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An Investigation of the Products of the Disintegration of Uranium by Neutrons

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Products of uranium fission which are chemically antimony, tellurium and iodine have been examined. The following activities have been found:

Antimony	Tellurium	Iodine	Atomic Weight
80 hr.	10 hr.		127
4.2 hr.	70 min.		129
	30 min.	8 days	131
	30 hr.		
5 min.	77 hr.	2.4 hr.	132, 134 or 136
<10 min.	43 min.	54 min.	
<10 min.	60 min.	22 hr.	

Three activities—the 10-hour tellurium, the 70-minute tellurium, and the 8-day iodine—have been identified with known bodies through comparison of half-lives and beta-ray spectra. Genetical relationships of the activities have been established by a technique of periodic separations and by the observation of growth in activity curves.

IN A classical paper in *Die Naturwissenschaften*[1] Hahn and Strassmann announced the discovery of active barium as a product of neutron irradiation of uranium. Meitner and Frisch[2] suggested an explanation of this phenomenon on the basis of the liquid-drop model of nuclei and predicted that the process of uranium fission should be accompanied by the liberation of 200 Mev of energy. Subsequent work in several laboratories[3–7] has demonstrated the production of heavy energetic particles by neutron irradiation of uranium. Further chemical work has shown that the products of uranium fission are numerous. Curie and Savitch[8] have found several rare earth activities and an alkali metal body, and Hahn and Strassman[9, 10] have stated that in addition to a number of barium activities they have found active strontium, yttrium, krypton and cesium. Numerous other workers have also reported activities.

When news of the discovery of the uranium cleavage reached this laboratory, the properties of the x-rays emitted by the 77-hour "trans-

[1] O. Hahn and F. Strassmann, Naturwiss. **27**, 11 (1939).
[2] L. Meitner and O. Frisch, Nature **143**, 239 (1939).
[3] O. Frisch, Nature **143**, 276 (1939).
[4] F. Joliot, Comptes rendus **208**, 341 (1939).
[5] R. D. Fowler and R. W. Dodson, Phys. Rev. **55**, 417 (1939).
[6] R. B. Roberts, R. C. Meyer and L. R. Hafstad, Phys. Rev. **55**, 417 (1939).
[7] G. K. Green and L. W. Alvarez, Phys. Rev. **55**, 417 (1939).
[8] I. Curie and P. Savitch, Comptes rendus **208**, 343 (1939).
[9] O. Hahn and F. Strassmann, Naturwiss. **27**, 89 (1939).
[10] O. Hahn and F. Strassmann, Naturwiss. **27**, 163 (1939).

uranic" activity were immediately examined. The radiations were shown to be characteristic iodine x-rays. Further work established that the 77-hour "transuranic" body is tellurium and that its 2.4-hour daughter is iodine.[11] A series of investigations was then made on those products of uranium cleavage which are chemically antimony, tellurium and iodine. The results[12, 13] have been published. The purpose of this paper is to give in detail a description of the work. The table in the abstract summarizes the activities. The values of half-lives given in this table take precedence over values previously quoted by the author.

Method

Uranium samples were activated by neutrons formed through the bombardment of beryllium by 8.0-Mev deuterons produced by the cyclotron. Samples were placed about one centimeter from the beryllium source and separated from it by a water-cooling brass plate. On all other sides the uranium was surrounded by paraffin. In other words, the uranium was bombarded by a mixture of slow and fast neutrons.

Several different chemical forms of uranium were employed, namely, uranyl nitrate ($UO_2(NO_3)_2 \cdot 6H_2O$), uranyl chloride (UO_2Cl_2) and uranium oxide (UO_3). In most cases the C.P. salts were repurified. Activities were for the most part measured on a Lauritsen type electroscope provided with an aluminum window 0.0005" thick. Calibration with a uranium standard showed one division per second to be equal to one-third of a microcurie. For the identification of x-rays a methyl-bromide-filled ionization chamber was used in conjunction with a d.c. amplifier of the type described by Dubridge. The ionization chamber was also used for the measurement of weak beta-activities.

