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# Comparative Study of Indion-AGR and Duolite A-638 Anion Exchange Resins by Application of <sup>131</sup>I and <sup>82</sup> Br as a Tracer Isotopes

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**Abstract** The present study deals with modeling of ion-isotopic exchange reaction kinetics by application of radioactive tracer isotopes 82Br and 131I. The bromide and iodide ion-isotopic exchange reactions was carried out by using weak base anion exchange resins Indion-AGR and Duolite A-638. It was observed that for both the resins, reaction rate decreases with rise in temperature and increases with increase in ionic concentration. The study was extended further for characterization of these resins based on their performance under different operational parameters. It was observed that the percentage/amount of ions exchanged and distribution coefficient values calculated for Duolite A-638 was higher than Indion-AGR resins under identical operational parameters, indicating superior performance of Duolite A-638 over Indion-AGR resins.

**Keywords** Indion-AGR, Duolite A-638, Tracer Applications, Reaction Kinetics, Ion-Isotopic Exchange Reactions, <sup>131</sup>I, <sup>82</sup>Br, Distribution Coefficient, Reaction Rate

### 1. Introduction

Radio isotopes find applications in several fields of which industrial applications constitute a major portion with respect to the quantum of activity used and the economic benefits accrued. Industrial applications of radioisotopes can be mainly categorized into two. The first one being the use of radiation from sealed sources of radioisotopes or from electron beam accelerators for industrial processing, non-destructive testing. The second major group of applications is the use of radiotracers in inventory control, study of process parameters, trouble shooting in industrial systems, flow measurements, leakage studies etc. The economic benefits that may be derived from the use of the radioisotope technology are great, a fact that is recognized by the governments of developing countries. Though radioisotopes have been applied to the solution of problems in industry for over 50 years, research and development of the technology continues unabated. There are two main reasons for the continuing interest. Firstly, it is industry driven. 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. Often, the information is obtained with the plant on-stream and

The fundamental principle in radiochemical investigations is that the chemical properties of a radio isotope of an element are almost the same as those of the other stable/radioactive isotopes of the element. When radioisotope is present in a chemical form identical to that of the bulk of the element in a chemical process, then any reaction the element undergoes can be directly traced by monitoring the radioisotope. Radiochemical work involves two main steps first is the sampling of chemical species to be studied and second is quantitative determination of the radiation emitted by the radioisotope in the sample[1]. In radiotracer study, a short lived radioisotope in a physico-chemical form similar to that of the process material is used to trace the material under study. The radioisotopes in suitable physical and chemical forms are introduced in systems under study. 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. The tracer concentration recorded at various locations also helps to draw information about the dynamic behavior of the system under study. The radioisotopes preferred for such studies are gamma emitters having half-life compatible with the

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without disrupting the process in any way. This can lead to substantial economic benefits, from shutdown avoidance to process optimization. Secondly, the methodology is derived from many fields of science and technology including radioisotope production, radiation detection, data acquisition, treatment and analysis, and mathematical modeling.

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duration of studies. The strength of radioactivity used varies depending on the nature of application. Applications of radiotracers in chemical research cover the studies of reaction mechanism, kinetics, exchange processes and analytical applications such as radiometric titrations, solubility product estimation, isotope dilution analysis and autoradiogrphy. Radioisotope tracers offer advantages such as high detection sensitivity, capability of detection, limited effects in-situ memory physic-chemical compatibility with the material under study. The radioisotopes have proved as a tool to study many problems in chemical, biological and medicinal fields. Radiotracers have helped in identification of leaks in buried pipelines and dams. Process parameters such as mixing efficiency, recidence time, flow rate, material inventory and movement in harbours are studied radioisotopes[1]. The efficiency of several devices in a wastewater treatment plant (primary and secondary clarifiers, aeration tank) is investigated by means of radiotracers[2].

