

STUDY OF RADIOIODINE SORPTION AND DIFFUSION ON MINERALS

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ABSTRACT

In this paper, the performance of adsorbing and retarding ^{125}I (substituted for ^{129}I) for mixed minerals as buffer, backfill material was investigated. The distribution coefficient K_d s by batch sorption experiments were determined for four kinds of minerals and one kind of bentonite under atmosphere. Sorption and desorption of radioiodine on several minerals were studied under low oxygen ambience at first time in the domestic, and apparent diffusion coefficient D_a of $^{125}\text{I}^-$ was determined for mixed minerals under atmosphere. The results as follows: Distribution coefficient K_d s of $^{125}\text{I}^-$ under atmosphere: bentonite is $3.23 \text{ ml}\cdot\text{g}^{-1}$, chalcopyrite is $72.42 \text{ ml}\cdot\text{g}^{-1}$, galena is $118.9 \text{ ml}\cdot\text{g}^{-1}$, pyrite is $1.93 \text{ ml}\cdot\text{g}^{-1}$, cinnabar is $55.48 \text{ ml}\cdot\text{g}^{-1}$, and the corresponding K_d s under low oxygen ambience: galena is $88.48 \text{ ml}\cdot\text{g}^{-1}$, chalcopyrite is $6.47 \text{ ml}\cdot\text{g}^{-1}$. when pH of solution was in the range of 2.25-12.26, K_d s of $^{125}\text{I}^-$ on chalcopyrite, galena, pyrite and cinnabar decreased with increase of pH under atmosphere. K_d s of $^{125}\text{I}^-$ on several minerals increased with increase of mineral ratio in mixed materials under atmosphere. Under the same condition, K_d s of $^{125}\text{I}^-$ on chalcopyrite and galena were larger than K_d s of $^{125}\text{IO}_3^-$. Sorption of $^{125}\text{I}^-$ on galena seems to be irreversible. Apparent diffusion coefficient D_a of $^{125}\text{I}^-$ in the mixed material was measured by the flow-through diffusion way, D_a values under atmosphere was given: $D_a=7.29\times10^{-12} \text{ m}^2\cdot\text{s}^{-1}$.

INTRODUCTION

The important long-lived fission product ^{129}I ($t_{1/2}=1.59\times10^7 \text{ a}$) normally exists as anion, and survives over a time span which is very long in comparison with the anthropogenic time frame. It was found that the migration of Iodine ions was very little retarded by geological barriers that retard cationic fission and actinides, therefore backfill materials are very important for radioiodine. Radioiodine can readily concentrate in the critical organ of human, which is very harmful to human. In America WRIT program for the assessment of the potential hazards of radionuclides in geological repositories, the term "Hazards Available Index(HA)" is used. The HA of ^{129}I is the largest. Barney and Wood (1980) used another safety index (HM) for radionuclide comparison. In terms of this hazard measure, ^{129}I is found in second

place among the radionuclides, surpassed only by ^{237}Np . The above considerations show that ^{129}I is one of the most critical radionuclides in a repository. It should therefore be considered with necessary care (1).

The migration of radionuclides in the geosphere is governed by three major mechanisms: (i) advection with the flowing water; (ii) diffusion, molecular or thermal; (iii) reaction/interaction with the fluid constituents or the medium. The subject of this work has been to search for minerals with good sorption properties for iodine, in view of their mixing with backfill materials-bentonite, study iodine sorption, desorption and diffusion on them, and research mechanism of iodine sorption on them preliminarily. Some minerals have been investigated in recent years. It seems that iodine can be absorbed by minerals containing metallic oxides and sulphides (2). From the various minerals of this kind available in China, Pyrite, chalcopyrite, galena, cinnabar have been chosen in this study, and sorption, desorption and diffusion experiments have been performed.

Sorption and desorption experiments under atmosphere.

The characteristics of the minerals, preequilibrated water and the experimental procedures are given below:

Preequilibrated water and minerals.

