Production of ^{128}I Atoms through ^{127}I (n,γ) ^{128}I Reactions in some Solid Iodocompounds Using an (^{241}Am-Be) Neutron Source

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Abstract

Present work reports the production of ^{128}I atoms through ^{127}I (n,γ) ^{128}I reactions in six iodine containing compounds (*viz.* NaI, NaIO$_3$, NaIO$_4$, KI, KIO$_3$ and KIO$_4$) in solid state. An ^{241}Am-Be neutron source has been used for the production of ^{128}I atoms. The radioactivities produced have been measured by a thin end-window G.M. Counter. The irradiation time and counting geometry were constant throughout the present sets of experiments. Results obtained have been analyzed through time-decay analyses. From the decay curves, the half life of ^{128}I has been estimated ~25 minutes. The average zero time activities of ^{128}I from three independent measurements in each of NaI, NaIO$_3$.H$_2$O, NaIO$_4$, KI, KIO$_3$ and KIO$_4$ target, have been found 116±2, 92±4, 88±6, 102±6, 98±1 and 93±5 cpm, respectively. It is interesting to note that the activities show a decreasing trend while passing through either from NaI to NaIO$_4$ or from KI to KIO$_4$. Results are attributed to the quantity of ^{128}I produced in a specified volume element and, self absorption and self scattering of radiations within the target compounds studied.

Keywords: Iodocompounds; ^{128}I; ^{241}Am-Be neutron source; Chemical effects of nuclear transformation.

1. Introduction

Iodine is one of the fission products (FPs) that might be produced in the core of light water reactors and is recognized as the most relevant to public health consequences in case of an accident with the environmental release of FPs [1]. There exist 37 well characterized isotopes of iodine and of these, only one, ^{127}I is stable [2]. The recoil study of hot iodine in iodocompounds is less perplexing because of its monoisotopic nature [3]. A high thermal neutron capture cross section (6.2±0.26 b)

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[4,5] of iodine and convenient half life (25 minutes) of ^{128}I enable researchers to study favorably the high energy iodine chemistry following ^{127}I (n,γ) ^{128}I reaction. Moreover, the characteristics of ^{128}I compound nucleus and its subsequent de-excitation through photon emission or electron conversion and the associated recoil spectra [6] have made the subject interesting. The spin and parity *i.e.* 1^+ [7] of the ground state of ^{128}I is different from the spin and parity of ^{127}I *i.e.* $5/2^+$ [8]. These properties are much helpful in explaining and understanding the (n,γ) consequences in iodine containing compounds.

　　One of the earlier studies on the production of ^{128}I is due to Benczer *et al.* [9]. Maruyama and Idenawa [10] produced ^{128}I atoms by small research reactor. Alfassi [11] produced ^{128}I by neutron activation of KIO_3. Dissolution effect on recoil ^{128}I retention in the form of IO_4^- ion following (n,γ) reaction in KIO_4 target has been studied by Sharma and Mishra [12]. Sharma and Mishra [13] also studied the behavior of energetic radioiodine atoms arising in $NaIO_3$-$NaNO_3$ solid solutions. Rhode [14] performed a series of measurements of nuclear beta and gamma radiation arising in neutron irradiated NaI(Tl) crystal. Islam *et al.* [15] studied the ^{127}I(n,γ)^{128}I reaction and observed 248 transitions in the photon energy range 4.5 - 6.8 MeV. Zaman *et al*. [16-22] studied ^{128}I reactions in several iodine containing organic compounds in liquid phase under various chemical environment. The present work deals with the production of ^{128}I atoms through (n,γ) activation of NaI, $NaIO_3.H_2O$, $NaNO_4$, KI, KIO_3 and KIO_4 using an (^{241}Am-Be) neutron source in order to study the chemical effects of nuclear transformations in these solid iodocompounds of inorganic origin.

2. Materials and methods

2.1. *Chemicals*

All target materials viz. sodium iodide (99.5%, E. Merck, Germany), sodium iodate (99-100%, BDH, England), sodium periodate (BDH, England), potassium iodide (99%, E. Merck, India), potassium iodate (Matheson Coleman and Bell), potassium periodate (99.8%, Loba Chemie, India) with natural ^{127}I were of AR Grade and used as received.