Considering the above wide use of radioactive isotopes in various industrial and technical applications, in the present investigation, they are applied to assess the performance of industrial grade anion exchange resins Indion-AGR and Duolite A-638 under different operational parameters like temperature and ionic concentrations. It is expected that the tracer technique used here can also be used for characterization of other organic ion exchange resins which are synthesized for their specific technical applications [3-8]. The present technique can also be extended further to standardize the operational parameters so as to bring about the most efficient performance of those resins in their specific industrial applications.

## 2. Experimental

#### 2.1. Conditioning of Ion Exchange Resins

Ion exchange resin Indion-A GR (by Ion Exchange India Ltd., Mumbai) and Duolite A-638 (by Auchtel Product Ltd., Mumbai) are weakly basic anion exchange resin in chloride form having tertiary ammonium 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<sub>3</sub> solution. These resins in bromide and iodide form were then dried separately over P<sub>2</sub>O<sub>5</sub> in desiccators at room temperature.

#### 2.2. Radioactive Tracer Isotopes

The radioisotope 131I and 82Br used in the present experimental work was obtained from Board of Radiation and Isotope Technology (BRIT), Mumbai. Details regarding the isotopes used in the present experimental work are given

in Table 2.

**Table 1.** Properties of ion exchange resins

Ion exchange resin	Matrix	Moisture content (%)	Maximum operating temperature (°C)	Total exchange capacity (meq./mL)		
Indion-AGR	Crosslinked Polystyrene	51	60	1.3		
Duolite A-638	Crosslinked Polystyrene	<2.0	60	4.0		

**Table 2.** Properties of <sup>131</sup>I and <sup>82</sup>Br tracer isotopes[9]

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 <sup>131</sup>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  $\gamma$  -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<sub>3</sub> solution. The above labeled solution of known initial activity  $(A_i)$  was kept in a thermostat adjusted to  $30.0^{\circ}$ 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:

$$R-I + I^*_{(aq.)} = R-I^* + I_{(aq.)}$$
 (1)

Here R-I represents ion exchange resin in iodide form;  $I^{*}_{(aq.)}$  represents aqueous iodide ion solution labeled with  $^{131}I$  radiotracer isotope.

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[10-24]. 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  $30.0^{\circ}C$ . The same experimental sets were repeated for higher

temperatures up to 45.0°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 82Br 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:

$$R-Br + Br^*_{(aq.)} = R-Br^* + Br_{(aq.)}$$
 (2)

Here R-Br represents ion exchange resin in bromide form; Br\*-(aq.) represents aqueous bromide ion solution labeled with 82Br radiotracer isotope.

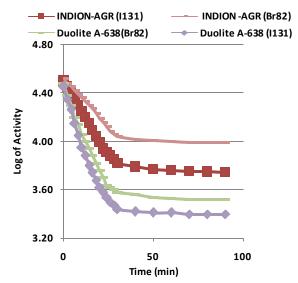
#### 3. Results and Discussion

## 3.1. Comparative Study of Ion-Isotopic Exchange Reactions

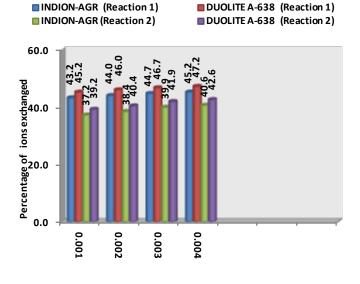
In the present investigation it was observed that due to the rapid ion-isotopic exchange reaction taking place, the activity of solution decreases rapidly initially, then due to the slow exchange the activity of the solution decreases slowly and finally remains nearly constant. Preliminary studies show that the above exchange reactions are of first order[25, 26]. 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[10-24]. 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<sup>-1</sup>), the initial rate of ion exchanged (mmol/min) was calculated.

Because of larger solvated size of bromide ions (310 pm) as compared to that of iodide ions (300 pm), it was observed that the exchange of bromide ions occurs at the slower rate than that of iodide ions[27]. Hence under identical experimental conditions, the values of specific reaction rate (min<sup>-1</sup>), 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, the values of specific reaction rate increases with increase in concentration of ionic solution from 0.001M to 0.004M (Table 3). However, with rise in temperature from 30.0°C to 45.0°C, the specific reaction rate was observed to decrease (Table 4). 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).