The sorption of iodine was studied on the following mineral species, and their main chemical compositions were given in Table I, II and III.

Table I. Average chemical composition of bentonite as possible back-fill material

Average composition as oxides, wt. %											
SiO ₂	Fe ₂ O ₃	FeO	Al ₂ O ₃	TiO ₂	MnO	CaO	MgO	K ₂ O	Na ₂ O	P ₂ O ₅	lost by
											total
burning											
64.11	2.29	0.08	11.59	0.077	0.051	1.54	1.97	0.132	0.104	0.018	18.03
											99.99

Tracer

$^{125}\text{I}^-$: Na^{125}I solution, carrier-free, obtained from the Radioisotope Division of CIAE.

$^{125}\text{IO}_3^-$: Na^{125}I was oxidized by concentrated nitric acid and dried by oven several times.

The tracers were diluted with preequilibrated water and approximate $2 \times 10^3 \text{ Bq}$ radioiodine was used for each sorption test. The liquid phase was obtained by equilibrating deionised water with bentonite for 35 days.

Table II. Average chemical composition of minerals choosed for sorption experiment

Names of minerals	Average composition as elements, wt. %							
	Cu	Pb	Fe	S	Na	Ca	Si	Hg
Chalcopyrite	33.27	0.024	29.27	33.98	0.34	1.94	0.038	/
Pyrite	0.044	0.022	52.96	45.98	0.094	0.079	0.047	/
Galena	0.018	85.99	0.063	13.20	0.014	0.012	0.034	/
Cinnabar	/	/	/	13.50	/	/	/	85.74

Table III. Chemical composition of preequilibrated water

Compositions	Content, mg·l ⁻¹	Compositions	Content, mg·l ⁻¹
H ₂ SiO ₃	24.5	Ti	0.005
Al	0.018	Mn	0.005
Fe	0.65	Sr	0.75
Ca	1.00	Ba	0.001
Mg	0.56	Li	0.065
K	2.00	B	0.013
Na	1.22	Zn	0.050

Sorption experiment

Batch sorption experiment were performed by shaking 0.25g of the crushed and sieved mineral or mixed minerals with 5 ml of spiked and preequilibrated water in the 10 ml centrifugation tube, varying the contact time from 1 to 43 days. At the end of shaking period the aqueous phase was separated by high speed centrifugation at 16000 rpm. An aliquot of the final solution was removed, and was measured for activity of radioiodine. The pH and Eh of the solution before and after contact with minerals were also recorded.

Desorption experiment

Desorption experiments were made from the samples previously used for the sorption experiments. The aqueous phase was removed from the sorption samples, and 5ml of non-spiked preequilibrated water was used for each experiment. The same procedure that was used in the sorption was used for separation of the phases and for the radioactivity measurement.

Radioactivity measurement

A thin window NaI detector was used to measure the γ -activity of the experimental samples. 2 ml of aqueous phase was taken for the measurement.

Calculation

Sorption ratio R_S and desorption ratio R_D were used in this work to express the sorption property of the minerals. The following equations were used to calculate sorption ratios and desorption ratios for radioiodine:

$$R_S = \frac{(C_0 - C_t)}{C_t} \cdot \frac{V}{W} \quad (\text{Eq.1})$$

Where

R_S = sorption ratio [$\text{ml} \cdot \text{g}^{-1}$]

C_0 = initial concentration of radioiodine in the spiked solution [$\text{Bq} \cdot \text{ml}^{-1}$]

C_t = concentration of radioiodine in the supernatant solution after the contact time t [$\text{Bq} \cdot \text{ml}^{-1}$]

V = volume of spiked solution [ml]

W = weight of materials used [g]

Because concentration of radioiodine in the solution is direct proportion to γ -activity of radioiodine in the solution, Eq.1 can be written as:

$$R_S = \frac{(A_0 - A_t)}{A_t} \cdot \frac{V}{W} \quad (\text{Eq.2})$$

Where

A_0 = initial \tilde{a} -activity of radioiodine in the spiked solution [$\text{counts} \cdot \text{min}^{-1}$]