2.2. *Preparation of paraffin cylinders*

Five paraffin cylinders were prepared with the help of commercial paraffin wax bought from local market (Shaheb Bazar, Rajshahi City). The diameters of these cylindrical paraffin blocks were 0.5 cm, 1 cm, 1.5 cm, 2.0 cm and 2.5 cm.

2.3. *Sample preparation*

Requisite quantities of solid iodocompounds were weighed through a digital balance (precision 0.0001gm) so that each sample target contains 3.626×10^{21} iodine atoms. The calculated weights of these targets were 1 gm KI, 1.2883 gm KIO_3, 1.3846 gm KIO_4, 0.9030 gm NaI, 1.2999 gm $NaIO_3.H_2O$ and 1.2876 gm $NaIO_4$. The measured samples were then separately encapsulated into a 2.25 sq. cm. polyethylene envelope and sealed by an electric sealer machine.

2.4. *Thermal neutron irradiation*

The encapsulated iodocompounds were irradiated with the help of an [241]Am-Be neutron source for the production of [128]I through [127]I (n,γ) [128]I reaction. Specification of the source and irradiation detail is described elsewhere [23]. The source ([241]Am-Be) was taken out from the hole of the neutron housing and then placed into a paraffin cylinder. The samples were then attached with the wall of the paraffin cylinder in the middle position. In this way, the solid iodocompounds prepared as described above were subjected to thermal neutron activation for 3 hours in dark. After 3 hours of irradiation, the irradiated samples were removed from the wall of the paraffin cylinder and carried to the laboratory for counting with the help of an end window G.M. Counter. Among those five paraffin cylinders, the cylinder having diameter 2.5 cm showed comparatively high activities as compared to remaining other four paraffin cylinders. So, the cylinder of diameter 2.5 cm was used for all subsequent experiments.

2.5. *Counting and analysis*

A G.M. Counter was used to count the activity (counts per minute) of [128]I in the irradiated samples. The counter was set at the operating voltage of 850 volts. The irradiated samples containing radioactive iodine were placed on the first shelf of the sample holder mounted under the G.M. tube for the measurement of radioactivities. The counting (counts per minute) of the samples were counted up to 3 to 4 half lives of [128]I as applicable to individual target. The counting geometry remained unaltered throughout the present series of activity measurements. The activities were subjected to background correction. The background activities were recorded as 50 cpm. The corrected activities due to each sample were analyzed separately by plotting activity *vs*. time elapsed on a semi log graph paper. Decay curves obtained which are typical of numerous observations, show that [128]I atoms are produced during the neutron bombardment. A sample analysis has been shown in Fig. 1. In this way, the yields of [128]I species due to each sample were computed for zero time activities.
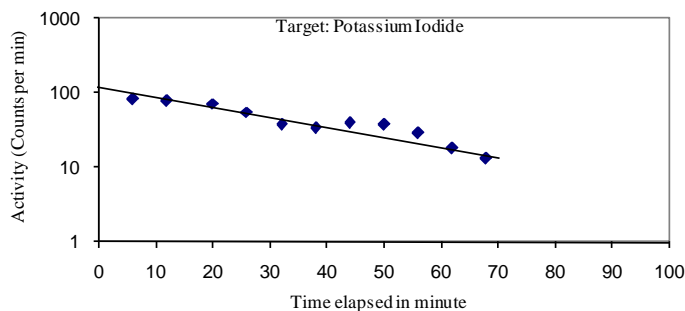

Fig. 1. Time decay analysis of the activity of [128]I species.

3. Results and Discussion

The production of ^{128}I species was effected by an ^{241}Am-Be neutron source through the reaction ^{127}I (n, γ)^{128}I. Paraffin cylinder was used for thermalizing the fast neutrons in order to produce a substantial amount of ^{128}I species. The activity of radioiodine produced during the (n,γ) reaction in potassium iodide is graphically shown in Fig. 1. It is evident from the figure that the ^{128}I species have been produced. It is also apparent from the figure that, the activity of ^{128}I species is decreasing with time. It has been observed from the figure that the zero time activity is 110. The half life of ^{128}I measured in this experiment is found 25 minutes.

The half life of ^{128}I has been calculated with respect to zero time activity. It has been seen from the figure that, activities persisted up to two half lives of ^{128}I. After first and second half lives the activities are 55 and 27.5 respectively and thereafter attain the background counts. For each target, three independent irradiation experiments and counting were performed.

