Chemical Effects of the Isomeric Transitions of ^{127m}Te and ^{129m}Te in Inorganic Tellurium Compounds

Shizuko AMBE and Nobufusa SAITO

The Institute of Physical and Chemical Research, Wako-shi, Saitama

Received October 16, 1971

The chemical effects of the isomeric transitions of ^{127m}Te and ^{129m}Te have been investigated in solid ammonium and potassium halo-tellurates(IV), tellurium dioxide, and tellurous acid, and also in aqueous solutions of telluric acid containing halide ions. Almost no daughter nuclides were found in the tellurium(VI) fraction in the case of solid halo-complex salts, tellurium dioxide, and tellurous acid. For aqueous solutions and ammonium fluoride solutions of telluric acid, however, 60~70% of the daughter nuclides were found in the tellurium(IV) fraction after the isomeric transition. In the other solutions, more than 90% of the daughter nuclides were found in the tellurium(IV) fraction.

1. Introduction

The chemical effects of the isomeric transitions of ^{127m}Te and ^{129m}Te in tellurium compounds have been studied by many investigators both in aqueous solutions^{1-8,20)} and in solids^{5,7-14)}. However, the effect of ligand halogen in solid halo-tellurates(IV) on the chemical behavior of ^{127}Te or ^{129}Te after the isomeric transition has not yet been investigated. In a previous study of the chemical effects of the β -decay of ^{125}Sb in ammonium halo-antimonates(V), the present authors found that the distribution of ^{125m}Te is dependent on the electronegativity of the ligand halogens¹⁵⁾. In the present experiment, the investigation has been extended to the isomeric transitions of ^{127m}Te and ^{129m}Te in solid halo-tellurates(IV) and in solutions of telluric acid containing halide ions. For the sake of comparison, similar experiments were also carried out on tellurium dioxide and tellurous acid.

2. Experimental

2.1 Production of ^{127m}Te and ^{129m}Te

The ^{127m}Te and ^{129m}Te were produced by the neutron irradiation of, respectively, enriched ^{126}Te and ^{128}Te metal obtained from ORNL.

2.2 Preparation of labeled compounds and storage

Telluric acid labeled with ^{127m}Te or ^{129m}Te was prepared by the oxidation of metallic tellurium¹⁶⁾. Tellurium dioxide and tellurous acid were prepared by the method described in the literature^{17,18)}. Tellurous acid was stored in water to prevent decomposition. Ammonium and potassium halo-tellurates(IV) labeled with ^{127m}Te or ^{129m}Te were prepared by the addition of ammonium or potassium halide to a hydrohalogenic acid solution of tellurium dioxide. The sample solutions were prepared by dissolving telluric acid in 5 M hydrofluoric acid, 5 M hydrochloric acid, 5 M hydrobromic acid, 5 M ammonium fluoride, 5 M ammonium chloride and 5 M ammonium bromide. The concentrations of telluric acid were 0.7 mg/ml for telluric acid labeled with ^{127m}Te and 2 mg/ml for telluric acid labeled with ^{129m}Te . These solutions and tellurous acid were stored in ice water for 1 day in the case of ^{129m}Te -labeled compounds and for 4 days in the case of ^{127m}Te -labeled compounds in order to attain radioactive equilibrium. The hexahalo-tellurates(IV) were sealed in ampoules and were stored in liquid nitrogen for 1 day in the case of ^{129m}Te -labeled compounds and for 4 days in the case of ^{127m}Te -labeled compounds.

2.3 Separation procedure

Solid halo-complex salts were dissolved in ice-cooled 4M hydrochloric acid. Tellurous acid and tellurium dioxide were dissolved in 8M hydrochloric acid, and then the solutions were diluted to 4M in hydrochloric acid. The halide solutions of telluric acid were adjusted to 4M in hydrochloric acid by the addition of distilled water or conc. hydrochloric acid. The solution was put on a column of an anion exchange resin, Diaion SA-100, treated previously with 5M hydrochloric acid. Tellurium(VI) was passed through the column. The tellurium(IV) adsorbed on the resin was eluted with 1M hydrochloric acid.

