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The Oxidation State of ^{126}I Arising from the (n,2n) Reaction in Inorganic Iodine Compounds

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A comparison of the chemical effects of the (n,γ) and (n,2n) reactions has been thought to present a basis for deciding whether or not the final state of recoil atoms is dependent on the initial recoil energy, since the (n,2n) reactions are estimated to impart a far larger recoil energy to the atoms than the (n,γ) reactions induced by thermal neutrons. In an earlier paper, we reported that ^{78}Br arising from the (n,2n) reaction gave nearly the same retention as ^{80m}Br from the (n,γ) reaction in $NaBrO_3$.[1,2] However, almost concurrently, Aten and his co-

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[1] N. Saito, F. Ambe and H. Sano, *Nature*, **205**, 688 (1965).

[2] F. Ambe, H. Sano and N. Saito, *Radiochim. Acta*, **9**, 116 (1968).

workers reported that ^{126}I arising from the (n,2n) reaction showed a considerably lower retention than ^{128}I from the (n,γ) reaction in KIO$_3$.[3] Recently, Dupetit and Aten have reported similar experiments on CsIO$_3$[4] and CsIO$_4$.[5] The present work was undertaken in order to see if this difference found between the behavior of ^{126}I and ^{128}I is a general phenomenon in inorganic iodine compounds of different types.

KIO$_3$, KH(IO$_3$)$_2$, I$_2$O$_5$, and NaIO$_4$ were irradiated with fast neutrons obtained from the 66 cm cyclotron of this Institute, and the distribution of the ^{126}I produced by the ^{127}I(n,2n)^{126}I reaction among various oxidation states was determined. The results were compared with data found in literature on the ^{128}I from the ^{127}I(n,γ)^{128}I reaction.

Experimental

The samples were commercially-available reagents of a high purity. They were irradiated for 3 hr with fast neutrons obtained by bombarding a metallic lithium target with 3.5 MeV deuterons accelerated in the cyclotron. The temperature of the samples during the neutron irradiation was slightly higher than room temperature. The neutron flux was 10^6—10^7 neutrons/cm$^2\cdot$sec. The irradiated samples were subjected to the following chemical separation procedure within a few hours after neutron irradiation. The irradiated KIO$_3$ and NaIO$_4$ were dissolved in water, while the KH(IO$_3$)$_2$ and I$_2$O$_5$ were dissolved in aqueous solutions containing a large excess of KOH. The alkaline solutions were then carefully neutralized with dilute nitric acid. ^{126}I atoms in different oxidation states were separated by the following chemical procedures after an appropriate amount of a carrier (I$^-$, IO$_3^-$ or IO$_4^-$) had been added: (1) Iodine atoms in the lower oxidation states (I$^-$, I$_2$ and IO$^-$) were precipitated as AgI. (2) Iodine atoms in the lower oxidation states were separated by exchange extraction with I$_2$ dissolved in CCl$_4$. (3) IO$_3^-$ ions were precipitated as Ba(IO$_3$)$_2\cdot$H$_2$O. (4) IO$_3^-$ and IO$_4^-$ ions were separated from iodine atoms in the lower oxidation states by an anion-exchange procedure.[6] (5) IO$_4^-$ ions were precipitated as KIO$_4$. The radioactivity of ^{126}I was measured by means of a NaI scintillation counter after the short-lived nuclides had decayed. The data were reproducible with allowable deviations, and the results obtained by the different procedures were in good agreement.

Results

TABLE 1. OXIDATION STATE OF ^{126}I ARISING FROM THE (n,2n) REACTION (the present work)

Compound	I$^-$+I$_2$ (%)	IO$_3^-$ (%)	IO$_4^-$ (%)
KIO$_3$	44±1	56±1	<1
KH(IO$_3$)$_2$	60±1	40±1	<1
I$_2$O$_5$	44±1	56±1	<1
KIO$_4$	4±1	94±2	2±1

TABLE 2. OXIDATION STATE OF ^{128}I ARISING FROM THE (n,γ) REACTION (a summary of data of previous workers)

Compound	I$^-$+I$_2$ (%)	IO$_3^-$ (%)	IO$_4^-$ (%)	Reference
KIO$_3$	33	67	0	a)
KH(IO$_3$)$_2$	47	53	0	a)
I$_2$O$_5$	31	69	0	b)
KIO$_4$	10	86	4	c)

a) R. E. Cleary, W. H. Hamill and R. R. Williams, Jr., *J. Amer. Chem. Soc.*, **74**, 4675 (1952).

b) M. Vlatković and A. H. W. Aten, Jr., "Chemical Effects of Nuclear Transformations (Proc. Symp. Prague)," Vol. I, p. 551, IAEA, Vienna (1961).

c) A. H. W. Aten, Jr., G. K. Koch, G. A. Wesselink and A. M. de Roos, *J. Amer. Chem. Soc.*, **79**, 63 (1957).

The results obtained are shown in Table 1. Storage for several hours at room temperature was found to cause no detectable change in the chemical distribution of ^{126}I in the irradiated materials. In I(V) compounds a slight increase in the parent form was observed after storage exceeding 24 hr at room temperature. In Table 2 the data on the (n,γ) reaction reported by previous workers are summarized. A comparison of Tables 1 and 2 indicates that for both I(V) and I(VII) compounds the distribution of radioactive recoil iodine atoms is considerably different for the (n,γ) and (n,2n) reactions. It may thus be concluded that, for all the I(V) compounds studied, the retention of ^{126}I is lower than that of ^{128}I; this conclusion is in accordance with the previous report on KIO$_3$ and CsIO$_3$.[3,4] Since we found no significant difference between the behavior of the recoil bromine arising from the ^{79}Br(n,γ)^{80m}Br and ^{79}Br(n,2n)^{78}Br reactions in NaBrO$_3$,[1,2] it is unlikely that the difference observed in the iodine compounds stems from the difference in recoil energy accompanying each nuclear reaction. We also reported previously that the ^{80m}Br and ^{82}Br arising from the (n,γ) reactions showed slightly different retention values in various bromates, and interpreted the difference as resulting from the highly-converted isomeric transition of ^{82m}Br to ^{82}Br after the (n,γ) reaction.[7,8]

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[3] A. H. W. Aten, Jr., M. Lindner-Groen and L. Lindner, "Chemical Effects of Nuclear Transformations (Proc. Symp. Vienna)," Vol. II, p. 125, IAEA, Vienna, 1965.

[4] G. A. Dupetit and A. H. W. Aten, Jr., *Radiochim. Acta*, **7**, 165 (1967).

[5] G. A. Dupetit, *ibid.*, **7**, 167 (1967).

[6] M. L. Good, M. B. Purdy and T. Hoering, *J. Inorg. Nucl. Chem.*, **6**, 73 (1958).

[7] N. Saito, F. Ambe and H. Sano, *Nature*, **206**, 505 (1965).

[8] N. Saito, F. Ambe and H. Sano, *Radiochim. Acta*, **7**, 131 (1967).

Thus, it may be possible that the difference observed in the retention of ^{126}I and ^{128}I is also a result of an internal conversion process of the excited ^{126}I or ^{128}I.

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