A_t = \tilde{a} -activity of radioiodine in the supernatant solution after the contact time t [$\text{counts} \cdot \text{min}^{-1}$]

$$R_D = \frac{(A_0 - A_t - A'_t)}{A'_t} \cdot \frac{V}{W} \quad (\text{Eq.3})$$

Where

R_D = desorption ratio [$\text{ml} \cdot \text{g}^{-1}$]

A'_t = \tilde{a} -activity of radioiodine in the supernatant solution after the desorption experimental time t' [$\text{counts} \cdot \text{min}^{-1}$]

Results and discussion

Determination of R_S and sorption equilibrium time

R_S and sorption equilibrium time were investigated. The results are shown in table 4. the contact time varied from 1 to 43 days. It was close to sorption equilibrium when the contact time reached 32 days. 32-35 days was adopted as contact time in the

following sorption experiment, and experiments considered that R_s was nearly equal to K_d (sorption equilibrium constant).

Table IV. variation of R_s with contact time

Composition of materials	Contact time, days								
	2	5	9	13	20	24	32	36	43
Bentonite	2.14	1.29	2.62	2.38	3.01	3.36	3.23	3.43	3.86
10%Chalcopyrite 90%Bentonite	3.4	4.9	5.15	5.35	6.83	9.98	10.12	11.31	11.25
Chalcopyrite	20.1	23.42	31.83	36.49	52.80	69.63	72.42	79.11	82.41
10%Galena 90%Bentonite	6.56	10.41	11.40	13.00	13.70	12.37	12.91	12.94	13.62
Galena	36.18	62.96	88.4	103.9	118.3	126.2	118.9	124.7	130.8
10%Pyrite 90%Bentonite	1.58	1.22	1.64	0.93	1.13	2.18	2.47	2.82	2.36
Pyrite	9.95	5.30	5.13	2.81	2.82	2.81	1.93	2.99	2.65
10%Cinnabar 90%Bentonite	6.12	5.97	11.34	13.71	14.85	28.42	29.31	32.77	35.72
Cinnabar	13.39	8.56	15.63	34.70	40.60	51.46	55.48	63.72	59.34

pH of solution influence on K_d

0.1mol·l⁻¹HCL and 0.1mol·l⁻¹NaOH were used for adjusting pH of solutions. Influence of pH of solutions on K_d was show in table V. When pH was in the range of 2.36-12.57, K_d of ¹²⁵I⁻ on the materials decreased with increase of pH of solution. Eh values of solutions before and after contact with the materials were measured and shown in table 6. The Eh value of solution is a little high under atmosphere. When pH of solution increased, I⁻ becomes more easily to transfer to IO₃⁻ according to theoretical relation of pH and Eh of iodine in solution, K_d of ¹²⁵I⁻ decreased, and the amount of ¹²⁵I⁻ absorbed by materials diminished. This show that iodine absorbed by the experimental materials was possibly I⁻, not IO₃⁻.

Table V. K_d variation with pH of solution

Composition of materials	pH of solutions					
	2.36	4.42	6.22	8.35	11.05	12.57
10%Chalcopyrite 90%Bentonite	15.61	13.62	13.58	10.12	8.27	2.95
10%Galena 90%Bentonite	17.69	17.50	17.07	11.94	6.07	2.62
10%Pyrite 90%Bentonite	6.29	5.19	4.81	2.47	3.11	1.44

Table VI. Eh values of solutions before and after contact with the materials

Composition of materials	Before contact		After contact	
	pH	Eh, mV	pH _e	Eh _e , mV
10%Chalcopyrite 90%Bentonite	2.36	345.2	7.45	396.4
	4.42	328.5	7.58	374.5
	6.22	314.0	7.69	368.9
	11.05	302.7	7.83	351.2
	12.57	116.5	12.53	143.3
10%Galena 90%Bentonite	2.36	371.6	5.31	409.5
	4.42	348.2	6.03	398.2
	6.22	360.8	6.08	394.1
	7.91	332.6	6.22	388.7
	12.59	172.7	11.18	228.6

Influence of ratio of solid phrase to liquid phrase on Kd

Experiments have changed the amount of solid phrase and made liquid phrase unvaried to change ratio of solid phrase to liquid phrase. Influence of ratio of solid phrase to liquid phrase on Kd is shown in table VII. Kd of iodine on materials decreased with relative decrease of materials.