Chemical Separations

In making chemical separations several factors were considered. First, it was highly desirable to make the separation a specific one. This is not always possible without resorting to a fairly complicated procedure. Second, it was necessary to make the separations in the shortest possible time. Finally, it was desirable to have separations which were semi-quantitative.

The separation employed for antimony was a volatilization of stibine (SbH_3) and the subsequent passage of this gas into a silver nitrate solution. Under these circumstances one obtains a precipitate of silver antimonide (Ag_3Sb).

In the antimony experiments a 10-gram sample of uranium oxide was irradiated with neutrons. After the activation had ceased, the oxide was dissolved in 4 normal sulfuric acid.

The sulfate solution was introduced into a hydrogen generator (employing zinc and 4 normal sulfuric acid). Under these conditions stibine is formed, which is carried along in the stream of hydrogen. The stibine, on coming in contact with a silver nitrate solution, forms a black precipitate of silver antimonide. The only element which reacts similarly is germanium. A later experiment showed that the activity was due to antimony and not to germanium. Besides being an almost specific separation, the stibine volatilization can be performed very quickly. In one experiment it was possible to start reading an antimony precipitate within four minutes after the conclusion of an irradiation. The sole disadvantage of this separation is the fact that only ten percent of the antimony is reduced to stibine. The remainder is precipitated in the hydrogen generator in the metallic state.

In order to separate tellurium from all the other elements, an extensive procedure is necessary. However, a fairly specific precipitation is obtained through the reduction of the element by sulfurous acid. If one passes sulfur dioxide into a boiling 3 normal hydrochloric acid solution containing appropriate carriers, gold, palladium, tellurium and selenium are precipitated completely in the elemental state. A large fraction of the polonium is likewise carried down. At first glance the precipitation of gold, palladium, selenium and polonium seems a great handicap. However, supplementary experiments showed that those four elements were not among the strongly active products of uranium cleavage. Thus, gold and palladium were separated from tellurium by reduction with formic acid. Selenium was precipitated from 12 normal hydrochloric acid by sulfur dioxide. Tellurium is not

[11] P. Abelson, Phys. Rev. **55**, 418 (1939).
[12] P. Abelson, Phys. Rev. **55**, 670 (1939).
[13] P. Abelson, Phys. Rev. **55**, 876 (1939).

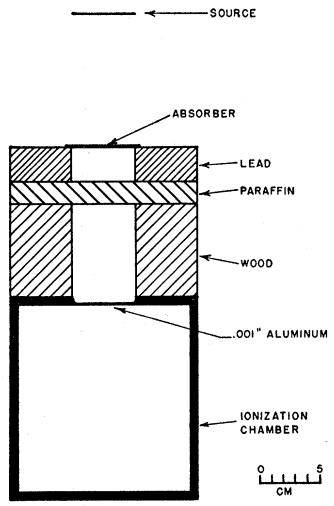

FIG. 1. Experimental arrangement used for determination of x-ray absorption.

precipitated under these conditions. On the other hand, hydrazine hydrochloride precipitates tellurium but not polonium. The reduction of tellurium by sulfur dioxide possesses the further advantages that it is quantitative and can easily be performed in ten minutes.

Several methods were employed for the separation of iodine. In one the iodine was distilled from a dilute nitric acid solution into dilute sodium sulfite. The iodine was then precipitated as silver iodide from a solution acidified by nitric acid.

In a second procedure the iodine was extracted from dilute nitric acid solution with carbon tetrachloride. To insure complete extraction the process was repeated. The iodine was extracted from the carbon tetrachloride by a dilute sodium sulfite solution, and subsequently precipitated as silver iodide. A third procedure—which gave the most satisfactory results in the periodic iodine extractions from tellurium—was carried out in the following manner. The metallic tellurium precipitate was dissolved in nitric acid, to which concentrated sulfuric acid was added, and the mixture was boiled until fumes of sulfuric acid were evolved. The solution was diluted to 6 normal, and silver sulfate was added. The mixture was then saturated with sulfur dioxide. Under these conditions tellurium remains in solution and iodine is reduced from either iodate or iodine to iodide. Any iodine liberated from the tellurium is, therefore, reduced from the valence state in which it was formed to the iodide state in which it is precipitated as silver iodide. The two substances which might interfere in all three of these separations are bromine and element 85. Bromine was eliminated by virtue of the solubility in concentrated ammonia of silver bromide. Element 85 was most easily disposed of by proving that the halogen activities were not genetically related to polonium. The validity of the chemical separations is enhanced by the fact that most of the activities studied possess genetical relationships with other activities. Thus, in the case of the antimony activities decaying into tellurium, if one shows that the daughter substance is uniquely tellurium, one confirms the chemical evidence that the parent is antimony.

