1 of 1

NOTICE

**CERTAIN DATA
CONTAINED IN THIS
DOCUMENT MAY BE
DIFFICULT TO READ
IN MICROFICHE
PRODUCTS.**

DECLASSIFIED

HW- 3-3366

- #1 D.A. Miller - T.M. Stapleton - M.H. Smith - 700
- #2 D.A. Miller for The Area Engineer
Attention: Patent Group
- #3 J.N. Tilley - Wilmington
- #4 L. Squires
- #5 W.C. Kay - F.B. Vaughan
- #6 J.D. Ellett - W.K. MacCready
- #7 M.P. Asken - G.E. Desetti
- #8 G.W. Struthers
- #9 W.H. Sullivan
- #10 R.S. Apple - E.R. Gilbert - J.B. Work
- #11 300 Area File
- #12 300 Area File
- #13 Pink Copy
- #14 Yellow Copy

FEB 26 1990

JANUARY 8, 1946

SE-FC-481

Classification Change

DECLASSIFIED

FISSION PRODUCT STUDIES WITH PLANT DISSOLVER SOLUTIONS

IX - Iodine
X - Arsenic

By Authority of T.D. 1161

Oct 6 11/15/57
By J. H. K. 12/5/57

Introduction

In continuing studies to characterize and identify fission product activities formed in the piles and appearing in plant dissolver solutions, as outlined in report SE-FC-475, work was done to determine whether or not long-lived isotopes of certain elements were present. Two of the elements in this category were iodine and arsenic. The iodine isotope of particular interest was I¹²⁹. Its tellurium parent has been identified but no daughter activity attributable to iodine has been observed. The arsenic activities of interest were the long-lived isotopes (16 days and 90 days) which have been produced artificially by several different nuclear reactions.

Summary

An iodine sample, which was collected from the stack gases in a scrubber column and which at the end of bombardment contained more than 40 curies of 8day I¹³¹ as based on an extrapolation of the experimental decay curves, was found to contain only ~30 counts/minute of a long-lived activity after 371 days of decay. From calculations of the lower limit for the half-life of isotope I¹²⁹ ($>3.8 \times 10^9$ years), it was concluded that the observed residual activity was not due to an iodine activity but to some impurity. It was concluded also that the beta radiations associated with the I¹²⁹ activity must be very soft (less than 0.1 Mev) or are not observable.

No long-lived arsenic activity with a half-life of 50-400 days was found to be formed in fission with a yield greater than $5 \times 10^{-6}\%$. From this value, as an upper limit, it is calculated that the yield for the direct formation of the 90 day As^{72,73} in fission is at least 20 times less than that expected for the fission fragment most probably formed (having a Z value nearer 29).

MAS

DECLASSIFIED

DISTRIBUTION RESTRICTED TO U.S. ONLY

Experimental Work

IX - Iodine

DECLASSIFIED

A - Summary of Previous Work

Ever since the discovery by Jentschke⁽¹⁾ of an orderly relation between the fission yield and mass number of fission fragments, there has been considerable interest in the problem of finding radio-isotope I^{129} , which should occur with a yield of $\sim 1.2\%$. Also, the fact that two preceding members of the mass number 129 decay chain are well known (2,3) has contributed to speculations concerning its existence.

On the Metallurgical Project several attempts have been made previously to place limiting values on the half-life for the isotope but only very few of the results have been recorded in the project literature. The most recent available reference was found in report CS-2124. Here, the half-life was estimated to be 10,000 to 16,000 days.

Since the opportunity existed at HEW to accumulate large amounts of iodine activity present in the stack gases, the present investigation was undertaken to find the I^{129} activity and/or re-estimate the lower limit for its half-life.

B - Chemical Operations for the Isolation of Iodine

In order to obtain an adequate amount of fission product iodine, as a source of any very long-lived iodine activity which might be present, a scrubber column was set up in the stack gas monitoring building of the T-Area Plant. A small fraction of the stack gases were drawn continuously through an alkaline sodium bisulfite solution in the glass ring-packed column whenever dissolving operations were in progress. The scrubber operated with interruptions during February and the first week of March. During this period, three tons of 26gt metal, pushed November 20, 1944, and three tons of 44gt metal, pushed December 20, 1945, were dissolved for processing. The scrubber solution, when brought to the laboratory a month after cessation of operations, showed a ~ 20 mr/hr reading at ~ 3 inches.

The solution was allowed to stand until July 28, 1945 at which time the iodine activity was isolated from the sample and purified by the procedure given in report CM-1312, p. 56. The silver iodide precipitate as finally isolated was too active for counting measurements, but portions of it were studied to determine the decay rate and the beta ray absorption characteristics in aluminum.