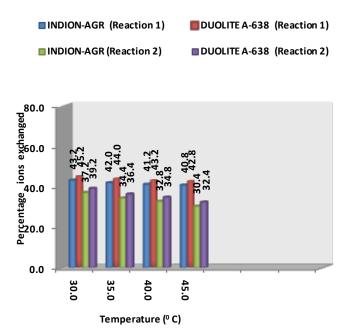


**Figure 1.** Kinetics of Ion-Isotopic Exchange Reactions Amount of ion exchange resin = 1.000 g, Concentration of labeled exchangeable ionic solution = 0.001M, Volume of labeled ionic solution = 250 mL, Temperature =  $30.0^{\circ}$ C

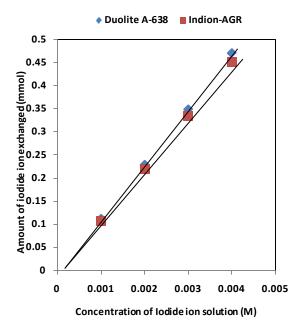


**Figure 2.** Variation in Percentage Ions Exchanged with Concentration of Labeled Ionic Solution Amount of ion exchange resin = 1.000 g, Volume of labeled ionic solution = 250 mL, Temperature = 30.0 ℃

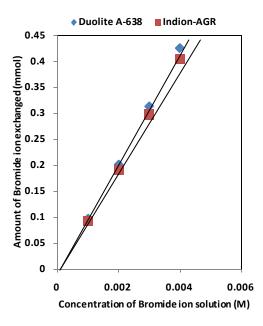
Concentration of labeled ionic solution (M)



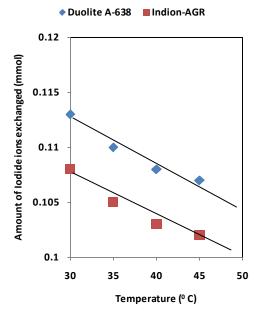
**Figure 3.** Variation in Percentage Ions Exchanged with Temperature of Labeled Ionic Solution Amount of ion exchange resin = 1.000 g, Concentration of labeled exchangeable ionic solution = 0.001M, Volume of labeled ionic solution = 250 mL, Amount of exchangeable ions in 250 mL labeled solution = 0.250 mmol



**Figure 4.** Correlation between concentrations 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 =  $30.0^{\circ}$ C Correlation coefficient (r) for Indion-AGR = 1.0000 Correlation coefficient (r) for Duolite A-638 = 1.0000



**Figure 5.** Correlation between concentrations 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 = 30.0°C Correlation coefficient (r) for Indion-AGR = 0.9998 Correlation coefficient (r) for Duolite A-638 = 0.9998



**Figure 6.** Correlation between Temperatures of exchanging medium and amount of iodide ion exchanged Amount of ion exchange resin = 1.000 g, Concentration of labeled exchangeable ionic solution = 0.001M, Volume of labeled ionic solution = 250 mL, Amount of exchangeable ions in 250 mL labeled solution = 0.250 mmol Correlation coefficient (r) for Indion-AGR = -0.9759 Correlation coefficient (r) for Duolite A-638 = -0.9759