For examination of the x-rays emitted by the 3-day tellurium and the 8-day iodine a methyl-bromide-filled chamber was used. The experimental arrangement is shown in Fig. 1. It can readily be seen that the geometrical conditions are excellent. The effect of fluorescent radiation from the absorber is negligible. The cadmium, indium and tin absorbers were used in the form of metallic foils. Antimony as SbO_2 was precipitated in uniform layers on filter paper.

It is well known that the shape of a beta-ray absorption curve is a function of the geometrical conditions employed in obtaining the curve. Hence in showing that one body has the same beta-ray energy as another, it is very desirable to measure the absorption of the radiation from the two substances under the same geometrical conditions. In this work the active samples were of the same thickness and were prepared for examination in similar manners. Furthermore, the intensities of the activities were made nearly equal.

THE 77-HOUR ACTIVITY

Of the fragments of uranium cleavage one of the most thoroughly investigated bodies is a 77-hour activity. Examination of the radiation of this body reveals the emission of x-rays. The emission of these x-rays was first established in March, 1938. Examination of the radiation at that time through use of copper absorbers gave

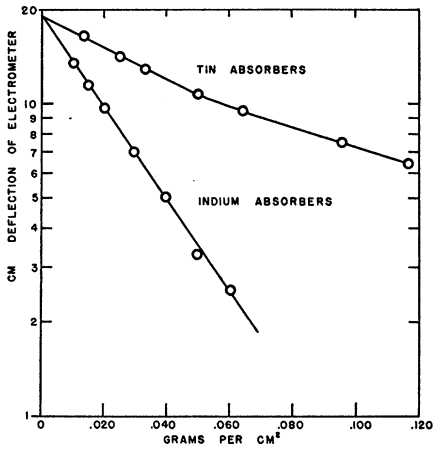

FIG. 2. Absorption of x-rays emitted by 77-hour tellurium. The gamma-background which has been subtracted was about equal to the x-ray effect.

an absorption coefficient which was compatible with the hypothesis that the x-rays were L radiation from a "transuranic" element. The absorption coefficient found was less than expected but geometrical conditions were considered to be poor enough to explain the low result. At that time an electroscope filled with methyl bromide was employed as the detecting instrument, and the available intensity of the 77-hour body was small. When the news of the work of Hahn and Strassmann was heard, the characteristics of the x-rays were at once re-examined. On that occasion a much stronger sample ($\sim$50 microcuries) was available. A number of absorbers were used—copper, bromine, strontium, molybdenum, silver—all of which showed an absorption which increased with atomic number. On trying iodine it was found that the absorption coefficient had markedly decreased. Further experiments revealed an absorption discontinuity lying between tin and indium (Fig. 2). This discontinuity is compatible with the assumption that the x-rays are the characteristic x-rays of iodine. A confirmation of that hypothesis is seen in Fig. 2. Here indium whose K level can be excited by both iodine K_α and K_β shows a single absorption coefficient. On the other hand, tin, whose K level can be excited only by iodine K_β, shows two absorption coefficients in its absorption curve. More recently these x-rays have been investigated by Feather and Bretscher[14] who also find a discontinuity in absorption between indium and tin. They likewise conclude that the radiations are the characteristic x-rays of iodine.