3.1. *Searching an optimum irradiation arrangement*

The optimum irradiation arrangement was searched with the help of five paraffin cylinders of 0.5 cm, 1.0 cm, 1.5 cm, 2.0 cm and 2.5 cm diameters using potassium iodide. Three independent irradiation experiments were carried out for each of 5 paraffin cylinder. Thus, altogether 15 irradiation experiments have been carried out and the results were computed through time-decay analyses. Average zero time activities (ZTA) in counts per minute (cpm) are arranged in Table 1.

Table 1. Thermal neutron activation of KI in paraffin cylinders of different thickness.

Observation	Thickness of paraffin cylinder (cm)	Average ZTA (cpm)
1	0.5	87±5
2	1.0	75±7
3	1.5	90±5
4	2.0	93±7
5	2.5	102±6

It is seen from the table that, the zero time activities are comparatively high in case of paraffin cylinder of diameter 2.5 cm. So, this paraffin cylinder was assumed to be suitable for (n,γ) activation and was used for all the subsequent experiments.

3.2. (*n,γ*) *Activation of sodium containing solid iodocompounds*

Results on sodium containing iodocompounds have been arranged in Table 2. It is to be noted from Table-2 that the (n,γ) activation of sodium containing iodocompounds (*i.e.* NaI, $NaIO_3.H_2O$ and $NaIO_4$) produce the average ^{128}I activities of 108±2, 93±4 and 88±6,

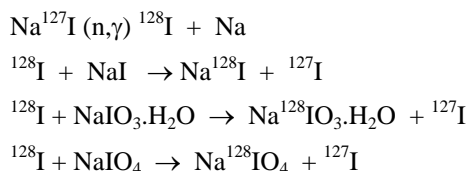respectively. Recorded zero time activities (ZTA) show a decreasing trend while passing through NaI to $NaIO_4$.

Table 2.   (n,γ) activation of sodium containing iodocompounds.

Compounds	Molecular Weight	Target Thickness (mg cm^{-2})	1st Irradiation	2nd Irradiation	3rd Irradiation	Average ZTA (cpm)
NaI	150	401.33	110	105	110	108 $\pm$ 2
$NaIO_3.H_2O$	215.92	577.73	98	90	90	93 $\pm$ 4
$NaIO_4$	213.89	572.27	95	90	80	88 $\pm$ 6

The fate of a hot atom depends, initially upon the particular way of deexcitation and, at the end of the process, on its chemical properties. When an iodocompound undergoes thermal neutron activation, the ^{127}I nucleus becomes activated by neutron capture; the excitation energy is equal to the neutron binding energy. The compound nucleus thus formed de-excites its excess energy through the emission of one or more gamma quanta and a net atomic recoil results. More specifically, atoms produced in solids by neutron capture as in the case under discussion, possess energy in excess of the thermal vibration energy of the crystal lattice. In turn, their chemical properties differ from those of the same atoms under normal temperature conditions. Other than KI and NaI, the other iodocompounds used in the present investigation are oxy-salts of iodine. Since the energies of chemical bonds in the present cases are usually of the order of a few eV, it is apparent that the majority of neutron capture reactions will result in bond breaking, leaving the ^{128}I atoms in a quasi-free state [24].

Assuming that the target molecule is initially torn down [25] there are the following general stages during which reactions might occur between the radioactive atoms and various fragments and molecules to give the observed products: i) Hot reactions take place with recoil atoms either in flight [26] or at the end of their trail, while the immediate surrounding is still thermally or electronically excited [27,28], ii) Reactions occur in the target after initial thermal excitation has been dissipated and iii) Reactions may also occur during the chemical work up of the sample. In our case, we have taken equal number of ^{127}I atoms to undergo (n,γ) activation; the difference is that ^{127}I atoms exist under different chemical environment. After production of ^{128}I species in the target compounds, radiations from β-decay (electron capture) and γ-transitions cause various reactions to occur mostly towards the end of the track. In our solid iodotargets, initial recoil energy would not be so important and the decisive region for chemical reaction might be terminal 'hot zone' in which energy would be dissipated in a small volume, causing melting, chemical decomposition and vacancy production around the recoiling species. Partial intermingling of the material is speculated in a volume comprising ~3×10^6 atoms; the molten region exists for a time of about 10^{-9} s, and this is sufficient for thermal equilibrium to be established and for many chemical processes to occur in the molten region, since the