Table VII. Influence of ratio of solid phrase to liquid phrase on Kd

Composition of materials	Ratio of solid phrase to liquid phrase, g.ml ⁻¹			
	1:5	1:10	1:20	1:40
10%Chalcopyrie 90%Bentonite	29.64	22.24	10.12	5.68
10%Galena 90%Bentonite	44.98	19.67	11.67	5.59
10%Pyrie 90%Bentonite	6.68	3.78	2.47	2.52
10%Pyrite 90%Bentonite	71.35	45.88	29.31	17.69

Influence of mixing ratio of mineral and bentonite on Kd

Experiments have mixed minerals and bentonite and changed mixing ratio of mineral and bentonite. Influence of mixing ratio of mineral and bentonite on Kd was shown in table VIII. Kd of iodine on materials increased with increase of effective constituent in the mminerals.

Table VIII. Influence of mixing ratio of mineral and bentonite on Kd

Composition of materials	Mixing ratio of mineral and bentonite , [W _{mineral} /(W _{bentonite} +W _{mineral})×100%]					
	5%	10%	20%	50%	80%	100%
Chalcopyrite+bentonite	5.94	11.29	34.42	72.95	98.33	118.9
Galena+bentonite	3.58	10.12	30.25	48.59	63.69	72.42
Pyrite+bentonite	2.27	2.41	2.83	2.25	2.23	1.93
Cinnabar+bentonite	16.38	27.31	29.26	36.81	47.45	55.48

Sorption of $^{125}\text{IO}_3^-$ on minerals

Kd of $^{125}\text{IO}_3^-$ on minerals has been determined in the experiments, and is shown in table IX. For the same mineral, Kd of $^{125}\text{IO}_3^-$ on it is smaller than Kd of $^{125}\text{I}^-$ on it.

Table IX. Kd of $^{125}\text{IO}_3^-$ and $^{125}\text{I}^-$ on the same mineral

Composition of materials	Kd($^{125}\text{IO}_3^-$), ml·g ⁻¹	Kd($^{125}\text{I}^-$), ml·g ⁻¹
Chalcopyrite	22.27	72.42
Galena	15.95	118.9

Desorption experiment

Samples which show strong sorption capability to iodine were selected for desorption experiments. Desorption ratio R_D of iodine on chalcopyrite and galena are shown in table X. Desorption of iodine on galena is weaker than that on chalcopyrite.

Table X. Desorption ratio R_D of iodine on chalcopyrite and galena

Composition of materials	$R_d(^{125}\text{IO}_3^-)$, ml·g ⁻¹	$R_d(^{125}\text{I}^-)$, ml·g ⁻¹
Chalcopyrite	11.65	65.47
Galena	22.64	157.0

Sorption and desorption of iodine on materials under low oxygen ambience

Determination of R_S and sorption equilibrium time under low oxygen ambience

R_S and sorption equilibrium time under low oxygen ambience were investigated. The results are shown in table XI. the contact time varied from 1 to 20 days. It was close to sorption equilibrium when the contact time reached 15 days under low oxygen ambience. 17 days was adopted as contact time in the following sorption experiment, and experiments considered that R_S was nearly equal to Kd. Approximate equilibrium time of iodine on materials under low oxygen ambience is shorter than that under atmosphere.