On separating tellurium from a tellurium precipitate which has aged for a week, one can observe a growth in the separated activity (Fig. 3). The activity of the tellurium reaches a maximum about 10 hours after separation. The growth indicates the formation of a daughter substance whose half-life is about $2\frac{1}{2}$ hours. This daughter substance can be easily separated from the tellurium; it has all the chemical properties of iodine, and it decays with a 2.4-hour half-life. The value obtained here for the half-life of the 77-hour body was somewhat different from the 66-hour value quoted by Meitner, Hahn and Strassmann.[15] It was found that a tellurium precipitate made from activated uranium a day after irradiation showed an initial half-life close to 66 hours. In less than a week the slope of the decay curve had become that of a 77-hour activity. In view of the fact that a 30-hour body has been found among the active tellurium fragments, it was assumed that the 66-hour value results from a combination of 30-hour and 77-hour activities. Accordingly, the following experiment was performed. Tellurium was pre-

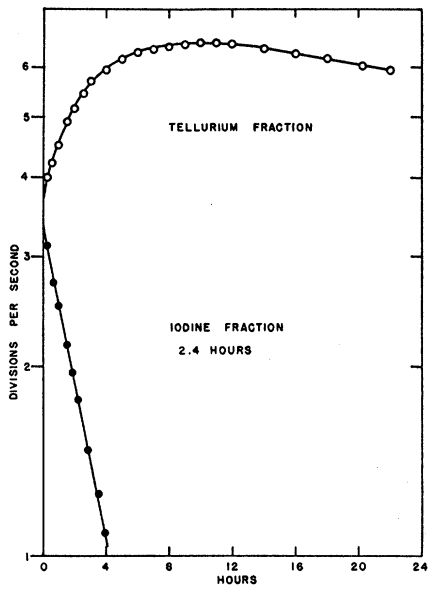

FIG. 3. Decay of 2.4-hour iodine separated from 77-hour tellurium and growth of activity in 77-hour tellurium.

[14] N. Feather and E. Bretscher, Nature **143**, 516 (1939).

[15] L. Meitner, O. Hahn and F. Strassmann, Zeits. f. Physik **106**, 249 (1937).

cipitated from activated uranium at the conclusion of a neutron irradiation. After 12 hours had elapsed, a series of periodic iodide separations was made, and the decay curve of each extract was followed. The iodide activity was found to consist almost entirely of a 2.4-hour body together with a small fraction possessing a half-life of 8 days. The results of these extractions give a decay curve of the parent 77-hour tellurium (Fig. 4).

The 77-hour body with its 2.4-hour daughter constitute a pair of activities which were considered to be "transuranic" elements. The genetical relationships given by Hahn, Meitner and Strassmann[15] list the 77-hour period as the daughter of a 59-minute body. This assignment was investigated thoroughly and found to be in error. The experiment was conducted as follows: A 100-gram sample of uranyl chloride was activated for 30 minutes with neutrons. The salt was dissolved in 3 normal hydrochloric acid, and 100 milligrams of tellurium in the form of tellurous acid were added. To sustain antimony 750 milligrams of antimony chloride were added. Tellurium was precipitated at once by passing sulfur dioxide into the boiling solution. A series of precipitations of tellurium was made at 10-minute intervals with 100 milligrams of the carrier being added before each precipitation. Decay curves of the active tellurium precipitates were followed and the relative amounts of the 77-hour activity were found both by analysis of the beta-decay curves and by a measurement of the x-rays. The results are given in Table I. It is evident that the parent of the 77-hour tellurium must have a half-life of much less than the 59-minute value given by Meitner, Hahn and Strassmann. In fact, the apparent half-life of the longest-lived ancestor is 5 or 6 minutes. Moreover, any lack of completeness of the precipitation of the tellurium would lengthen the apparent half-life of the parent rather than shorten it. Weighing of the tellurium precipitate showed that the precipitations were practically complete.

TABLE I. *Decay of active tellurium precipitates.*

	TIME AFTER START OF IRRADIATION	ACTIVITY OF 77-HOUR COMPONENT
Conclusion of irradiation	30 min.	
1st tellurium precipitation	40 min.	3.4 div./sec.
2nd tellurium precipitation	50 min.	0.25 div./sec.
3rd tellurium precipitation	60 min.	0.07 div./sec.
4th tellurium precipitation	70 min.	0.02 div./sec.