normal lattice vibration period is $\sim 10^{-13}$ s [26,29]. As ^{128}I nuclei are normally produced with a very high energy in the present target compounds they lose excess energy in successive interactions with the medium and should become stabilized through various reactions. Reactions between a recoil fragment and the parent molecule (*i.e.* the ion-molecule reactions), exchange reactions, and diffusional jumps by interstitial or vacancy mechanisms are possible. The ^{128}I atoms or ions produced in our iodo-targets loss its kinetic energy and is stabilized in various chemical forms within the solid matrices. Following propositions can be made for the present sets of compounds; after bond rupturing, the energetic atoms may re-enter the parent molecule similar to the thermal exchange:

$$Na^{127}I \ (n,\gamma) \ ^{128}I \ + \ Na$$
$$^{128}I \ + \ NaI \ \rightarrow Na^{128}I \ + \ ^{127}I$$
$$^{128}I + NaIO_3.H_2O \ \rightarrow \ Na^{128}IO_3.H_2O \ + \ ^{127}I$$
$$^{128}I + NaIO_4 \ \rightarrow \ Na^{128}IO_4 \ + \ ^{127}I$$

Various types and varied amount of diverse iodine species are produced during the activation process (see later). Energetic atoms may possess varying degrees of excitation that determines their subsequent chemical fate. In gaseous or liquid phases, hot atoms generally react with the intact molecules of the medium. However in solid systems, as in the present case, the displacement of the recoiled atoms is much shorter and the lifetime of lattice defects, excited states and molecule fragments is longer, radiation damage (randomly distributed chemical changes) plays an important part [30]. It is reported [31] that neutron irradiation of iodate and periodate solutions produce IO_4^-, IO_3^-, IO_2^-, IO^-, I^- species with the distribution as shown in Table 3.

Table 3. Species produced in the neutron radiation of iodate and periodate solutions.

Reaction	Species	%	Reaction	Species	%
	IO_3^-	6		IO_4^-	6
$IO_3^- \ + \ n \rightarrow$	IO_2^-	34	$IO_3^- \ + \ n \rightarrow$	IO_3^-	28
	IO^-	54		IO_2^-	60
	I^-	6		IO^-	6

However, it is difficult to ascertain with the present sets of experiments, in what chemical forms are hot atoms stabilized in the crystals of NaI, $NaIO_3.H_2O$ and $NaIO_4.$ Presence of water of crystallization as in the case of hydrated iodate ($NaIO_3.H_2O$) may have remarkable influence on the product yield [32]. The difficulties of determining the chemical nature and environment of a recoil atom arise because of the very low concentrations of such atoms. Batalha and Bellido [33] investigated the state of ^{128}I hot atoms in the crystalline periodates of $NaIO_4$, $Na_2H_2IO_4$ and $Na_4I_2O_9.3H_2O$ containing ^{127}I.

They postulated that the excitation of hot atoms through Auger processes and subsequent relaxation steps in a particular chemical and structural environment seems to explain the results better than recoil in the de-excitation of [128]I atoms. In our case we must take into consideration two other factors that can affect the relative abundance of the recoil fragments: a) mass of the iodine atom that influences the recoil energy, the number of atoms lost and the extent to which the recoil fragment is reduced; b) the enthalpy of formation of the IO_n^- ion that governs the tendency towards reduction or breakdown of parent molecule. The probability of regenerating the parent compound, partly disintegrated by the radioactive recoil, decreases with increasing structural complexity since the number of possible chemical forms produced by the decomposition of a few molecules by recoil energies might be quite large. In the case of iodates, a restricted distraction and disarray would justify the fact that each oxy-salt reveals its own behavior during and after each nuclear event. Our present results ratify the fact that the lattice surrounding the radioiodine species appears to be an important aspect reported earlier for iodate-periodate crystalline mixtures [32, 34]. Crystal structure of sodium periodates has been reported [35]. Evidently, the explanation for the results observed in the present study can be worked out viewing to the direct participation of the medium in the changes occurring during the nuclear transformation and, in particular, in the final stages when the process ends up by chemical reactions. Near the end of the track, the recoil species loses energy predominantly by ejecting atoms from the medium forming interstitials [24]. It is reported [36] that in hard ionic solids containing a simple molecular ion, such as the salts of the oxy-anions, the process probably requires about 25 eV. The recoil atom may then react with their parent fragments or interact with a neighboring parent molecule or may remain trapped in the lattice site.

