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EFFECT OF GAMMA IRRADIATION ON KINETICS OF ION EXCHANGE OF Zn (II), Co (II), Pb (II), Mg (II) ON DOWEX 50 WX_8 (NH_4^+) IN AQUEOUS ACETONE – AMMONIUM BUTYRATE MEDIA.

***Kamble R. S.**

Department Of Chemistry, Bhogawati Mahavidyalaya, Kurukali (MS) – 416001.

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***Corresponding Author**
Kamble R. S.
Department Of Chemistry,
Bhogawati Mahavidyalaya,
Kurukali (MS) – 416001.

ABSTRACT

During the last few years many varieties of organic and inorganic exchangers have been synthesized and employed with notable success in every field of science and industry. Of the large number of ion exchangers identified as potentially useful in nuclear process industry, the vast majority of ion exchangers used today are synthetic organic resins. However, these materials are quite susceptible to radiation chemical decomposition and present knowledge is inadequate to provided meaningful mechanistic interpretations of many of the problems resulting from their radiation damage. It was thus of interest to examine the physiochemical parameters of resins. Many factors change the ion exchange kinetics including exchange capacity, moisture content, cross linkages, rate of diffusion, concentrations of ionic species in solution which in turn are influenced by radiation.[1-4] Hence the kinetics also should be affected by gamma irradiation treatment. With this view kinetic studies ion exchange reactions with various metal ions like Zn, Pb, Mg, and Co have been made on un-irradiated and gamma irradiated resins.

In case of ion exchangers having a polymeric organic matrix, the cation of radiation leads to the following characteristics reactions.

1. Breakdown or loss of functional groups.
2. Formation of new functional groups.
3. De-polymerization of the matrix.
4. Cross linkage of the matrix.
5. Colour change of the resin.

As a result of these reactions, there can be observed changes in the capacity and basicity of the functional groups as well as variations in the swelling and consequently sorption

characteristics of the exchanger. Increase or decrease in the moisture contents, loss in weight of the resin are some of the changes which can occur as a result of gamma irradiation of the resin.

KEYWORDS: Gamma irradiated and un-irradiated resin, aqueous acetone - ammonium butyrate, diffusion coefficient (D), rate constant (K), half exchange time ($t^1/_2$), inter-diffusion coefficient (D).

INTRODUCTION

The radiation stability of porous exchangers was found to be considerably higher than that of non porous exchangers.[5] The effect of gamma irradiation on the physical properties of Dowex 50 WX_8 resin in H^+ and NH^+_4 forms studied. The effect of percentages of acetone on the kinetics of exchange of metal ions on irradiated and un-irradiated samples of resin was compared. The kinetic parameters such as half exchange time ($t^1/_2$), inter-diffusion coefficient (D), and the exchange rate constant (K) were computed.

The resin on irradiation, the rates of exchange of Zn, Pb, Mg, Co on Dowex 50 WX8 (NH+4) in aqueous acetone ammonium butyrate (0.02M) was studied. The effect of percentages of acetone on the kinetic of exchange of metal ions on irradiated and un-irradiated samples of resins was compared. The probable properties of irradiated and normal resin are discussed.

MATERIAL AND METHODS

The soluble impurities from resin were removed by repeated soxhlet extractions using water. The resin sample soaked in water was placed in a 3 cm X 50 cm column and repeatedly 1N NaOH, 1 M HCl solutions were passed through it. Between each cycle, the resin was washed with de-ionized water and occasionally with ethanol which removes the non polymerized organic impurities. After five, six cycles, the resin was converted into the desired form.

Irradiation of resin

Cobalt -60 source was used for irradiation of resin sample.^{60} Co decays by emission of β particles to an excited ^{60} Ni nucleus and it gets stability by emitting two gamma photons, each of 1.7323 MeV and 1.33248 MeV energy. This decay scheme is shown in fig no 1.

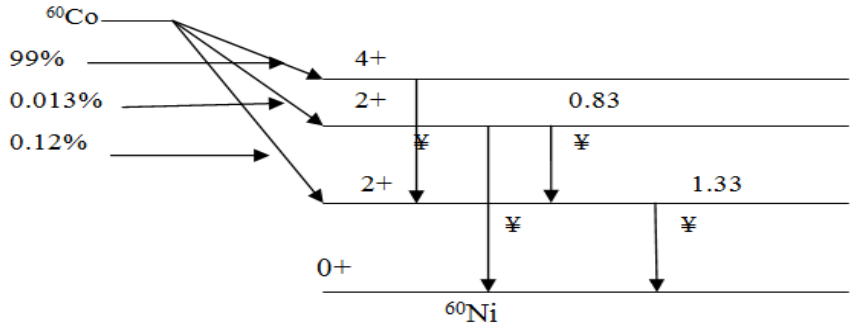


Fig 1. ^{60} Co Decay Scheme.

