrier for the diffusion of ions is reduced leading to an increase in the diffusion coefficient with concentration of the electrolyte as observed.

3.2 Obstruction effect

The variation of the diffusion coefficient with weight fraction of agar as a function of the supporting electrolyte is shown in Figs. 4–6, which shows a linear relationship between the diffusion coefficient and ω obeying the following equation [15, 16].

$$D_g = D_s - a\omega \tag{9}$$

where D_g is the diffusion coefficient in the gel medium, D_s is the extrapolated value of D_g to zero agar content at a given concentration of the electrolyte and a is the slope of the plot of D_g *versus* ω.

Agar macromolecules have a much larger volume and a much smaller diffusion coefficientx than the diffusing ions. Due to the large size of the agar macromolecules these stationary agar molecules obstruct the path of ions, *i.e.*, the ions near an agar macromolecule have to diffuse along longer paths in order to get to the other side of the agar macromolecule. In the actual experimental work we compute the diffusion coefficient of ions by taking the macroscopic dimension of the diffusion tube as the length of the diffusion path, irrespective

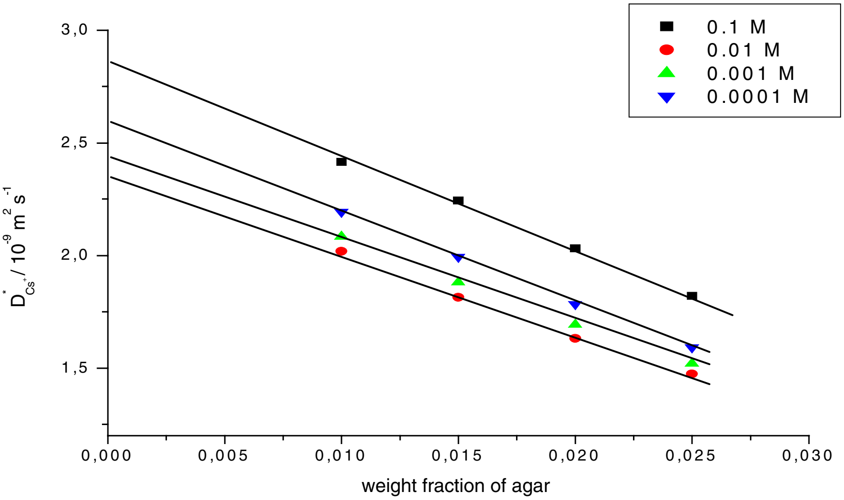


Fig. 4 Obstruction effect in the trace diffusion of Cs^+ ions at different concentrations of LiBr at 25 °C

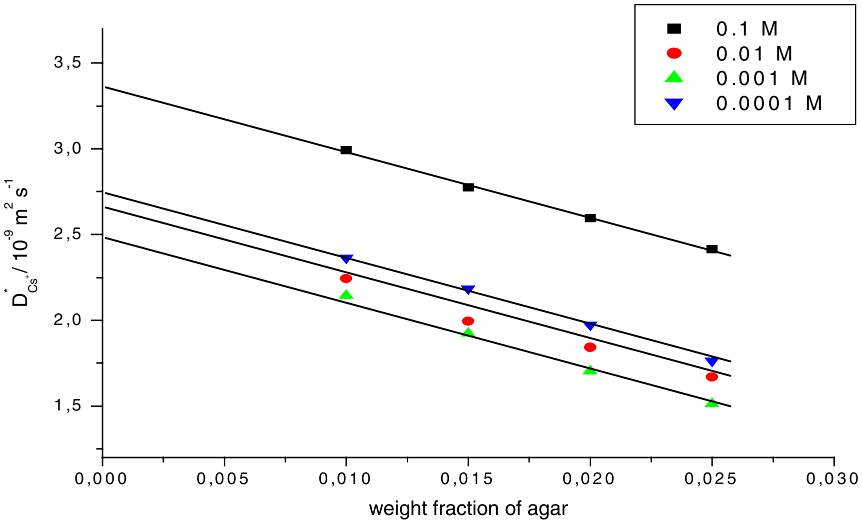


Fig. 5 Obstruction effect in the tracer-diffusion of Cs^+ ions at different concentrations of NaBr at 25 °C

of whether or not the diffusion path is blocked microscopically. This leads to a decrease in the value of the diffusion coefficient in the gel solution with increasing gel concentration, C. Thus, the obstruction effect appears to be due to the lengthening of the effective path of the diffusing ions because of the irregular network of macromolecules.