Table 3. Concentration effect on Ion-Isotopic Exchange Reactions

	DUOLITE A-638	Log K <sub>d</sub>		4.1	4.5	4.7
		Initial rate of bromide ion (mim/lomm)	0.010	0.022	0.038	0.056
		noi əbirmond to tranom A (Iomm) bəgnsrbəxə	0.098	0.202	0.314	0.426
REACTION -2		Specific reaction rate of rapid <sup>1</sup> -rim essong	0.099	0.110	0.121	0.132
EACT		Log Ka	3.0	3.8	4.1	4.3
R	INDIO N-AGR	Initial rate of bromide ion (mim/lomm)	800.0	0.020	0.034	0.050
		noi əbimord To InnomA (Iomm) bəgnahəxə	0.093	0.192	0.299	0.406
		Specific reaction rate of rapid <sup>1</sup>	0.091	0.104	0.112	0.123
	DUOLITE A-638	$ m Log~K_d$	6.7	8.7	67	8.1
		noi əbiboi fo ətar laitirl (mim\lomm) bəgarahəxə	0.014	0.030	0.051	0.072
		noi əbiboi to tnuomA (Iomm) bəgnsıbxə	0.113	0.230	0.350	0.472
ION -1		Specific reaction rate of rapid process	0.120	0.131	0.145	0.153
REACTION -	INDIO N-AGR	Log K <sub>d</sub>	6.1	9.9	7.1	9.7
I		Initial rate of iodide ion exchange (mmol/min)	0.011	0.025	0.041	0.061
		noi əbiboi to innomA (Iomm) bəgnadəxə	0.108	0.220	0.335	0.452
		Specific reaction rate of rapid <sup>1</sup> -mim seconq	0.103	0.112	0.123	0.134
	noitulos Jm 002 ni anoi 10 tnuom A (lomm)		0.250	0.500	0.750	1.000
noitulæ sinoi to noitsunænoO (M)			0.001	0.002	0.003	0.004

Amount of ion exchange resin =  $1.000~\mathrm{g}$ . Volume of labeled ionic solution =  $250~\mathrm{mL}$ , Temperature =  $30.0^\circ\mathrm{C}$ 

Table 4. Temperature effect on Ion-Isotopic Exchange Reactions

ION -2	DUOLITE A-638	Log K <sub>d</sub>	3.2	2.5	2.3	2.1	
		noi abimord to atta lainil exchange (mm/lomm)	0.010	800.0	0.007	900.0	
		noi əbimond to tanomA (lomm) bəgasdəxə	0.098	0.091	0.087	0.081	
		Specific reaction rate of rapid frim sesonin	0.099	0.090	0.085	0.079	50 mL.
REACTION -2	INDIO N-AGR	Log K <sub>d</sub>	3.0	2.3	1.9	1.7	tion = 2
		noi əbirmorlətə əsra faitirl (mim/lormm) əgrashəxə	0.008	0.007	900.0	0.005	centration of label ed exchangeable ionic solution $= 0.001$ Wolume of labeled ionic solution $= 250$ mJ.
		noi əbimord To InnomA (Iomm)bəgnshəxə	0.093	980'0	0.082	920.0	lume of Jahe
		Specific reaction rate of rapid <sup>1</sup>	160.0	980.0	0.078	0.070	0.001M Vc
	DOOLITE A-638	$ m Log~K_d$	6.7	6.1	5.4	4.8	= uoiti
REACTION -1		Initial rate of iodide ion (mim/lomm)	0.014	0.012	0.010	0.010	los sinoi e
		Amount of iodide ion (Iomm) bagashx.3	0.113	0.110	0.108	0.107	exchangeahl
		Specific reaction rate of rapid <sup>1</sup> -mim seconq	0.120	0.108	0.097	060.0	of Jaheled
	INDIO N-AGR	Log K <sub>d</sub>	6.1	5.5	4.6	4.1	itration
		noi əbiboi to ətan laitinl (nim/lomm) əgnadəxə	0.011	0.010	600.0	0.007	9 Concer
		noi əbiboi 10 innomA (lomm) bəgnshəxə	0.108	0.105	0.103	0.102	Amount of ion exchange resin = 1 000 g Cor
		Specific reaction rate of rapid <sup>1</sup>	0.103	0.091	0.084	0.071	fion exchange
		30.0	35.0	40.0	45.0	nount o	

Amount of ion exchange resin = 1.000 g. Concentration of labered exchanges Amount of exchangeable ions in 250 mL labeled solution = 0.250 mmol.