From Table 4, one can also note that the activities decrease while passing from NaI to $NaIO_4$. This result points out that as [128]I atoms experience relatively complex and structurally different chemical environment, the activities of radioiodine decrease. Results indicate that self absorption of radiations might be important. Self absorption and self scattering depend not only on β-particle energy, but also on the chemical form of the sample and on the geometrical arrangement of sample and detector [37]. As the counting in the present work has been performed under constant geometry, chemical composition of the sample and the generated radionuclides in a specified volume element are accordingly responsible for the observed effects. Uniformity is also important insofar as the specification of maximum surface density, set by a given experimental situation, applies not only to the source as a whole but to any small portion of it. Self scattering and self absorption become negligible for samples $<< 1$ mg cm^{-2} [37]. Conversely, our targets are thick enough (401–577 mg cm^{-2}) in which the incident bombarding particles are significantly degraded in energy. The thickness values (mg cm^{-2}) of sodium containing iodocompounds in our cases are considerable and cannot be disregarded (*cf.* Table 4). In emission sources of finite thickness where radiant energy quanta emitted by atoms (or molecules) are absorbed by atoms of the same kind present in the same source, radiation level reduces. The absorbed energy is usually dissipated by collisional transfer, or through

emission of radiant energy of the same or other frequencies. In consequence, the observed radiant intensity of a spectral line (or band component) emitted by a source may be less than the radiant intensity would be from an optically thin source having the same number of emitting atoms. Self-absorption may occur in all emitting sources to some degree, whether they are homogeneous or not. Reduction of the intensity of the center of an emission line occurs by selective absorption by the cooler portions of the source of radiation [38, 39].

3.3. *(n,γ) activation of potassium containing solid iodocompounds*

Similar results have been recorded in case of potassium containing iodocompounds *viz.* KI, KIO_3 and KIO_4. Results are shown in Table 3.

Table 4. (n,γ) activation of KI, KIO_3 and KIO_4.

Compounds	Molecular Weight	Target Thickness $(mgcm^{-2})$	1st Irradiation	2nd Irradiation	3rd Irradiation	Average ZTA (cpm)
KI	166.1	444.44	110	96	98	102 ± 6
KIO_3	214.01	572.58	98	98	100	98 ± 1
KIO_4	230	615.38	90	90	100	83 ± 5

Results of Table 3 also show a decreasing trend in the ^{128}I activities as seen in sodium containing solid iodocompounds considered above. Radioactive ^{128}I recoils formed by neutron capture in crystalline KIO_3 and KIO_4 appeared as iodide, iodate and periodate ions on analysis of alkaline aqueous solutions of the irradiated solids. With KIO_3 more than two-thirds of the radioiodine was retained as radioiodate and small amounts of radioperiodate. In the KIO_3 crystal, neutral radicals of the type IO and IO_2 are most likely to be formed [23]. In KIO_4, 90% of the recoils appeared as radioiodate ($K^{128}IO_3$) while periodate was less than 10%. The radioactive iodide ion was readily oxidized to IO_3^- and IO_4^- thermally and by γ irradiation [31]. Potassium iodo-salts used in the present investigation undergo similar reactions as speculated for sodium containing iodocompounds. The question of complex chemical environment again comes. When iodine atoms are bound to structurally more complex chemical environment, the radioactivies would be less due to self absorption of γ-rays. Here the crystalline lattice seems to ensure a higher stability of the primary products. In solids, the Mossbauer effect has been used to study various unstable complexes formed during β disintegration by a daughter atom in the crystal lattice [24]. It is reported that $I^*O_2^-$ is a stable recoil product in the solid state, and is probably the precursor of $I^*O_3^-$ [40,41]. Neutron activation of oxygen containing rhenium compounds have been investigated [42,43]; the importance of composition and structure has been dealt in some detail. It is worth noting here that as

molecular weight of the substances used in the present investigation is increased, the activities get decreased. Reactions between a recoil fragment and the parent molecule, exchange reactions and diffusional jumps by interstitial or vacancy mechanisms are highly likely.

4. Conclusion

It is evident that ^{128}I atoms have been produced in the neutron irradiation of some solid iodo oxy-salts of sodim and potassium. Chemical nature and environment of the recoil atom govern the radioactivity produced in these oxygen containing solid iodocompounds. A good deal of information on possible exchange reactions will be needed before detailed statements could be made regarding the mechanisms of the radiochemical reactions in these solids.

Acknowledgements

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