2.4 Measurement of radioactivity

417 keV gamma rays of ^{127}Te or 460, 487 keV gamma rays of ^{129}Te in the tellurium(IV) and -(VI) fractions and the resin phase were counted by means of a single-channel pulse-height analyser or a multi-channel pulse-height analyser. The distribution of the oxidation states of tellurium was determined by analyzing the decay or growth curves of the tellurium(IV) and tellurium(VI) fractions. In the hydrobromic acid solution of telluric acid, a part of tellurium(VI) was found to be chemically reduced to tellurium(IV). The contribution of chemical reduction was corrected by the analysis of the decay curve of the tellurium(IV) fractions.

3. Results and Discussion

3.1 Ammonium and potassium halo-tellurates(IV)

In ammonium and potassium halo-tellurates(IV) (NH_4TeF_5 , $(\text{NH}_4)_2\text{TeCl}_6$, $(\text{NH}_4)_2\text{TeBr}_6$, KTeF_5 , K_2TeCl_6 and K_2TeBr_6), almost no ^{127}Te or ^{129}Te was found in the tellurium-(VI) fractions; consequently, no effect of the kind of halogen on the chemical effects of the isomeric transitions of ^{127m}Te and ^{129m}Te was observed. The conversion coefficients of ^{127m}Te and ^{129m}Te have been reported to be 470 and 250 respectively, indicating that essentially all the transitions are converted. The converted isomeric transition and the

Table 1 Percentage distribution of ^{127}Te and ^{129}Te in the Te(IV) fraction after the isomeric transition and chemical reduction of tellurate(VI) in various solutions containing halide ions

Species Sample	^{127}Te (IV)*1 (%)	Te(IV)*2 (%)	^{129}Te (IV)*1 (%)	Te(IV)*2 (%)
5M NH_4F	68±3	<0.02	64±3	<0.02
5M NH_4Cl	97±4	<0.02	89±9	<0.02
5M NH_4Br	104±7	<0.02	98±4	<0.02
5M HF	93±5	<0.02	92±4	<0.02
5M HCl	97±7	<0.02	98±4	<0.02
5M HBr	98±2	82±1	109±11	71±2
H_2O	74±3	<0.02	68±3	<0.02

*1 reduction of daughter nuclides arising from the isomeric transition.

*2 chemical reduction of inactive tellurium.

subsequent Auger process are considered to lead to the build-up of a highly-positive charge on the tellurium atoms. However, the results of the present investigation make it possible to conclude that the possible highly-positive charge of the tellurium atoms just after the transition is not reflected in the ultimate fate of the atoms in the halo-tellurates.

3.2 Tellurium dioxide and tellurous acid

In tellurium dioxide and tellurous acid, almost no radioactivity was found in the tellurium(VI) fraction, just as in the case of halo-tellurates(IV). These results show that a daughter tellurium nuclide arising from the isomeric transition can hardly be stabilized in a higher oxidation state than that of the starting material, irrespective of whether the ligands are halogens or oxygen.

3.3 Solutions of telluric acid

Table 1 shows the results obtained for telluric acid in various solutions containing halide ions. No experiment was carried out on solutions of telluric acid in hydriodic acid and ammonium iodide, since the telluric acid was found to be reduced completely to the tetravalent state by the iodide ions during storage. In hydrobromic acid solu-

tions, 70~80% of the telluric acid was chemically reduced to the tetravalent state during storage, while 20~30% remained in the hexavalent state. As has been mentioned in the section describing the experiments, the contribution of chemical reduction during storage could be corrected in this case by the analysis of the decay curves of the tellurium(IV) fraction. In the other solutions, the chemical reduction of telluric acid in the presence of halide ions was not observed.

In the ammonium halide solutions, the chemical distribution of daughter nuclides seems to depend on the kind of halogens. The lowest yield of daughter nuclides in the tellurium(IV) state was obtained in ammonium fluoride solutions. In ammonium chloride and ammonium bromide solutions, most of the daughter nuclides were found in the tellurium(IV) fraction, and no significant difference in the yields was observed. On the other hand, in all of the hydrohalogenic acid solutions, more than 90% of the daughter nuclides were reduced to tellurium(IV) after the isomeric transition, and no effect of the halogens on the chemical distribution of the daughter nuclides was found within the range of experimental error. The difference in the results obtained in the hydrofluoric acid solutions and ammonium fluoride solutions suggests that not only halide ions, but also hydrogen ions, take part in the reduction of hot radio-tellurium. The daughter nuclides in the tellurium(IV) state are about 70% of those in the aqueous solutions containing no halide ions. The fact that almost the same values were obtained in an aqueous solution and in an ammonium fluoride solution may suggest that fluoride ions in ammonium fluoride solutions do not induce the reduction to tellurium(IV) of radio-telluric acid, which is considered to be in an activated state immediately after the isomeric transition. On the other hand, chloride and bromide ions in ammonium halide solutions promote the reduction of the activated telluric acid. The reduction of radio-telluric acid in the solutions of various salts and even in an aqueous