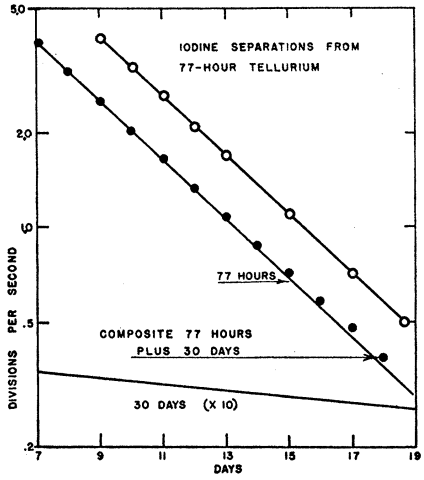

FIG. 4. Determination of half-life of 77-hour tellurium.

It is established, then, that the 77-hour activity is formed from a body whose quantity decays with a half-life of about five minutes. It must be borne in mind that this does not mean that the parent antimony is necessarily a 5-minute body. On the other hand, for instance, a 5-minute tin might decay into a shorter-lived antimony. On making the successive tellurium separations, one would find an apparent half-life of 5 minutes for the parent of the 77-hour activity. However, a 5-minute antimony activity has been found which probably is the parent of the 77-hour substance.

It is quite possible that the 2.4-hour body is the final active member of a chain of beta-decays. There are three stable xenon isotopes—132, 134 and 136—which could terminate the series of activities. If the 2.4-hour body has a daughter substance, the half-life of that activity must be greater than several months or shorter than one minute. The evidence for the above statement follows: If one makes a speedy iodine separation from the 77-hour activity, one finds that the 2.4-hour activity decays following a simple exponential law into the background. A sample of iodine whose intensity was twenty divisions per second was placed on the electroscope three

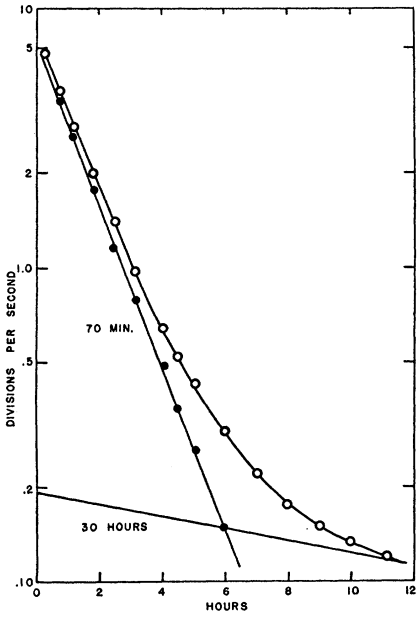

FIG. 5. Decay curve of tellurium separated from parent antimony activities showing the 70-minute daughter of a 4.2-hour activity.

minutes after the beginning of an iodine separation. If the half-life of the daughter were in the range from one minute to 100 days, the curve would not have been a simple exponential.

THE 70-MINUTE ACTIVITY

Among the active fragments of uranium fission is an antimony isotope whose half-life is 4.2 hours. If an activated uranium sample is allowed to age for six hours after termination of bombardment and then antimony is separated, the activity curve shows a growth. The intensity reaches a maximum in two hours. This indicates the growth of a tellurium daughter whose half-life is of the order of an hour. Separation of tellurium from the antimony revealed that the tellurium had a half-life of 70 minutes (Fig. 5). The determination of the parent half-life in the antimony is complicated by the presence of other long-lived antimony isotopes which themselves produce tellurium isotopes. Hence, the most accurate determination of the half-life of the 4.2-hour body is obtained through making periodic extractions of tellurium from antimony and then following decay curves of the separated tellurium fractions to determine the amount of seventy-minute body present at the time of extraction. The results are shown in Fig. 6. The equality of the value of the 70-minute half-life to that found by Seaborg, Livingood and Kennedy[16] for the lower isomer of Te^{129} suggested that the 70-minute body produced from uranium might be Te^{129}. Absorption curves of the beta-rays emitted by both of the tellurium isotopes have been obtained. The known Te^{129} was prepared from a sample of tellurium which had