In the present studies we have calculated the obstruction effect in terms of $\alpha(\alpha = a/D_s)$, representing the relative lowering of the diffusion coefficient with the weight fraction of agar. The values of α obtained in various systems under various experimental conditions are

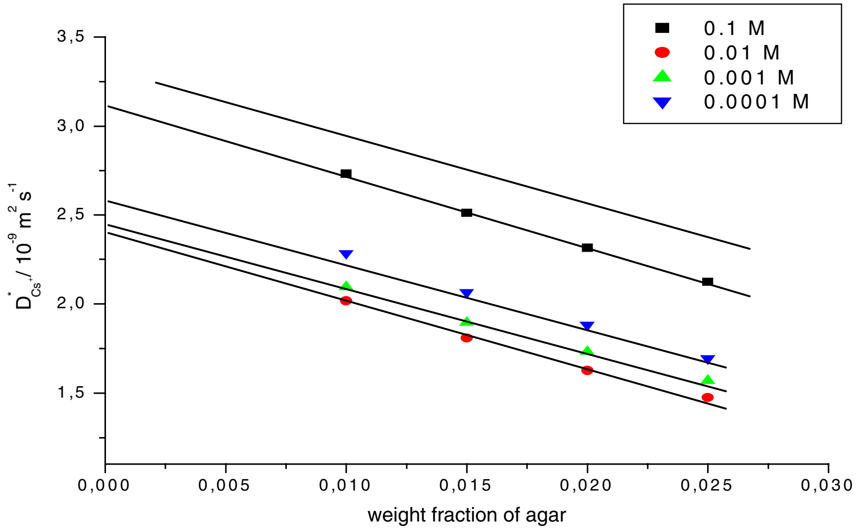


Fig. 6 Obstruction effect in the tracer diffusion of Cs^+ ions at different concentrations of KBr at 25 °C

Table 5 Variation of α and H for the tracer diffusion of Cs^+ ions in different alkali metal bromides

C/mol·dm^{-3}	α	H
Cs^+ in LiBr		
10^{-4}	17.76	26.28
10^{-3}	16.25	23.99
10^{-2}	15.21	22.42
10^{-1}	13.96	20.52
Cs^+ in NaBr		
10^{-4}	18.10	26.79
10^{-3}	17.6	26.04
10^{-2}	15.38	22.67
10^{-1}	14.51	20.81
Cs^+ in KBr		
10^{-4}	18.46	27.34
10^{-3}	17.85	26.42
10^{-2}	16.91	24.99
10^{-1}	14.86	21.89

recorded in Table 5 along with the H values that represents the extent of hydration of agar in terms of grams of bound water per gram of anhydrous agar.

H values listed in Table 5 are calculated using the equation [17]

$$\alpha = [1/d_a + H/d_w]\, d_w(\beta - 1) \tag{10}$$

where β is the shape factor for agar molecules = 5/3 for the needle-shaped agar macromolecules, and d_a and d_w are the densities of anhydrous agar and pure water, respectively.

An examination of Table 5 reveals that the observed trend in α at a given electrolyte concentration, C, for the tracer-diffusion of Cs^+ ions in alkali metal bromides is in the order

$$\alpha_{\text{LiBr}} < \alpha_{\text{NaBr}} < \alpha_{\text{KBr}}$$

which is in the reverse order of charge densities of the cation of the supporting electrolyte.