The dose rate was calculated using the relation

$$\text{Dose} = \frac{\text{Gy min}^{-1}\ (\text{Cm}^3)\quad 0.647 \text{ x}\quad 106 \text{ x } \Delta A}{\text{Ed eC } (Fe^{3+})\ 2a}$$

The Δ A is the difference in absorption between the irradiated and non irradiated solution of density, d is the optical path length in centimeters and the value of G is taken as 15.6.

15 g air dried sample was irradiated up to the desired dose. The change in the exchange capacity of resin after irradiation calculated by,

$$\text{Capacity} = \frac{(50 \text{ M NaOH}) - (\text{X M Acid})}{\text{Sample weight}}$$

The weighed air dry sample after radiation treatment were washed thoroughly with distilled water to remove degradation. Products were dried over P_2O_5 in a desicator for long time and reweighed. The difference in weight is considered as a measure of degradation.

Solutions

1. 0.1 N stock solutions of magnesium, zinc, lead and cobalt were prepared from A. R. grade salts of B. D. H. and S. R. L by using double distilled water.
2. Acetone- 0, 30, and 50 %.
3. Ammonium butyrate solution – 2M.
4. E. D. T. A. – 0.001M.

Procedure

Exactly 1 g of air dried resin Dowex 50 W X8 (NH_4^+ form) was taken in an Erlenmeyer flask. The mixture of acetone-ammonium butyrate (0.02M) was then added. Proper quantity of metal ion solution was poured into it at noted time, so that the total volume of the mixture

becomes 50 ml. the amount of metal ions exchanged on the resin was estimated at different time intervals from the concentration difference before and after the ion exchange. The kinetic studies for Mg, Zn, Pb and Co (0.004 M) were carried out at 296^{o} K. The experiments were performed using un irradiated and gamma irradiated (0.45 X 10^{6} rads) resins.

Table 1: Effect of the capacity of the state of resin during irradiation (Dose – 0.45 MGY).

Ion Exchanger	Form	Decrease in capacity %	
		Air dry	Under water
Dowex 50 WX_8	NH_4^+	5.0	8.7

Table 2: kinetic parameters for the exchange of Mg, Zn, Co, Pb on un irradiated and Irradiated Dowex 50W X8 (NH_4^+) in Aqueous Acetone Ammonium Butyrate (o.02M) at 296^{o}K.

Acetone % V/V	$t^1/_2$ (min)		$Dx10^{-10}$ m^2 sec^{-1}		Kx 10^{-3} (min^{-1})	Kx 10^{-4} (min^{-1})	Kx 10^{-3} (min^{-1})	Kx 10^{-4} (min^{-1})
	a	b	a	b	a	a	b	b
Mg (II)								
0	75	85	0.36	0.317	8.29	-	5.18	9.7
30	115	95	0.234	0.284	5.06	9.90	4.03	9.8
50	125	130	0.216	0.207	5.0	9.90	5.4	7.2
Zinc (II)								
0	160	150	0.168	0.18	4.8	-	7.67	15.3
30	230	160	0.117	0.168	5.06	6.90	4.60	8.22
50	190	135	0.142	0.2	5.0	-	3.54	-
Cobalt (II)								
0	130	115	0.207	0.234	13.3	-	8.22	8.29
30	125	120	0.216	0.225	23.03	-	4.42	8.21
50	120	125	0.225	0.216	32.2	-	2.99	8.06
Lead (II)								
0	145	130	0.186	0.207	20.26	-	6.05	9.07
30	220	215	0.122	0.125	20.75	-	4.21	17.27
50	225	215	0.12	0.125	17.27	-	4.14	8.84

a = gamma irradiated resin, b = un-irradiated resin.