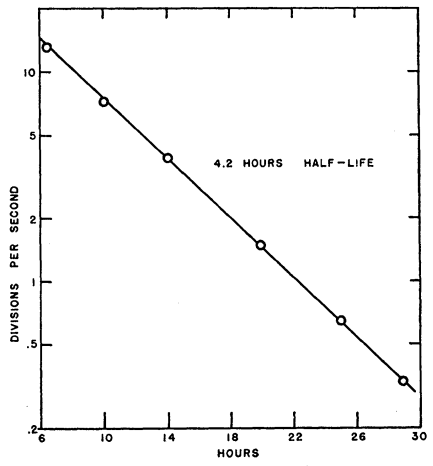

FIG. 6. Decay of a 4.2-hour antimony activity established through periodic tellurium separations of the daughter substance.

been bombarded by deuterons. A mixture of the 70-minute Te^{129} and the 10-hour Te^{127} was obtained separately from the parent isomers of 30-day and 90-day half-lives by means of an extraction devised by Seaborg, Livingood and Kennedy.[16] A beta-ray absorption curve was taken immediately after separation. The decay curve of the mixture of 70-minute and 10-hour bodies was followed and the fraction of each present thus determined. Fifteen hours after the isomer separation with the 70-minute body completely gone a second beta-ray absorption curve was followed. The characteristics of the beta-rays of the 10-hour body were thus obtained. The true absorption characteristics of the 70-minute body were then found by correcting the first absorption curve for the effect of the presence of the 10-hour activity. Actually this correction was very small because the 10-hour

[16] G. T. Seaborg, J. J. Livingood and J. H. Kennedy, Phys. Rev. **55**, 794 (1939).

body had an activity only five percent of that of the 70-minute body. The absorption curves of the 70-minute bodies obtained from both uranium and tellurium are shown in Fig. 7. It is seen that the two curves are parallel over a factor of 100 in intensity. It seems probable then that the two 70-minute bodies are identical.

The 10-Hour Activity

Another cleavage product of uranium which can be identified with a known activity is the 10-hour tellurium. Beta-ray absorption curves of this body and of the lower Te^{127} isomer[16] are identical. These curves are shown in Fig. 7. The 10-hour substance is obtained as the daughter of an 80-hour antimony isotope. If antimony is

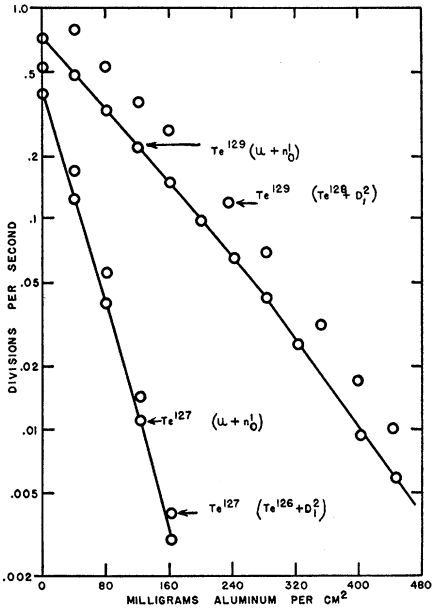

FIG. 7. Beta-ray absorption curves of 70-minute Te^{129} and 10-hour Te^{127}.

precipitated from an active sample which has been allowed to age for a week, the decay curve shows a growth which indicates the production of a daughter substance whose half-life is of the order of 10 hours (Fig. 8). On separating tellurium from the active antimony a 10-hour tellurium body was obtained. Furthermore, a series of tellurium precipitations showed that the 10-hour daughter was growing from an 80-hour parent (Fig. 8). No experiments have yet been made to determine the parent of the 80-hour antimony.

The 8-Day Activity

Still another fragment of uranium disintegration which may be identified with a known activity is an 8-day iodine body. The decay curve of this substance is shown in Fig. 9. An 8-day iodine activity can be produced through deuteron or neutron activation of tellurium and has been shown to be I^{131}.[16] Several lines of evidence indicate that the two 8-day iodine bodies are identical. Beta-ray absorption curves have been made on both bodies. These are identical. Further evidence is the fact that both of the 8-day iodines were found to emit characteristic xenon x-rays. It has previously been shown that I^{131} emits a gamma-ray of 0.4-Mev energy. Corresponding to any given geometrical arrangement of a source of I^{131} with respect to the ionization chamber, a certain value of the ratio of the gamma-ray to x-ray ionization was observed. When a source of the 8-day body obtained from uranium was placed in the same geometrical configuration as that of the I^{131}