Among the various supporting electrolytes, the lithium ion has the smallest size and the highest charge density as compared to the other two ions. As a result it would attract more water molecules towards it as compared to the other two ions. Hence, in the LiBr system fewer water molecules are available for hydration of agar than are available for the NaBr and KBr systems resulting in a lower obstruction value.

3.3 Activation energy

The Arrehenius plots for the tracer diffusion of cesium ions in various alkali metal bromides are shown in Figs. 7–9. The values of the activation energies are calculated by least-square fitting of the data presented in this figure and are given in Table 6. It can be seen from Table 6 that in a given system the activation energy decreases with increasing concentration of the electrolyte.

This observation is in agreement with our earlier explanation of the diffusion coefficient on the basis of Wang's model, which states that the total energy barrier for diffusion decreases with concentration. Similar findings were reported earlier from our laboratory in various systems [18–20].

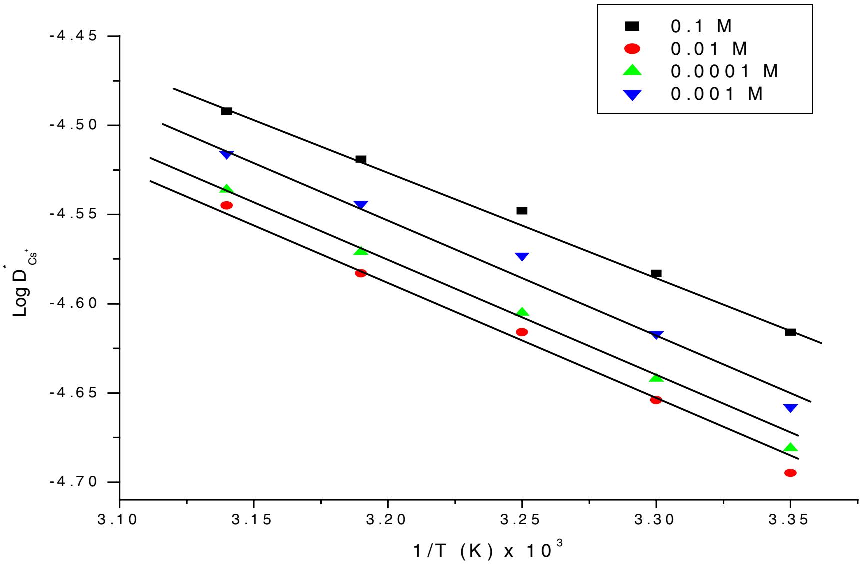


Fig. 7 Activation energy for the tracer-diffusion of Cs^+ ions in LiBr as a function of electrolyte concentration in 1% agar gel

It is also seen from Table 6 that at a given electrolyte concentration, the activation energy for the tracer diffusion of cesium ions in alkali metal bromides exhibits the following order:

$$E_{\text{KBr}} > E_{\text{NaBr}} > E_{\text{LiBr}}$$

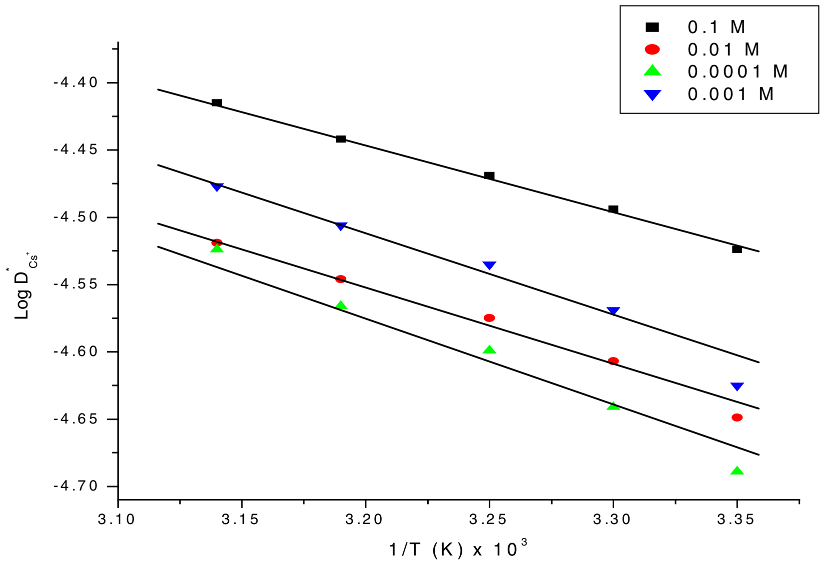


Fig. 8 Activation energy for the tracer diffusion of Cs^+ ions in NaBr as a function of electrolyte concentration in 1% agar gel