Under low oxygen ambience, galena still has strong sorption capability to $^{125}\text{I}^-$, while sorption capability of chalcopyrite to $^{125}\text{I}^-$ become very weak. This is possible because that in the condition of low Eh value of solution CuFeS in the chalcopyrite is more easily destroyed through redox reaction than PbS in the galena, which makes R_s of $^{125}\text{I}^-$ on chalcopyrite decrease. Sorption of $^{125}\text{IO}_3^-$ on chalcopyrite and galena were also investigate under low oxygen ambience, and Kd of $^{125}\text{IO}_3^-$, $^{125}\text{I}^-$ on two minerals were shown in table XI. Two minerals show stronger sorption capability to $^{125}\text{I}^-$ than to $^{125}\text{IO}_3^-$.

Table XI. Variation of R_s with contact time under low oxygen ambience

Composition of materials	Contact time, days							
	2	4	6	10	12	15	17	20
Chalcopyrite	0.57	1.72	3.48	5.53	6.02	6.19	6.48	6.33
Galena	7.21	17.58	33.67	58.29	79.57	86.34	88.48	90.35

Table XII. Kd of $^{125}\text{IO}_3^-$, $^{125}\text{I}^-$ on chalcopyrite and galena under low oxygen ambience

Composition of materials	Kd($^{125}\text{IO}_3^-$), ml·g ⁻¹	Kd($^{125}\text{I}^-$), ml·g ⁻¹
Chalcopyrite	2.39	6.47
Galena	42.8	88.48

Desorption experiment under low oxygen ambience

Deorption experiment was only carried out with galena. Desorption ratio R_D of iodine on galena are shown in table XIII. Sorption of $^{125}\text{I}^-$ on galena seems to be irreversible.

Table XIII. Desorption ratio R_D of iodine on galena

Composition of material	$R_d(^{125}\text{IO}_3^-)$, ml·g ⁻¹	$R_d(^{125}\text{I}^-)$, ml·g ⁻¹
Galena	75.69	197.6

Diffusion of iodine in the mixing material

Calculation of apparent diffusion coefficient D_a

Schematically the flow-through diffusion cell consists of a thick disc of the porous material under investigation, in the present case the mixed materials consisted of wt.20% galena and wt.80% bentonite, sandwiched between two thin porous membrane (Fig.1). The concentration of the diffusion species is initially zero in the disc as well as in the constant volume V_2 of well-stirred solution in the right compartment. The concentration of the left compartment is kept at a constant C_0 ($C_1=C_0$) throughout the test and takes a value C_2 in the right compartment, determined by the quantity transported through the disc.

The diffusion through the disc is governed by the transport equation (3), (4), (5):

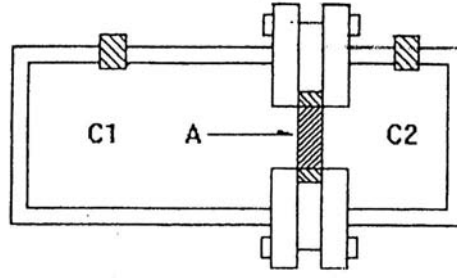


Fig.1 Schematic representation of the flow-through diffusion test

$$\frac{\partial C}{\partial t} = Da \frac{\partial^2 C}{\partial^2 x} \quad (\text{Eq.4})$$

designating the cross-sectional area of the disc by A and its thickness by L, the initial and boundary conditions can formally be written as:

$$C(t = 0, x > 0) = 0$$

$$C(t > 0, x = 0) = C_1 \quad (\text{Eq.5})$$

$$-\epsilon R A D a (\partial C / \partial x) \big|_{x=L} = V (\partial C / \partial t) \big|_{x=L}$$

where

C(x,t) or C = concentration of the considered species in the interstitial liquid phase [Bq·m⁻³].

Da = apparent diffusion coefficient of the considered species [m²·s⁻¹].

t = diffusion time of the considered species [s]

x = diffusion length of the considered species [m].

ε = porosity of the disc accessible by diffusion [m³·m⁻³].

R = retardation factor

A = cross-sectional area of the disc [m²].

V = volume of the left compartment [m³].

L = thickness of the disc [m].

