

Non Destructive Application of Radioactive Tracer Isotopes for Performance Evaluation of Industrial Grade Anion Exchange Resins Tulsion A-33 and Indion NSSR

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ABSTRACT

The present study deals with non-destructive application of radioactive tracer isotopes to evaluate the performance of Tulsion A-33 (nuclear grade) and Indion NSSR (non-nuclear grade) anion exchange resins. The performance evaluation was done by carrying out the iodide and bromide ion-isotopic exchange reactions using the above resins. It was observed that at a constant temperature of 40.0 °C, as the concentration of labeled iodide ion solution increases 0.001 M to 0.004 M, the percentage of iodide ions exchanged increases from 58.0 % to 64.0 % for Tulsion A-33 resins; and from 48.4 % to 50.8 % for Indion NSSR resins. Similarly in case of bromide ion-isotopic exchange reaction under identical experimental conditions, the percentage of bromide ions exchanged increases from 45.6 % to 50.4 % for Tulsion A-33 resin; and from 39.8 % to 44.6 % for Indion NSSR resin. It was also observed that during iodide ion-isotopic exchange reaction at 40.0 °C, using 1.000 g of ion exchange resins and 0.003 M labeled ionic solution, using Tulsion A-33 resin the values of specific reaction rate (min^{-1}), amount of iodide ion exchanged (mmol), initial rate of iodide ion exchange (mmol/min) and $\log K_d$ were 0.229, 0.469, 0.107 and 10.6 respectively, which was higher than the values of 0.167, 0.375, 0.063 and 7.6 respectively as obtained by using Indion NSSR resins under identical experimental conditions. The overall results indicate superior performance of Tulsion A-33 over Indion NSSR resin under identical operational parameters.

Keywords: radioisotopes; non-destructive technique; Tulsion A-33; Indion NSSR; nuclear grade resin; anion exchange resins; performance evaluation; operational parameters; ^{131}I ; ^{82}Br

1. INTRODUCTION

The availability of a wide spectrum of radioisotopes from the reactors and cyclotrons led to the quest for their beneficial applications in diverse fields such as medicine, industry and agriculture. Though radioisotopes have been applied to the solution of problems in industry for over 50 years, research and development of the technology continues unabated. Because of their unique properties, radioactive isotopes can be used to obtain information about plants and processes that cannot be obtained in any other way. It is possible to apply the radiotracers in order to investigate the efficiency of several devices in a wastewater treatment plant (primary and secondary clarifiers, aeration tank) [1]. Often, the information is

obtained with the plant on-stream and without disrupting the process in any way. By monitoring the radioactivity both continuously or after sampling (depending on the nature of study), the movement, adsorption, retention etc. of the tracer and in turn, of the bulk matter under investigation, can be followed. This can lead to substantial economic benefits, from shutdown avoidance to process optimization. Application of radioisotopes in certain manufacturing processes to induce desired chemical reactions is also reported in the literature [2]. As a result radioisotopes have become useful tool in various industrial applications [2]. Unlike many other advanced technologies, isotope applications have a very low gestation period. A technology developed in the laboratory is often successfully applied in the field within a very short time span.

This make isotope technology one of the highly visible peaceful uses of nuclear energy and a major reason for continuing with nuclear research. Radiotracer methodology is described extensively in the literature [3-5]. Generally in most of these applications the radioisotopes preferred are gamma emitters having half-life compatible with the duration of studies; also the strength of radioactivity used varies depending on the nature of application. Considering the widespread non-destructive application of radioactive isotopes, in the present investigation attempts are made to apply them for performance evaluation of industrial grade ion exchange resins Tulsion A-33 and Indion NSSR under different experimental conditions.

2. EXPERIMENTAL

2. 1. Conditioning of ion exchange resins

Tulsion A-33 a nuclear grade resin in hydroxide form (by Thermax India Ltd., Pune, India), and Indion NSSR a non-nuclear grade resin in chloride form (by Ion Exchange India Ltd., Mumbai, India) are strongly basic anion exchange resin having quaternary ammonium $N^+(CH_3)_3$ functional group. Details regarding the properties of the resins used are given in Table 1. These resins were converted separately in to iodide / bromide form by treatment with 10 % KI / KBr solution in a conditioning column which is adjusted at the flow rate as 1 mL / min. The resins were then washed with double distilled water, until the washings were free from iodide/bromide ions as tested by $AgNO_3$ solution. These resins in bromide and iodide form were then dried separately over P_2O_5 in desiccators at room temperature.

Table 1. Properties of ion exchange resins.

Ion exchange resin	Matrix	Particle Size (mm)	Moisture content (%)	Operating pH	Maximum operating temperature (°C)	Total exchange capacity (meq./mL)
Tulsion A-33	Polystyrene copolymer	0.3-1.2	60	0-14	60	1.00
Indion NSSR	Polystyrene copolymer	0.3-1.2	50	0-14	100	0.9

2. 2. Radioactive Tracer Isotopes

Details regarding the isotopes used in the present experimental work are given in Table 2.

Table 2. Properties of ^{131}I and ^{82}Br tracer isotopes [6].