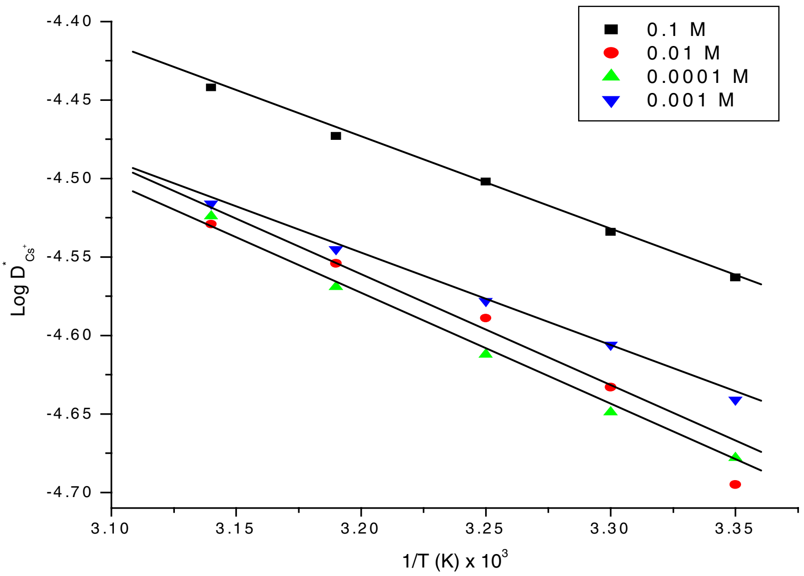


Fig. 9 Activation energy for the tracer-diffusion of Cs^+ ions in KBr as a function of electrolyte concentration in 1% agar gel

This trend in activation energy is explained qualitatively by considering the relative distortion in water structure caused by the ions from the supporting electrolyte and agar molecules [22, 23]. As mentioned earlier, the charge density of the different cations from the different electrolytes is in the order:

$$C.D._{\mathrm{Li}^+} > C.D._{\mathrm{Na}^+} > C.D._{\mathrm{K}^+}$$

Thus, if we consider diffusion of Cs^+ ions in various alkali metal halides, lithium ions melt the gel-water structure to a greater extent as compared to other metal ions. The distortion

Table 6 Activation energy as a function of electrolyte concentration for the tracer diffusion of Cs^+ ions in alkali metal bromides

Concentration/mol·dm^{-3}	E/kJ·mol^{-1}
LiBr	
10^{-4}	15.05 ± 0.2
10^{-3}	13.91 ± 0.1
10^{-2}	12.29 ± 0.1
10^{-1}	11.17 ± 0.1
NaBr	
10^{-4}	15.91 ± 0.3
10^{-3}	14.64 ± 0.2
10^{-2}	12.56 ± 0.1
10^{-1}	11.63 ± 0.1
KBr	
10^{-4}	16.76 ± 0.1
10^{-3}	15.06 ± 0.1
10^{-2}	13.87 ± 0.4
10^{-1}	11.98 ± 0.1

thus caused in the short-range crystalline structure of water leads to a decrease in the local dielectric constant of the medium which in turn increases the self energy of the ions in the normal state. Therefore, the total energy barrier for diffusion decreases with increasing distortion. This picture indicates that the activation energy required for the diffusion of a particular ion in different supporting electrolytes should be in the reverse order of the charge density of the cation from the supporting electrolyte. Our observation is in agreement with this prediction.

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