Eq.4 has to be solved, and the solution for 0 ≤ x ≤ L is given as follows(5) :

$$\frac{C(x,t)}{C_{0(1)}} = 1 - \frac{x}{L} - \frac{2}{\pi} \sum_{n=1}^{\infty} \frac{1}{n} \sin \frac{n\pi x}{L} \exp\left(-\frac{D_i n^2 \pi^2 t}{\alpha L^2}\right) \quad (\text{Eq.6})$$

The total quantity Q(t) diffused in the outlet volume V₂ after time t can be calculated as the time integral of the flux F_L through the boundary x=L, and can be written as:

$$Q(t) = -D_i A \int_0^t \frac{\partial C}{\partial x} dt = C_2(t) V_2 \quad (\text{Eq.7})$$

$$\frac{Q(t)}{A L C_1} = \frac{D_i t}{L^2} - \frac{\alpha}{6} - \frac{2\alpha}{\pi^2} \sum_{n=1}^{\infty} \frac{(-1)^n}{n} \exp\left(-\frac{D_i n^2 \pi^2 t}{\alpha L^2}\right) \quad (\text{Eq.8})$$

If the concentration in the outlet volume C₂ « C₀, and diffusion time is sufficient long,

the third item of Eq.8 will be close to zero. Eq.8 can be approximately written as:

$$\frac{C_2 V}{ALC_1} = \frac{D_i t}{L^2} - \frac{\alpha}{6} \quad (\text{Eq.9})$$

Eq.9 is transformed to:

$$C_2 = \frac{D_i C_1 A}{LV_2} \cdot t - \frac{\alpha C_1 AL}{6V_2} \quad (\text{Eq.10})$$

Then, D_i can be calculated through the slope k of the straight line(C_2 - t):

$$D_i = kV_2 L / (C_1 A) \quad (\text{Eq.11})$$

$$\alpha = 6t_g D_i / L^2 \quad (\text{Eq.12})$$

where

D_i = intrinsic diffusion coefficient of considered species [$\text{m}^2 \cdot \text{s}^{-1}$]. $Da = D_i / \alpha$

t_g = the intercross time value of the straight line and x axis [s].

α = capacity factor.

k = slope of the straight line of steady state in diffusion curve.

$$Da = L^2 / (6t_g) \quad (\text{Eq.13})$$

Because the two thin porous membranes effected the diffusion of considered species, Da determined in the experiment should be corrected by Eq.14, and correction curve of Da can be seen Fig.2 (5), (6):

$$\frac{\langle Da \rangle}{Da} = \frac{1 + 2BK}{1 + 6BK + [6 + 2(3 + BK / B^2)] \cdot (BK)^2} \quad (\text{Eq.14})$$

$$B^2 = \frac{\langle D_i \rangle^2}{D_{f,i}^2} \times \frac{h^2 / t_{gf}}{L^2 / t_g} \quad (\text{Eq.15})$$

$$BK = \frac{2h}{L} \times \frac{\langle D_i \rangle}{D_{f,i}} \quad (\text{Eq.16})$$

where

$\langle Da \rangle$ = apparent diffusion coefficient uncorrected [$\text{m}^2 \cdot \text{s}^{-1}$].

Da = apparent diffusion coefficient corrected [$\text{m}^2 \cdot \text{s}^{-1}$].

$D_{f,i}$ = intrinsic diffusion coefficient of considered species while single membrane diffusion experiment [$\text{m}^2 \cdot \text{s}^{-1}$].

$t_{g,f}$ = the intercross time value of the straight line and x axis while single membrane diffusion experiment [s].

h = thickness of membrane [m].