Isotopes	Half-life	Radioactivity / mCi	γ - energy / MeV	Chemical form	Physical form
131I	8.04 d	5	0.36	Iodide*	Aqueous
82Br	36 h	5	0.55	Bromide**	Aqueous

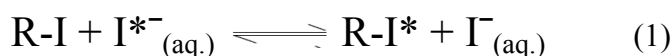
* Sodium iodide in dilute sodium sulphite.

** Ammonium bromide in dilute ammonium hydroxide.

2. 3. Study on kinetics of iodide ion-isotopic exchange reaction

In a stoppered bottle 250 mL (V) of 0.001 M iodide ion solution was labeled with diluted ^{131}I radioactive solution using a micro syringe, such that 1.0 mL of labeled solution has a radioactivity of around 15,000 cpm (counts per minute) when measured with γ -ray spectrometer having NaI (Tl) scintillation detector. Since only about 50-100 μL of the radioactive iodide ion solution was required for labeling the solution, its concentration will remain unchanged, which was further confirmed by potentiometer titration against AgNO_3 solution.

The above labeled solution of known initial activity (A_i) was kept in a thermostat adjusted to 30.0 $^\circ\text{C}$. The swelled and conditioned dry ion exchange resins in iodide form weighing exactly 1.000 g (m) were transferred quickly into this labeled solution which was vigorously stirred by using mechanical stirrer and the activity in cpm of 1.0 mL of solution was measured. The solution was transferred back to the same bottle containing labeled solution after measuring activity. The iodide ion-isotopic exchange reaction can be represented as:



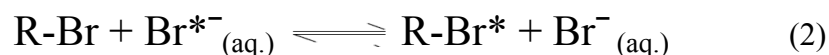
here R-I represents ion exchange resin in iodide form; $\text{I}^*_{(\text{aq})}$ represents aqueous iodide ion solution labeled with ^{131}I radiotracer isotope [17-19].

The activity of solution was measured at a fixed interval of every 2.0 min. The final activity (A_f) of the solution was also measured after 3h which was sufficient time to attain the equilibrium [7-12]. The activity measured at various time intervals was corrected for background counts.

Similar experiments were carried out by equilibrating separately 1.000 g of ion exchange resin in iodide form with labeled iodide ion solution of four different concentrations ranging up to 0.004 M at a constant temperature of 30.0 $^\circ\text{C}$. The experimental sets were also repeated for higher temperatures up to 45.0 $^\circ\text{C}$.

2. 4. Study on kinetics of bromide ion-isotopic exchange reaction

The experiment was also performed to study the kinetics of bromide ion- isotopic exchange reaction by equilibrating 1.000 g of ion exchange resin in bromide form with labeled bromide ion solution in the same concentration and temperature range as above. The labeling of bromide ion solution was done by using ^{82}Br as a radioactive tracer isotope for which the same procedure as explained above was followed. The bromide ion-isotopic exchange reaction can be represented as:



here R-Br represents ion exchange resin in bromide form; $\text{Br}^{*-}(\text{aq.})$ represents aqueous bromide ion solution labeled with ^{82}Br radiotracer isotope [17-19].