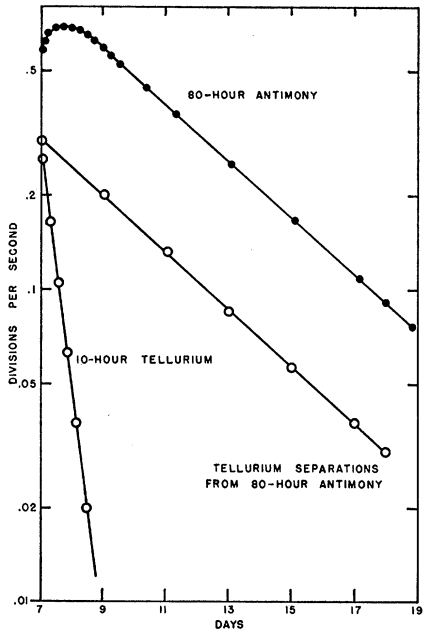

FIG. 8. Decay of 10-hour tellurium daughter extracted from 80-hour antimony and growth of activity in the 80-hour mother substance. The half-life of the antimony activity was also determined by periodic tellurium extractions of its daughter substance.

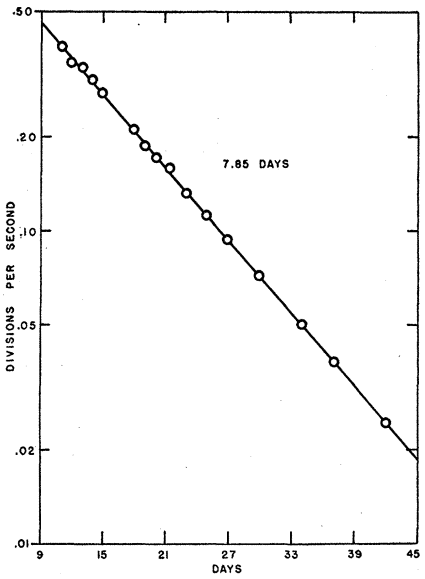

FIG. 9. Decay curve of 8-day iodine.

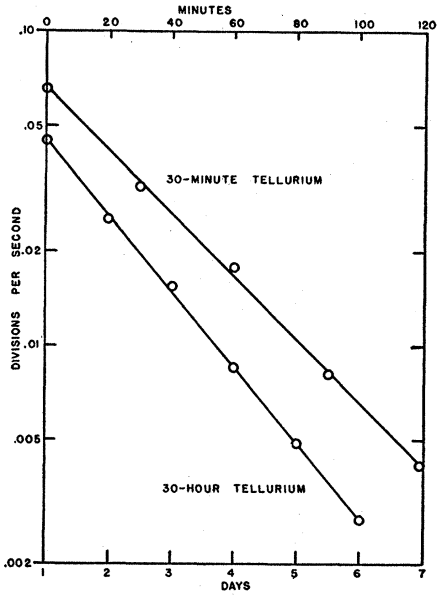

FIG. 10. Determination of half-lives of tellurium parents of 8-day iodine through periodic separations of iodine from tellurium.

identical values of the ratio of gamma-ray to x-ray intensities were found.

It has been shown[16] that Te^{131} possesses two isomers of 30-hour and 25-minute periods with the higher energy state belonging to the 30-hour body. Activities of these same half-lives have been found in tellurium precipitated from activated uranium. The periods of these parent tellurium activities were established by making a series of periodic iodine extractions from the precipitated tellurium. The results are shown in Fig. 10. In both cases the first iodine extract was discarded because some iodine could be carried down with the original tellurium precipitate. These experiments show that a 30-hour and a 30-minute Te^{131} are formed by neutron irradiation of uranium. Presumably these bodies are identical with those produced by activation of tellurium itself.