solution seems to originate in the characteristic behavior of hot atoms arising from the nuclear transformation; the phenomena are not observed in ordinary solution chemistry. No isotope effect between ^{127m}Te and ^{129m}Te was observed in any compounds or solutions, as might be expected from the large conversion coefficient of ^{127m}Te and ^{129m}Te .

References

- 1) G.T. Seaborg and J.W. Kennedy: *Phys. Rev.*, **55**, 410 (1939)
- 2) G.T. Seaborg, G. Friedlander and J.W. Kennedy: *J. Am. Chem. Soc.*, **62**, 1309 (1940)
- 3) R.R. Williams: *J. Chem. Phys.*, **16**, 513 (1948)
- 4) R.R. Williams: Natl. Nuclear Energy Ser., Div. IV, 9, Radiochem. Studies, The Fission Product, Book 1, 220 (1951)
- 5) A.N. Murin, V.D. Nefedov, E.N. Sinotova and O.V. Larinov: *Zh. Neorg. Khim.*, **3**, 181 (1958)
- 6) R.L. Hahn: *J. Chem. Phys.*, **39**, 3482 (1963)
- 7) R.L. Hahn: *J. Chem. Phys.*, **41**, 1986 (1964)
- 8) M. Bertet and R. Muxart: Chemical Effects of Nuclear Transformations, Symp. (Vienna), IAEA, Vol. II, p. 13 (1965)
- 9) H.J. Riedel and E. Merz: *Radiochimica Acta*, **6**, 144 (1966)
- 10) D. Danciewicz and A. Halpern: *Nature*, **203**, 856 (1964)
- 11) T. Andersen, L. Johansen and K. Olesen: *Trans. Faraday Soc.*, **63**, 1730 (1967)
- 12) L. Carrillo and S.T. Massiff: *Radiochimica Acta*, **8**, 124 (1967)
- 13) C.H.W. Jones and J.L. Warren: *J. Inorg. Nucl. Chem.*, **30**, 2289 (1968)
- 14) A. Halpern, D. Danciewicz: *Radiochimica Acta*, **11**, 31 (1969)
- 15) S. Ambe and N. Saito: *Radiochimica Acta*, **16**, 40 (1971)
- 16) G. Brauer: Handbook of the Preparative Inorg. Chem., Vol. 1, 2nd ed. p. 451 (1963)

- 17) C. Gh Macarovici: *Chem. Abst.*, **41**, 6494a (1947) Chemie, 8th Aufl. Ammonium, S. 318, Kalium, S. 802.
18) E.D.R. Prideaux and J.O. Millott: *J. Chem. Soc.*, 2703 (1929) 20) M. Hillman and A.J. Weiss: *Radiochimica Acta*, **15**, 79 (1971)
19) Gmelins Handbuch der anorganischen

要 旨

無機テルル化合物中の ^{127m}Te および ^{129m}Te の核異性体転移の化学効果

安部 静子, 斎藤 信房
理化学研究所

NH_4TeF_6 , $(\text{NH}_4)_2\text{TeCl}_6$, $(\text{NH}_4)_2\text{TeBr}_6$, KTeF_6 , K_2TeCl_6 , K_2TeBr_6 , 二酸化テルル, 亜テルル酸の固体およびテルル酸水溶液につき, ^{127m}Te および ^{129m}Te の核異性体転移の化学効果を研究した。Te(IV) の固体の試料の場合には, 生成した娘核種は, Te(VI) の酸化状態にはほとんど見出されなかった。テルル酸の水溶液およびフッ化アンモニウム溶液では, 核異性体転移の結果, 娘核種の60~70%が Te(IV) の酸化状態に見出され, その他の溶液では90%以上が Te(IV) の状態に見出された。