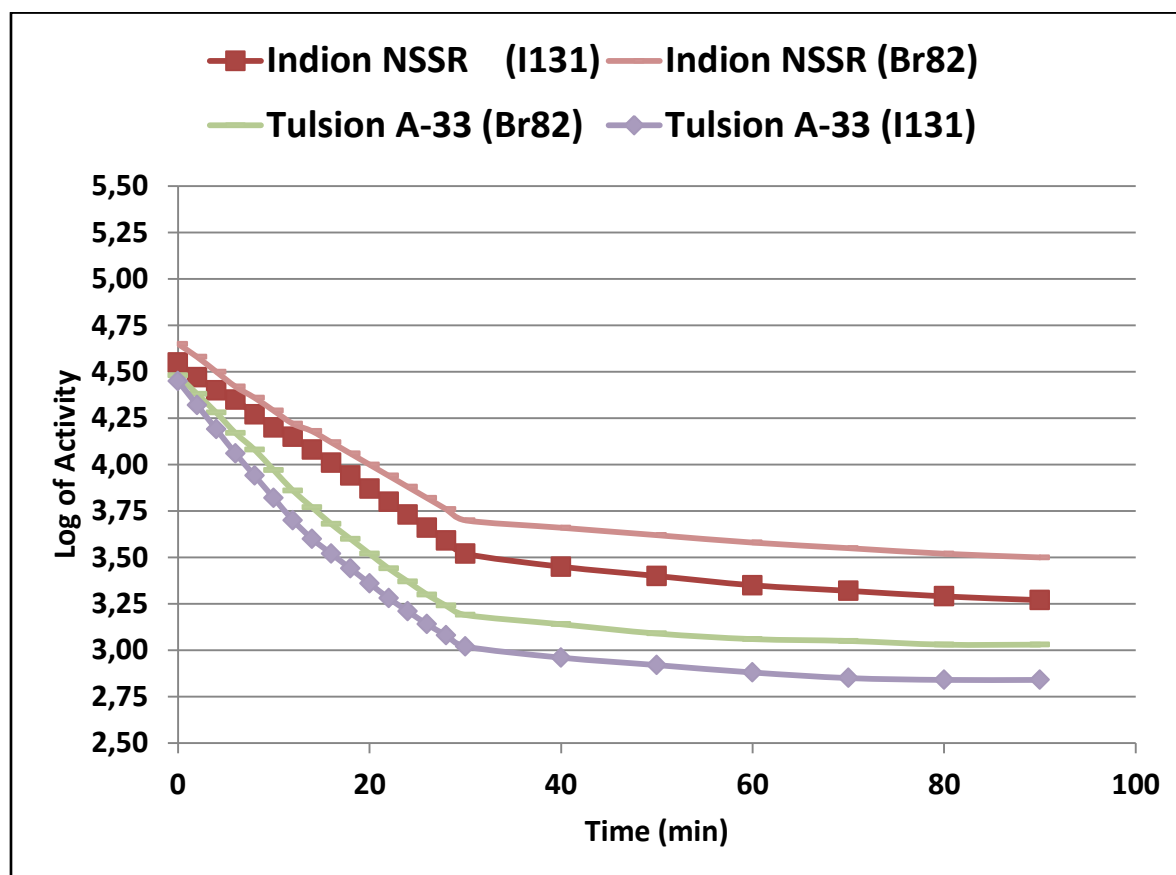


Figure 1. Kinetics of Ion-Isotopic Exchange Reactions.

Amount of ion exchange resin = 1.000 g,
 Concentration of labeled exchangeable ionic solution = 0.003M,
 Volume of labeled ionic solution = 250 mL,
 Temperature = 40.0 °C

Table 3. Concentration effect on Ion-Isotopic Exchange Reactions.

Amount of ion exchange resin = 1.000 g
 Volume of labeled ionic solution = 250 mL
 Temperature = 40.0 °C

Concentration of ionic solution (M)	Amount of ions in 250 mL solution (mmol)	REACTION -1				REACTION -2					
0.001	0.250	Tulsion A-33	Specific reaction rate of rapid process min ⁻¹	Amount of iodide ion exchanged (mmol)	Initial rate of iodide ion exchange (mmol/min)	Log K _d	Tulsion A-33	Specific reaction rate of rapid process min ⁻¹	Amount of bromide ion exchanged (mmol)	Initial rate of bromide ion exchange (mmol/min)	Log K _d
0.002	0.500		0.208	0.145	0.030	8.4		0.170	0.114	0.019	7.5
0.003	0.750		0.229	0.469	0.107	10.6		0.179	0.370	0.066	8.6
0.004	1.000		0.240	0.640	0.154	11.2		0.185	0.504	0.093	9.3
		Indion NSSR	Specific reaction rate of rapid process min ⁻¹	Amount of iodide ion exchanged (mmol)	Initial rate of iodide ion exchange (mmol/min)	Log K _d	Indion NSSR	Specific reaction rate of rapid process min ⁻¹	Amount of bromide ion exchanged (mmol)	Initial rate of bromide ion exchange (mmol/min)	Log K _d
0.001	0.250		0.143	0.121	0.017	6.5		0.120	0.100	0.012	3.1
0.002	0.500		0.151	0.246	0.037	7.0		0.127	0.208	0.026	3.8
0.003	0.750		0.167	0.375	0.063	7.6		0.135	0.322	0.043	4.3
0.004	1.000	0.179	0.508	0.091	8.0	0.141	0.446	0.063	4.8		

Table 4. Temperature effect on Ion-Isotopic Exchange Reactions.
 Amount of ion exchange resin = 1.000 g
 Concentration of labeled exchangeable ionic solution = 0.003M
 Volume of labeled ionic solution = 250 mL
 Amount of exchangeable ions in 250 mL labeled solution = 0.750 mmol

Temperature °C			
45.0	40.0	35.0	30.0
0.218	0.229	0.235	0.250
0.452	0.469	0.473	0.482
0.099	0.107	0.111	0.121
10.0	10.6	11.1	11.6
0.155	0.167	0.176	0.187
0.372	0.375	0.382	0.388
0.058	0.063	0.067	0.073
7.1	7.6	8.3	8.8
0.173	0.179	0.183	0.187
0.352	0.370	0.381	0.392
0.061	0.066	0.070	0.073
8.1	8.6	9.1	9.8
0.130	0.135	0.143	0.148
0.306	0.322	0.336	0.352
0.040	0.043	0.048	0.052
3.9	4.3	4.8	5.5