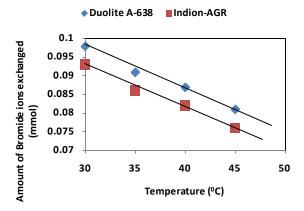


Figure 7. Correlation between Temperatures of exchanging medium and amount of bromide ion exchanged Amount of ion exchange resin = 1.000 g, Concentration of labeled exchangeable ionic solution = 0.001M, Volume of labeled ionic solution = 250 mL, Amount of exchangeable ions in 250 mL labeled solution = 0.250 mmol Correlation coefficient (r) for Indion-AGR = -0.9951 Correlation coefficient (r) for Duolite A-638 = -0.9951

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

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

Heumann et al. [28] in the study of chloride distribution coefficient on strongly basic anion exchange resin observed that the selectivity coefficient between halide ions increased at higher electrolyte concentrations. Adachi et al.[29] observed that the swelling pressure of the resin decreased at higher solute concentrations resulting in larger K<sub>d</sub> values. The temperature dependence of K<sub>d</sub> values on cation exchange resin was studied by Shuji et al.[30]; were they observed that the values of K<sub>d</sub> increased with fall in temperature. The present experimental results also indicates that the K<sub>d</sub> values for bromide and iodide ions increases with increase in ionic concentration of the external solution, however with rise in temperature the K<sub>d</sub> values were found to decrease. It was also observed that the K<sub>d</sub> values for iodide ion-isotopic reaction were calculated to be higher than that for bromide ion-isotopic reaction (Tables 3 and 4).

#### 3.2. Comparative Study of Anion Exchange Resins

From the Table 3, it is observed that for iodide ion-isotopic exchange reaction by using Duolite A-638 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 Kd were 0.120, 0.113, 0.014 and 6.7 respectively, which was higher than 0.103, 0.108, 0.011 and 6.1 respectively as that obtained by using Indion-AGR resins under identical experimental conditions of 30.00C, 1.000 g of ion exchange resins and 0.001 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 using Duolite A-638 resins, at a constant temperature of  $30.0 \,^{\circ}\text{C}$ , as the concentration of labeled iodide ion solution increases 0.001

M to 0.004 M, the percentage of iodide ions exchanged increases from 45.2 % to 47.2%. While using Indion-A GR resins under identical experimental conditions the percentage of iodide ions exchanged increases from 43.2 % to 45.2 %. Similarly in case of bromide ion-isotopic exchange reaction, the percentage of bromide ions exchanged increases from 39.2% to 42.6% using Duolite A-638 resin, while for Indion-A GR resin it increases from 37.2 % to 40.6 %. The effect of ionic concentration on percentage of ions exchanged is graphically represented in Figure 2.

From Table 4, it is observed that using Duolite A-638 resins, for 0.001 M labeled iodide ion solution, as the temperature increases 30.0°C to 45.0°C, the percentage of iodide ions exchanged decreases from 45.2 % to 42.8 %. While using Indion-A GR resins under identical experimental conditions the percentage of iodide ions exchanged decreases from 43.2 % to 40.8 %. Similarly in case of bromide ion-isotopic exchange reaction, the percentage of bromide ions exchanged decreases from 39.2 % to 32.4 % using Duolite A-638 resin, while for Indion-A GR resin it decreases from 37.2 % to 30.4 %. 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-AGR resins, Duolite A-638 resins shows higher percentage of ions exchanged. Thus Duolite A-638 resins show superior performance than Indion-AGR 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 using Duolite A-638 and Indion-A GR resins, the values of correlation coefficient (r) was calculated as 1.0000, while for bromide ion-isotopic exchange the values of r was calculated as 0.9998 for both the resins.

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 calculated for Duolite A-638 and Indion-A GR resins was -0.9759. Similarly in case of bromide ion-isotopic exchange reactions the r values calculated was -0.9951 for both the resins.

### 4. Conclusions

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 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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