Two other tellurium periods were demonstrated through the technique of repeated iodine separations from tellurium which had been precipitated from neutron activated uranium. The decay curve of a typical iodine extract is shown in Figs. 11 and 12. This curve can be analyzed into two main components of 54-minute and 22-hour half-lives. A series of such iodine separations was made at 30-minute intervals and the activity of each extract was followed. The

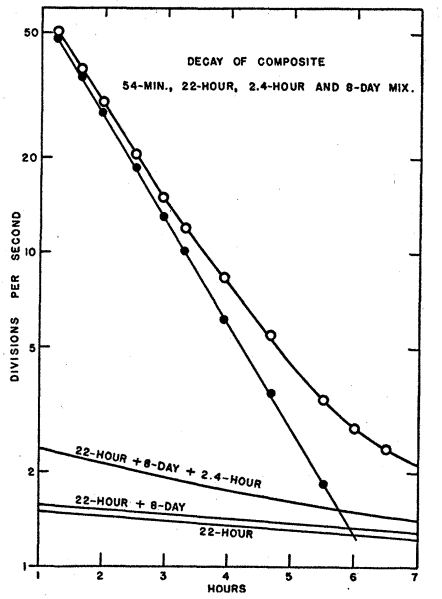

FIG. 11. Decay of a typical mixture of iodine activities separated from tellurium. The solid dots correspond to the 54-minute activity.

presence of the 2.4-hour and the 8-day bodies was taken into account. This is especially important in the case of separations made several hours or longer after bombardment. From these extractions it was determined that the 54-

minute iodine grows from a 43-minute tellurium and that the 22-hour iodine is the daughter of a 60-minute tellurium. Results are shown in Fig. 13. An experiment was made to determine the half-lives of the parents of the two tellurium activities. A series of tellurium precipitations was made at 10-minute intervals from an activated uranium sample. After an hour had elapsed these precipitates were dissolved and iodine extracted from them. The first two tellurium separations were discarded. Examination of the iodine activity removed from the third and subsequent precipitates revealed that neither the 22-hour nor the 54-minute body was present. This indicates that the grandparents (if any) of these two iodine substances possess a half-life less than 10 minutes.

A glance at the tellurium activities which have been found reveals four whose half-lives are so

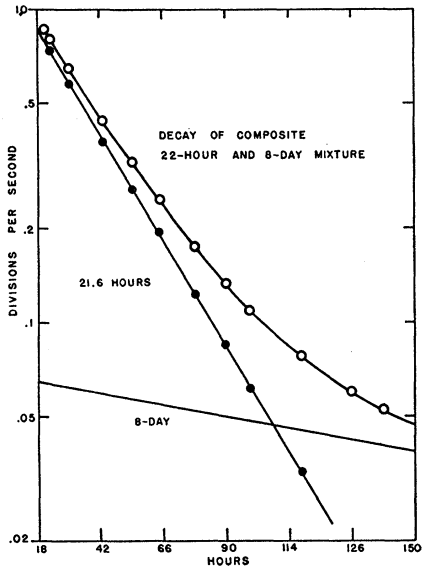

FIG. 12. Decay of a typical mixture of iodine activities separated from tellurium. The 54-minute and the 2.4-hour activities have practically disappeared.

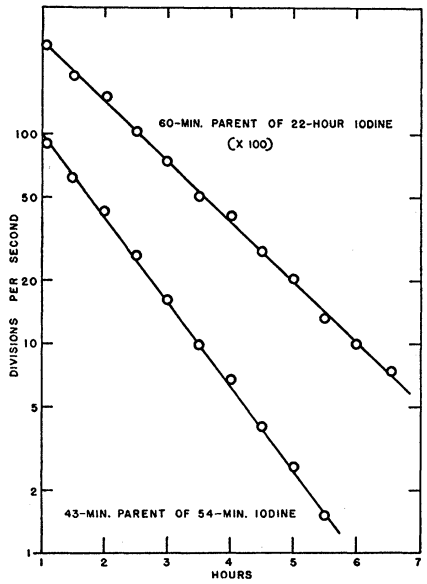

FIG. 13. Determination of half-lives of the 43-minute and 60-minute tellurium bodies through periodic separations of their daughter iodine activities.

close together as to be unidentifiable when lumped together in a single decay curve. The technique of periodic separations of daughter substances has proved successful in solving this problem. However, in order to follow decay curves of the extracts it is necessary to have large initial intensities. The writer feels that the accomplishment of this work is due in large measure to the huge neutron intensities of the cyclotron which have been made available to him.

ACKNOWLEDGMENT

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