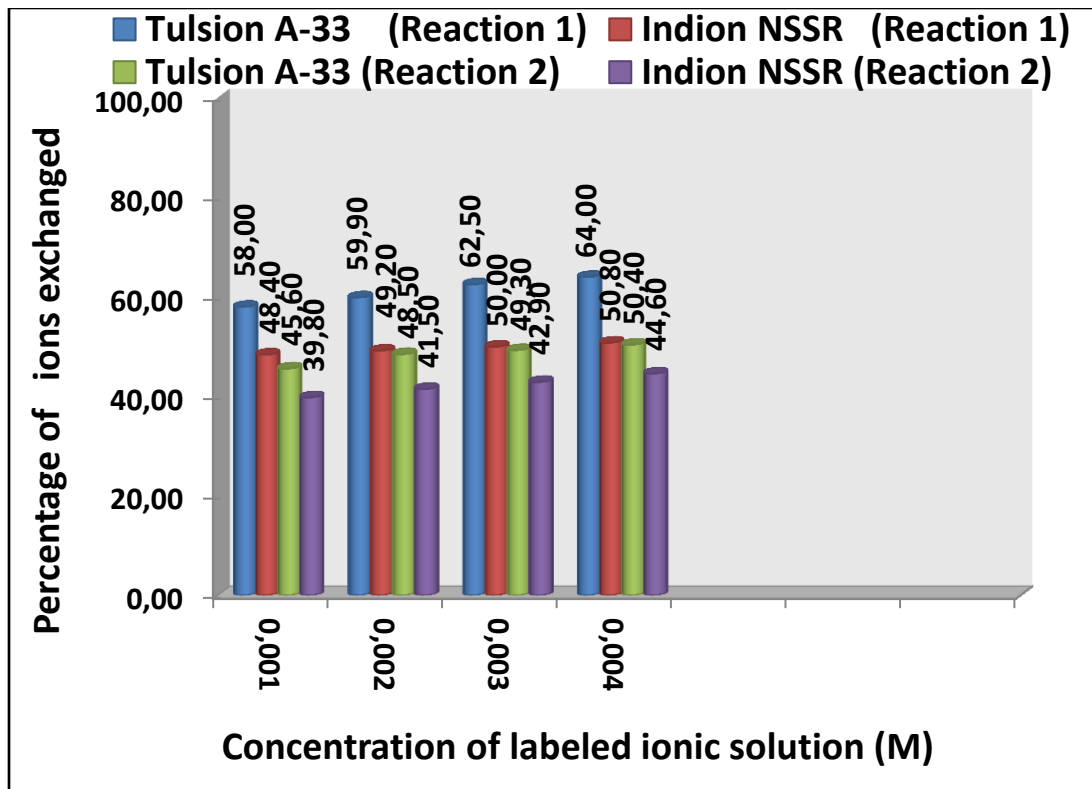


Figure 2. Variation in Percentage Ions Exchanged with Concentration of Labeled Ionic Solution. Amount of ion exchange resin = 1.000 g, Volume of labeled ionic solution = 250 mL, Temperature = 40.0 °C.

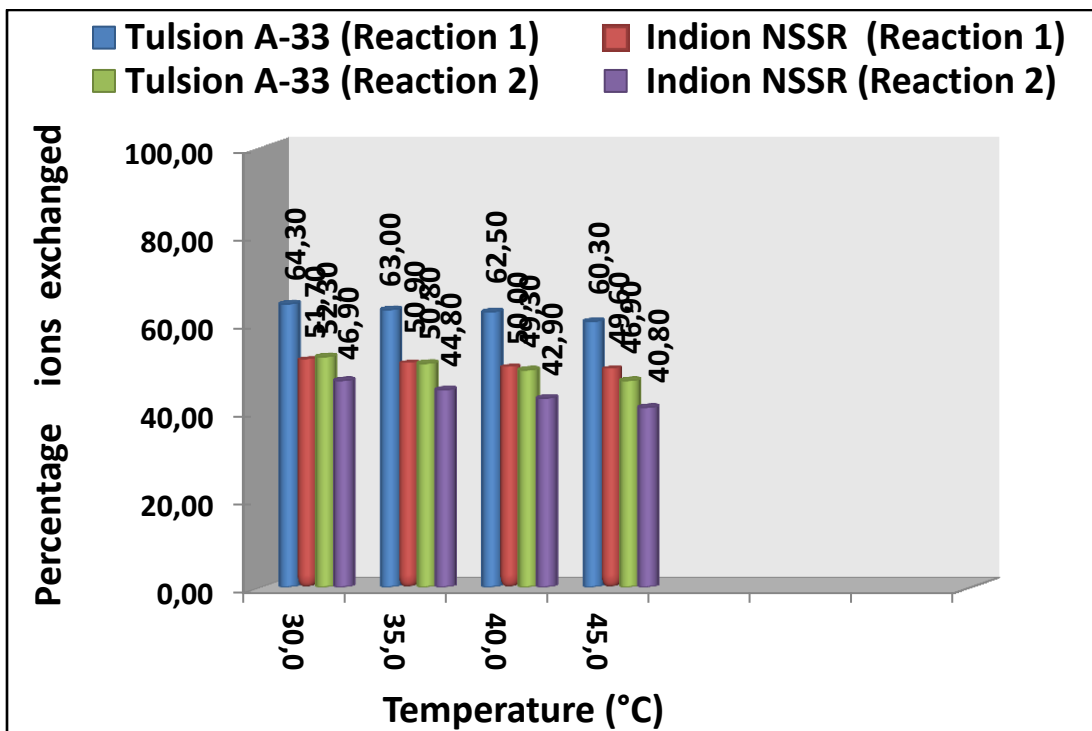


Figure 3. Variation in Percentage Ions Exchanged with Temperature of Labeled Ionic Solution. Amount of ion exchange resin = 1.000 g, Concentration of labeled exchangeable ionic solution = 0.003 M, Volume of labeled ionic solution = 250 mL, Amount of exchangeable ions in 250 mL labeled solution = 0.750 mmol

