Chemical Effects Following the (n, 2n) Reaction in Cobaltammine Bromides

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It has previously been reported that radiobromine, ^{80}Br, ^{80m}Br and ^{82}Br, arising from thermal neutron capture and isomeric transition in cobaltammine complexes $[Co(NH_3)_5X]Br_{2,3}$ and $[Co(en)_2X_2]Br_{1,3}$, could substitute for their ligands, X, to produce radiobromine-labeled complexes.[1,2] In order to compare the behavior of hot radiobromine atoms produced in different nuclear reactions, the present authors investigated the behavior of radiobromine, ^{78}Br, arising from $(n, 2n)$ reaction induced by fast-neutron irradiation of these complexes.

The samples listed in Table 1 were irradiated with 14 MeV neutrons produced by Toshiba model NT-20 Cockcroft-Walton type neutron generator at room temperature or liquid nitrogen temperature. The flux was approximately $1—2\times 10^7$ neutrons /cm^2·sec. The samples were dissolved in water immediately after the completion of irradiation and the solution was passed through columns of a cation exchange resin and anion exchange resin successively. The fraction adsorbed on the cation exchanger contained ligand bromine including radiobromine, whereas the fraction adsorbed on the anion exchanger contained bromide ions and anionic radiobromine. Measurements were made on each fraction with respect to the mixture of activities of ^{78}Br, ^{13}N and ^{60m}Co produced together, and with respect to the content of bromine. After correction for ^{13}N and ^{60m}Co activities, the ligand yield (the fraction of the total ^{78}Br activity retained in ligands of complex ions) was calculated. In Table 1 are shown the ligand yields as well as the enrichment factors.

Table 1 indicates that the ligand yields vary with the kind of ligand X. Comparison is made of the behavior of hot ^{78}Br atoms resulting from $(n, 2n)$ process with that of ^{80m}Br and ^{82}Br atoms arising from (n, γ) process. In Figs. 1 and 2 are shown the dependencies of the ligand yields in $(n, 2n)$ and (n, γ) cases upon some properties of ligand X. It is seen from these figures that the dependencies are very much alike for both nuclear processes. Therefore, it may be concluded that

TABLE 1. LIGAND YIELD OF IRRADIATED COMPLEX SALTS $[Co(NH_3)_5X]Br_{2,3}$ AND $[Co(en)_2X_2]Br_{1,3}$

No.	Irradiated salt	Ligand yield (%)* at room temp.	Enrichment factor
1	$[Co(NH_3)_6]Br_3$	3.5±0.1(6.9)	$>2\times10^2$
2	$[Co(NH_3)_5NCS]Br_2$	3.2	$>0.6\times10^2$
3	$[Co(NH_3)_5NO_2]Br_2$	3.5±0.1	$>0.8\times10^2$
4	$[Co(NH_3)_5OH_2]Br_3$	7.9±0.2	0.7×10^2
5	$[Co(NH_3)_5ONO]Br_2$	4.1±0.1	3×10^2
6	$[Co(NH_3)_5ONO_2]Br_2$	9.0±0.4	0.4×10^2
7	$[Co(NH_3)_5Cl]Br_2$	13.8±0.3(16.7)	$>6\times10^2$
8	$[Co(NH_3)_5I]Br_2$	12.3±0.3	$>1\times10^3$
9	$[Co(NH_3)_5Br]Br_2$	16.8±0.6(16.2±0.8)	—
10	$[Co(en)_3]Br_3\cdot3H_2O$	0.4±0.4(3.7)	0.8×10^2
11	*trans*-$[Co(en)_2(NH_3)_2]Br_3\cdot H_2O$	3.2±0.3	0.3×10^2
12	*trans*-$[Co(en)_2(NCS)_2]Br.1\frac{1}{2}H_2O$	1.1±0.2	
13	*cis*-$[Co(en)_2(NCS)_2]Br\cdot H_2O$	3.1±0.4	$>0.4\times10^2$
14	*trans*-$[Co(en)_2Cl_2]Br$	7.9±0.1(12.2)	1×10^2
15	*cis*-$[Co(en)_2Cl_2]Br\cdot H_2O$	10.8±1.3	1×10^2
16	*trans*-$[Co(en)_2Br_2]Br$	26.8±0.5(22.1±0.7)	—

* Nearly identical with the yield as $Co(NH_3)_5Br^{*2+}$, or as $Co(en)_2XBr^{*1,2+}$. Note: Figures in parentheses indicate the ligand yield obtained at liquid nitrogen temperature. The ligand yields obtained at liquid nitrogen temperature are about 3% higher than those obtained at room temperature for the same target complex except for $[Co(NH_3)_5Br]Br_2$ and $[Co(en)_2Br_2]Br$.

1) N. Saito, T. Tominaga and H. Sano, This Bulletin, **35**, 63 (1962).

2) N. Saito, S. Ito and T. Tominaga, *ibid.*, **38**, 504 (1965).

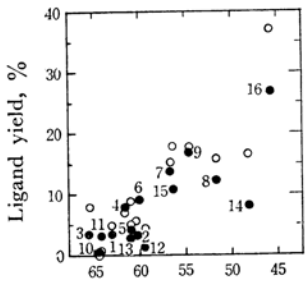

Fig. 1. The relationship between the ligand yield and the frequency of the maximum in the first absorption band.
○ Ligand yield in (n, γ) reaction
● Ligand yield in $(n, 2n)$ reaction
(Numbers in Figs. 1 and 2 correspond to those in Table 1.)

the large difference in maximum recoil energies of hot-atoms does not play a dominant role in deciding the fate of recoiling radiobromine atoms in these particular reactions.

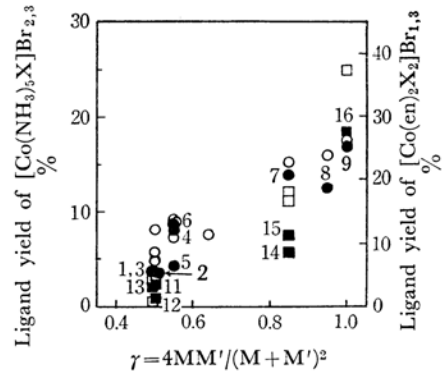

Fig. 2. The relationship between the ligand yield and the γ-value calculated for collision of a recoil atom (mass M) with a donor atom (mass M′) in ligand X*.
○ (n, γ) reaction } in $[Co(NH_3)_5X]Br_{2,3}$
● $(n, 2n)$ reaction } series
□ (n, γ) reaction } in $[Co(en)_2X_2]Br_{1,3}$ series
■ $(n, 2n)$ reaction }
* Different ligand yield scales are used for the two series on the basis of the estimation made in the previous paper.[1]