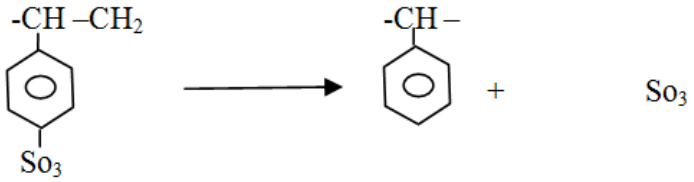
RESULTS AND DISCUSSION

The colour of the resin was changed from golden yellow to brownish on irradiation. The capacity of the resin was found to be decreased on gamma irradiation (**Table No 1).** Irradiation causes the decrease in the weight of the resin samples. The variation of loss in capacity and weight with dose rate was studied on air dried resin samples.[6] The results are

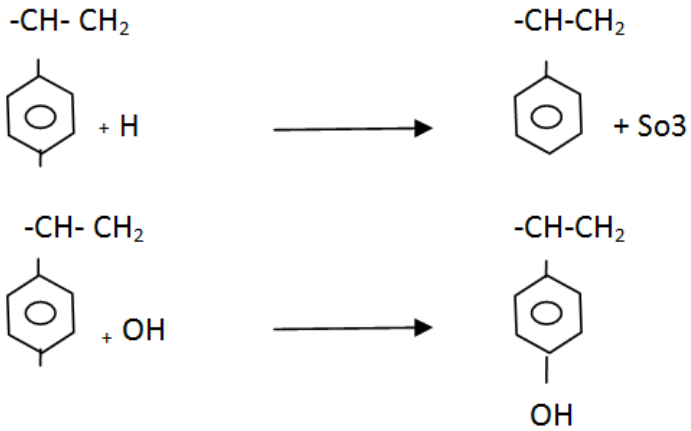
plotted in figure. The change in colour, capacity and weight loss are due to changes in skeleton of resin.

The kinetics of exchange of Zn, Pb, Mg and Co on irradiated (0.45×10^{-6} rads) and un irradiated Dowex 50 Wx_8 (NH_4^+) resin in aqueous acetone – ammonium butyrate (o.02M) was studied. The kinetic parameters such as half exchange time ($t^1/_2$) diffusion coefficient (D) and exchange rate constant (K) are presented in table no 2. The kinetic parameters were computed at o, 30, and 50 percentages of acetone.

The colour change and decrease in capacity of resin occur mainly as a result of some structural change in the skeleton of the resin such as cross linkage. Degradation and formation of new groups in Dowex 50 Wx8 may be explained as follow.



The H- atom also may bring about scission of So_3- group and the formation of weak acid exchange group takes place.



So_3 radicals may react with water molecule near the exchange group to form H- atoms and the resulting H- atoms can be in return induce further degradation of the resin. The loss of sulphonic acid group leads to capacity loss. Thus the loss in capacity can be determined either by measuring the residual capacity of the irradiated resin or by estimating the suphur contents in aqueous portion. The results in **figure (No 1)** were obtained by measuring the residual capacity.

$So_3^- + H_2O \longrightarrow HSo_4 + H$

The amount of degradation in terms of exchange capacity loss for Dowex 50 Wx_8 resins at a gamma ray dose of 0.4 x 10^{-6} rads/hr was found to be 15% for air dry resin in NH_4^+ form.

It is observed that the half exchange time (t1/2) increase on irradiation of the resin. It indicates that the exchange reaction becomes slower on irradiation. This effect is observed at all percentages of acetone. The half exchange time for all the metal ions, show the following order at 50% acetone for aqueous acetone ammonium butyrate media.

Pb > Zn > Mg > Co

It is observed that diffusion coefficient value decreased when the resin is exposed to gamma radiation for un irradiated and irradiated resins D decreases with increase in the acetone percentage for all metal ions. The change in chemical structure of the irradiated resin should account for the changes in diffusion properties on the resin. On exposure to ionizing radiation, macromolecular structures have been found undergoing changes such as bond breakage, bond formation, oxidation etc. breakage of C – S bonds might be resulting into splitting of ion exchange groups as indicated by decreased capacity and it is responsible for decrease in diffusion coefficient by irradiation. For all metal ion K decreases with rise in the acetone percentage at 296^o K.

CONCLUSION

The exchange reaction becomes slow due to irradiation and acetone content. The values of diffusion coefficients decrease on irradiation of the resin. Breaking of C – S bonds might be resulting into spitting of ion exchange groups and there is decrease in capacity and decrease in diffusion coefficient by irradiation.

APPLICATION

Ion exchange is often used for removing a certain ion form a solution or for replacing it by another ion. The quantitative separation of various metal ions can be effectively carried out at trace level also.

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