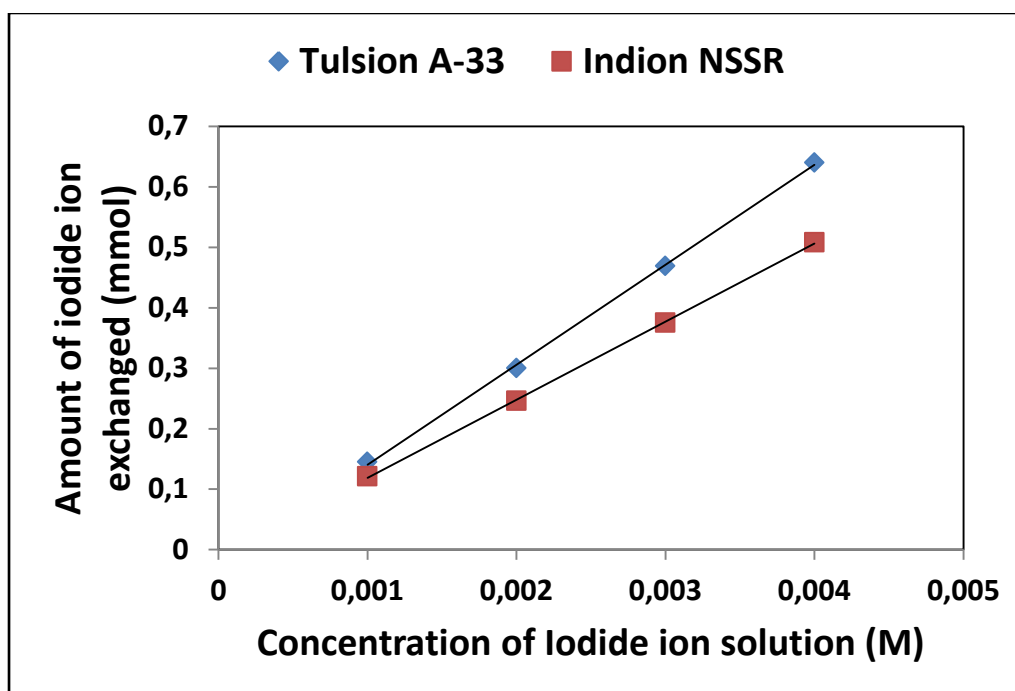


Figure 4. Correlation between concentration of iodide ion solution and amount of iodide ion exchanged.

Amount of ion exchange resin = 1.000 g, Volume of labeled ionic solution = 250 mL, Temperature = 40.0 °C
Correlation coefficient (r) for Tulsion A-33 = 0.9997, Correlation coefficient (r) for Indion NSSR = 0.9999

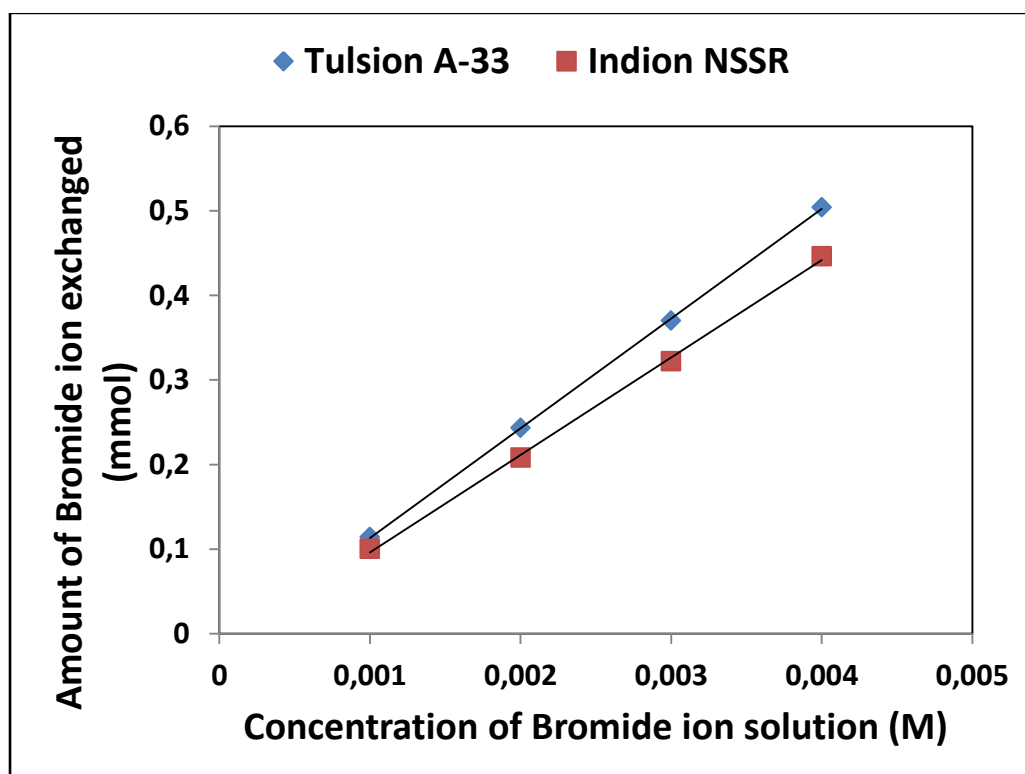


Figure 5. Correlation between concentration of bromide ion solution and amount of bromide ion exchanged.