Results and discussion

The disc compressed for diffusion experiment is consisted of galena (wt.20%) and bentonite (wt.80%). The flow-through diffusion experiments have been investigated,

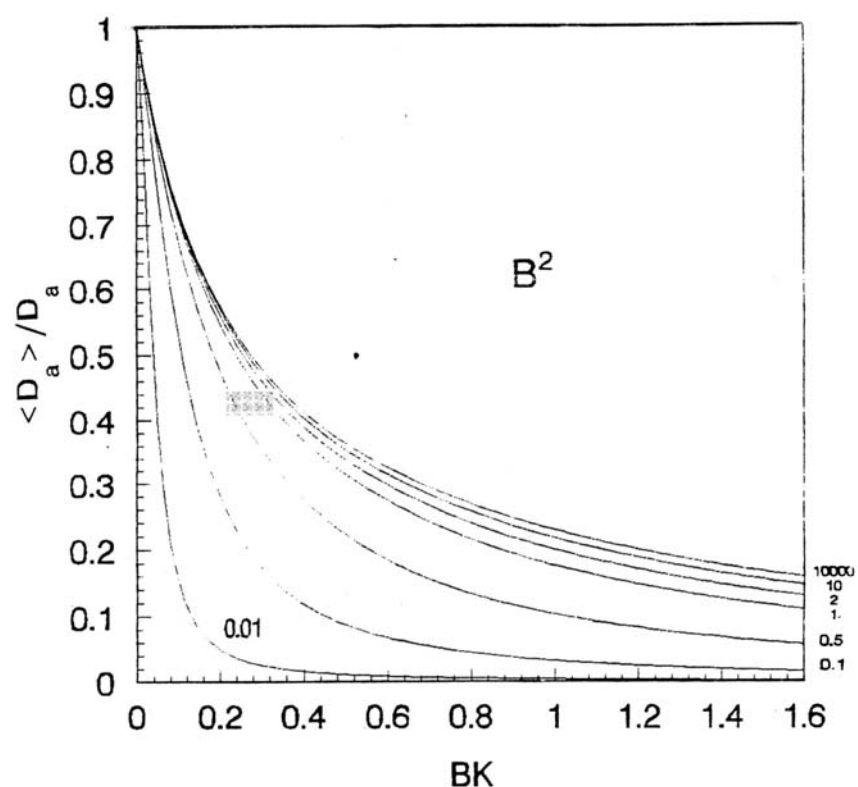


Fig.2 Correction curve of apparent diffusion coefficient D_a

and the total diffusion curve and single membrane diffusion curve of $^{125}\text{I}^-$ are shown in Fig.3 and Fig.4.

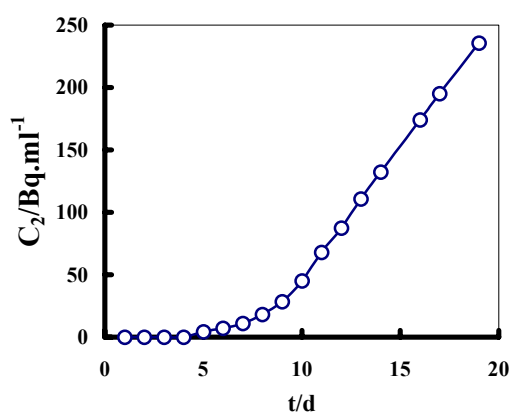


Fig.3 Total diffusion curve of $^{125}\text{I}^-$

Straight line equation of steady state in Fig.3 can be written as:

$$C_2 = 21.2t - 165.2 \quad (\text{Eq.17})$$

While $k_1 = 21.2 \text{ [Bq}\cdot\text{ml}^{-1}\cdot\text{d}^{-1}]$, $V_2 = 200 \text{ [ml]}$, $L = 5 \text{ [mm]}$, $C_{1(0)} = 1.15 \times 10^3 \text{ [Bq}\cdot\text{ml}^{-1}]$, $A = 12.64 \text{ [cm}^2]$, according to Eq.11, gives:

$$\langle D_i \rangle = 1.69 \times 10^{-10} \text{ [m}^2\cdot\text{s}^{-1}]$$

while $C_2 = 0 \text{ [Bq}\cdot\text{ml}^{-1}]$, $t_g = 7.8 \text{ [d]}$, and according to Eq.13, gives:

$$\langle Da \rangle = 6.18 \times 10^{-12} \text{ [m}^2\cdot\text{s}^{-1}]$$

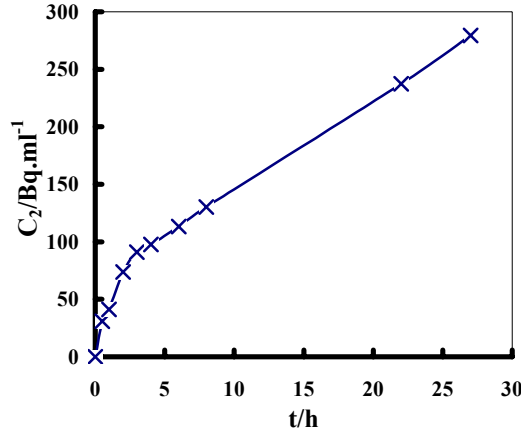


Fig.4 Single membrane diffusion curve of $^{125}\text{I}^-$

Straight line equation of steady state in Fig.4 can be written as:

$$C_2 = 7.75t + 66.89 \quad (\text{Eq.18})$$

While $k_2 = 7.75 \text{ [Bq}\cdot\text{ml}^{-1}\cdot\text{h}^{-1}]$, $V_2 = 200 \text{ [ml]}$, $h = 0.1 \text{ [mm]}$, $C_{1(0)} = 1.02 \times 10^3 \text{ [Bq}\cdot\text{ml}^{-1}]$, $A = 26.81 \text{ [cm}^2]$, according to Eq.11, gives:

$$D_{f,i} = 1.57 \times 10^{-10} \text{ [m}^2\cdot\text{s}^{-1}]$$

$t_{g,f}$ can't be measured in the experiment. It was gotten by trial and error approach.

$^{125}\text{I}^-$ flows through the single membrane in a very short time, so the $t_{g,f}$ is very small.

From Fig.2 we can see that the smaller $\langle Da \rangle / Da$ will changes, the smaller $t_{g,f}$ is and the larger B^2 is.

While $t_{g,f} = 60 \text{ [min]}$, $B^2 = 62.47$, $BK = 0.043$, and $\langle Da \rangle / Da = 0.848$.

While $t_{g,f} = 5 \text{ [s]}$, $B^2 = 0.087$, $BK = 0.043$, and $\langle Da \rangle / Da = 0.847$.

$\langle Da \rangle / Da$ changes very small with variation of $t_{g,f}$ in the range from 5 [s] to 60 [min].

trial and error approach used for estimating $t_{g,f}$ is feasible. In the experiment, 5 [s] was taken for $t_{g,f}$, gives:

$$Da = \langle Da \rangle / 0.848 = 7.29 \times 10^{-12} \text{ [m}^2\cdot\text{s}^{-1}]$$

$^{125}\text{I}^-$ can be retarded by the mixed material to some degree.

Conclusions

Under atmosphere, $^{125}\text{I}^-$ can be strongly retarded by chalcopyrite and galena; K_d of $^{125}\text{I}^-$ on chalcopyrite and galena is larger than that of $^{125}\text{IO}_3^-$; K_d of $^{125}\text{I}^-$ on chalcopyrite and galena decreased with increase of pH when pH in solution is in the range of 2.25-12.26; Sorption of $^{125}\text{I}^-$ and $^{125}\text{IO}_3^-$ on chalcopyrite and galena seems to be irreversible.

Under low oxygen ambience, $^{125}\text{I}^-$ can be strongly retarded by galena while chalcopyrite has weak sorption capability to $^{125}\text{I}^-$ and $^{125}\text{IO}_3^-$; Sorption of $^{125}\text{I}^-$ and $^{125}\text{IO}_3^-$ on galena seems to be irreversible.

Apparent diffusion coefficient D_a of $^{125}\text{I}^-$ in the mixed materials was measured by the flow-through diffusion way, D_a value under atmosphere was given: $D_a = 7.29 \times 10^{-12} [\text{m}^2 \cdot \text{s}^{-1}]$, and $^{125}\text{I}^-$ can be retarded by the mixed material to some degree.

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