Amount of ion exchange resin = 1.000 g, Volume of labeled ionic solution = 250 mL, Temperature = 40.0 °C
Correlation coefficient (r) for Tulsion A-33 = 0.9999, Correlation coefficient (r) for Indion NSSR = 0.9995

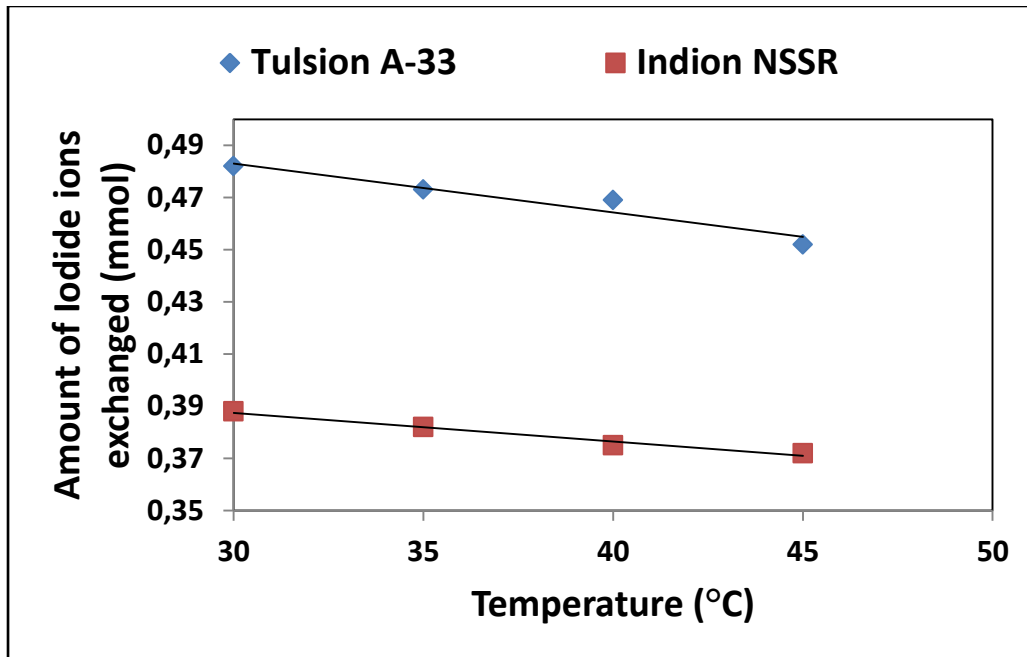


Figure 6. Correlation between Temperature of exchanging medium and amount of iodide ion exchanged.

Amount of ion exchange resin = 1.000 g, Concentration of labeled exchangeable ionic solution = 0.003M, Volume of labeled ionic solution = 250 mL, Amount of exchangeable ions in 250 mL labeled solution = 0.750 mmol, Correlation coefficient (r) for Tulsion A-33 = -0.9654, Correlation coefficient (r) for Indion NSSR = -0.9886

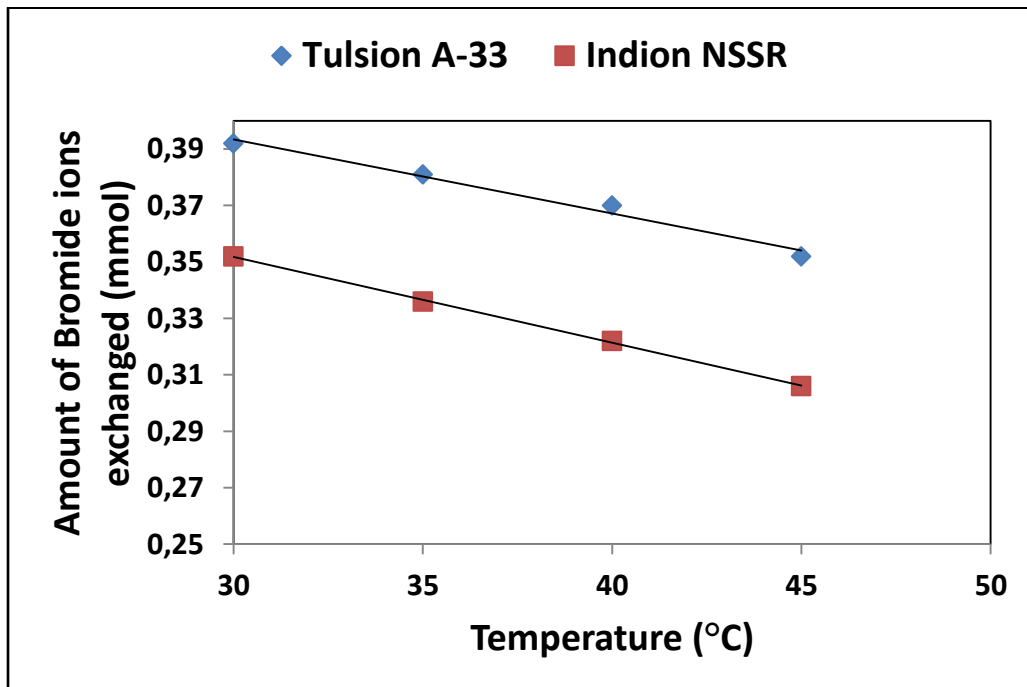


Figure 7. Correlation between Temperature of exchanging medium and amount of bromide ion exchanged.

Amount of ion exchange resin = 1.000 g, Concentration of labeled exchangeable ionic solution = 0.003 M, Volume of labeled ionic solution = 250 mL, Amount of exchangeable ions in 250 mL labeled solution = 0.750 mmol +9, Correlation coefficient (r) for Tulsion A-33 = -0.9915, Correlation coefficient (r) for Indion NSSR = -0.9997

3. RESULTS AND DISCUSSION

3.1. Comparative study of ion-isotopic exchange reactions

In the present investigation it was observed that due to the rapid ion-isotopic exchange reaction taking place, the activity of solution decreases rapidly initially, then due to the slow exchange the activity of the solution decreases slowly and finally remains nearly constant. Preliminary studies show that the above exchange reactions are of first order [7-12]. Therefore logarithm of activity when plotted against time gives a composite curve in which the activity initially decreases sharply and thereafter very slowly giving nearly straight line (Figure 1), evidently rapid and slow ion-isotopic exchange reactions were occurring simultaneously [7-12]. Now the straight line was extrapolated back to zero time. The extrapolated portion represents the contribution of slow process to the total activity which now includes rapid process also. The activity due to slow process was subtracted from the total activity at various time intervals. The difference gives the activity due to rapid process only. From the activity exchanged due to rapid process at various time intervals, the specific reaction rates (k) of rapid ion-isotopic exchange reaction were calculated. The amount of iodide / bromide ions exchanged (mmol) on the resin were obtained from the initial and final activity of solution and the amount of exchangeable ions in 250 mL of solution. From the amount of ions exchanged on the resin (mmol) and the specific reaction rates (min^{-1}), the initial rate of ion exchanged (mmol/min) was calculated.

Because of larger solvated size of bromide ions as compared to that of iodide ions, it was observed that the exchange of bromide ions occurs at slower rate than that of iodide ions. Hence under identical experimental conditions, the values of specific reaction rate (min^{-1}), amount of ion exchanged (mmol) and initial rate of ion exchange (mmol/min) are calculated to be lower for bromide ion-isotopic exchange reaction than that for iodide ion-isotopic exchange reaction as summarized in Tables 3 and 4. For both bromide and iodide ion-isotopic exchange reactions, under identical experimental conditions, while temperature remaining constant at 40.0 °C, the values of specific reaction rate increases with increase in ionic concentration from 0.001M to 0.004M (Table 3). However, at constant ionic concentration of 0.003 M, the specific reaction rate was observed to decrease with rise in temperature from 30.0 °C to 45.0 °C, (Table 4).

Thus in case of Tulsion A-33 at 40.0 °C when the ionic concentration increases from 0.001M to 0.004M, the specific reaction rate values for iodide ion-isotopic exchange increases from 0.208 to 0.240 min^{-1} , while for bromide ion-isotopic exchange the values increases from 0.170 to 0.185 min^{-1} . Similarly in case of Indion NSSR, under identical experimental conditions, the values for iodide ion-isotopic exchange increases from 0.143 to 0.179 min^{-1} , while for bromide ion-isotopic exchange the values increases from 0.120 to 0.141 min^{-1} . However when concentration of ionic solution was kept constant at 0.003 M and temperature is raised from 30.0 °C to 45.0 °C, in case of Tulsion A-33 the specific reaction rate values for iodide ion-isotopic exchange decreases from 0.250 to 0.218 min^{-1} , while for bromide ion-isotopic exchange the values decreases from 0.187 to 0.173 min^{-1} . Similarly in case of Indion NSSR, under identical experimental conditions, the specific reaction rate values for iodide ion-isotopic exchange decreases from 0.187 to 0.155 min^{-1} , while for bromide ion-isotopic exchange the values decreases from 0.148 to 0.130 min^{-1} . From the results, it appears that iodide ions exchange at the faster rate as compared to that of bromide ions which was related to the extent of solvation (Tables 3 and 4).

From the knowledge of A_i , A_f , volume of the exchangeable ionic solution (V) and mass of ion exchange resin (m), the K_d value was calculated by the equation

$$K_d = [(A_i - A_f) / A_f] \times V / m \quad (3)$$

Previous studies [13,14] on halide ion distribution coefficient on strong and weak basic anion exchange resins indicate that the selectivity coefficient between halide ions increased at higher electrolyte concentrations. Adachi *et al.* [15] observed that the swelling pressure of the resin decreased at higher solute concentrations resulting in larger K_d values. The temperature dependence of K_d values on cation exchange resin was studied by Shuji *et al.* [16]; were they observed that the values of K_d increased with fall in temperature. The present experimental results also indicates that the K_d values for bromide and iodide ions increases with increase in concentration of the external ionic solution, however with rise in temperature the K_d values were found to decrease. Thus in case of Tulsion A-33 at 40.0 °C when the ionic concentration increases from 0.001M to 0.004M, the log K_d values for iodide ions increases from 8.4 to 11.2, while for bromide ions the values increases from 7.5 to 9.3. Similarly in case of Indion NSSR, under identical experimental conditions, the log K_d values for iodide ions increases from 6.5 to 8.0, while for bromide ions the values increases from 3.1 to 4.8. However when concentration of ionic solution was kept constant at 0.003 M and temperature is raised from 30.0 °C to 45.0 °C, in case of Tulsion A-33 the log K_d values for iodide ions decreases from 11.6 to 10.0, while for bromide ions the values decreases from 9.8 to 8.1. Similarly in case of Indion NSSR, under identical experimental conditions, the log K_d values for iodide ions decreases from 8.8 to 7.1, while for bromide ions the values decreases from 5.5 to 3.9. It was also observed that the K_d values for iodide ion-isotopic exchange reaction were calculated to be higher than that for bromide ion-isotopic exchange reaction (Tables 3 and 4).

3.2 Comparative study of anion exchange resins From the Table 3 and 4, it is observed that for iodide ion-isotopic exchange reaction by using Tulsion A-33 resin, the values of specific reaction rate (min^{-1}), amount of iodide ion exchanged (mmol), initial rate of iodide ion exchange (mmol/min) and log K_d were 0.229, 0.469, 0.107 and 10.6 respectively, which was higher than 0.167, 0.375, 0.063 and 7.6 respectively as that obtained by using Indion NSSR resins under identical experimental conditions of 40.0 °C, 1.000 g of ion exchange resins and 0.003 M labeled iodide ion solution. The identical trend was observed for the two resins during bromide ion-isotopic exchange reaction.

From Table 3, it is observed that at a constant temperature of 40.0 °C, as the concentration of labeled iodide ion solution increases 0.001 M to 0.004 M, the percentage of iodide ions exchanged increases from 58.0 % to 64.0 % for Tulsion A-33 resins; and from 48.4 % to 50.8 % for Indion NSSR resins. Similarly in case of bromide ion-isotopic exchange reaction, the percentage of bromide ions exchanged increases from 45.6 % to 50.4 % for Tulsion A-33 resin; and from 39.8 % to 44.6 % for Indion NSSR resin. The effect of ionic concentration on percentage of ions exchanged is graphically represented in Figure 2.

From Table 4, it is observed that using Tulsion A-33 resins, for 0.003 M labeled iodide ion solution, as the temperature increases 30.0 °C to 45.0 °C, the percentage of iodide ions exchanged decreases from 64.3 % to 60.3 %. While using Indion NSSR resins under identical experimental conditions the percentage of iodide ions exchanged decreases from 51.7 % to 49.6 %. Similarly in case of bromide ion-isotopic exchange reaction, the percentage of bromide ions exchanged decreases from 52.3 % to 46.9 % using Tulsion A-33 resin, while for Indion NSSR resin it decreases from 46.9 % to 40.8 %. The effect of temperature on percentage of ions exchanged is graphically represented in Figure 3.

The overall results indicate that under identical experimental conditions, as compared to Indion NSSR resins, Tulsion A-33 resins shows higher percentage of ions exchanged. Thus

Tulsion A-33 resins show superior performance than Indion NSSR resins under identical operational parameters.

3. 3. Statistical Correlations

The results of present investigation show a strong positive linear co-relationship between amount of ions exchanged and concentration of ionic solution (Figures 4, 5). In case of iodide ion-isotopic exchange reaction, the value of correlation coefficient (r) was calculated as 0.9997 and 0.9999 respectively for Tulsion A-33 and Indion NSSR resins, while for bromide ion-isotopic exchange reaction, the respective values of r calculated was 0.9999 and 0.9995.

There also exist a strong negative co-relationship between amount of ions exchanged and temperature of exchanging medium (Figures 6, 7). In case of iodide ion-isotopic exchange reactions the values of r was calculated as -0.9654 and -0.9886 respectively. Similarly in case of bromide ion-isotopic exchange reactions the respective values of r calculated where -0.9915 and -0.9997 for both the resins.

4. CONCLUSION

The experimental work carried out in the present investigation will help to standardize the operational process parameters so as to improve the performance of selected nuclear grade ion exchange resins. The radioactive tracer technique used here can also be applied for characterization of different nuclear as well as non-nuclear grade ion exchange resins.

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