C - Experimental Results

The physical measurements showed that the activity decayed with an 8 day half-life and that the aluminum absorption curve was indistinguishable from that reported to be characteristic of 8.0 day I^{131} . (Memorandum MUC-NS-#230). The several smaller samples continued to show a uniform 8 day decay rate until August 24, 1945 by which time the main sample had decayed sufficiently to be counted. The decay curve for this sample after September 20, 1945 (first shelf counting measurements on Eek and Kribs counter) is shown in Figure 1. Graphical resolution of the complex curve gives an 8 day half-life for the shorter-lived component. An absorption curve for the long-lived activity showed that a soft beta radiation, with a $\sim 10\text{mg/cm}^2$ Al half-value thickness, was present.

- (1) Jentschke, Zeit. f. Physik 120, 165-84 (1943)
- (2) Seaborg, Livingood, and Kennedy, Phys. Rev. 57, 363 (1940)
- (3) Abelson, Phys. Rev. 55, 1 (1939)

DECLASSIFIED

D - Discussion of Results

From the above experimental data a lower limit for the half-life of isotope I¹²⁹ may be calculated, assuming that the residual activity is due to iodine and that the radiation characteristics of isotope I¹²⁹ are similar to those observed for the residual activity.

On September 20, the counting rate for the main sample was 1010 c/m, with 960 c/m being due to the 8 day I¹³¹ and ~50 c/m, to the long-lived activity. After correcting for absorption and geometry, it was found that there were $\frac{960 \times 1.25}{0.27} = 4470$ d/m of 8 day I¹³¹ and $\frac{\sim 50 \times 2}{0.27} = \sim 220$ d/m of long-lived iodine (I¹²⁹). From the value, 4470 d/m, it was calculated that the disintegration rate of the 8 day I¹²⁹ isotope at the end of bombardment, after correction for 274 days (34.2 half-lives) of decay, was $4470 \times 2^{34.2} = 8.9 \times 10^{13}$ d/m or a total of more than 40 curies.

To calculate the half-life of isotope I¹²⁹ from the above data, the relation

$$T_{1/2} (I^{129}) = \frac{Y (I^{129}) \times dN/dt (I^{131}) \times T_{1/2} (I^{131})}{Y (I^{131}) \times dN/dt (I^{129})}$$

was used. Here, $T_{1/2}$ is the half-life of the activity in days; Y, the fission yield; and dN/dt , the disintegration rate per minute for the two isotopes concerned, as indicated parenthetically. Accordingly, the lower limit for the half-life of I¹²⁹ was calculated to be $\frac{0.012 \times 8.9 \times 10^{13} \times 8}{0.027 \times 220} = 1.4 \times 10^{12}$ days or $> 3.8 \times 10^9$ years.

In considering the meaning of this result, it must be remembered that the assumption was made initially that the radiation characteristics of I¹²⁹ are similar to those observed for the long-lived residual activity. On this basis, one may conclude that the observed activity was not due to isotope I¹²⁹, but to some impurity, since a $> 3.8 \times 10^9$ year half-life would indicate that the isotope should occur naturally, reasoning by analogy from the natural occurrence of uranium (4.5×10^9 years half-life) and the estimated 10^9 to $> 2 \times 10^9$ years age of the earth (4) (to account for decay of the isotope after formation).

Assuming that the observed activity is due to an impurity, then one can conclude only that the long-lived I¹²⁹ isotope has a range of less than 10 to 15 mg/cm², since this is the limit of detection under the particular experimental conditions employed (total absorption from counter window, air, cover, and the sample itself was ~ 2mg/cm²).

However, since it is known that isotope I¹²⁹ must be formed in fission and artificial transmutations, the problem of finding the activity evolves in a search for very soft radiations, if they exist. It is possible, of course, for the isotope to have no detectable beta radiations, due to the possibility that they never escape from the field of orbital electrons after emission from the nucleus. In this latter case, neutrinos would be the only detectable radiation.

• In these rather crude calculations, corrections for the decay of I¹³¹ during bombardment and for the 30 day difference in cooling time for some of the metal, which was pushed November 20, were not taken, since the relative amounts of iodine from the two batches of metal were not known.

(4) Mier, Phys. Rev. 55 153 (1939)

I - ArsenicA - Summary of Previous Work

The most recent work to find long-lived arsenic isotopes produced in fission was described by Winsberg in report CG-2310. His results indicated that there were no long-lived arsenic activities with half-lives between 3 days and 1000 years with a fission yield of greater than 10^{-4} percent.

Since it was considered possible that the 90 day $As^{72,73}$ and 18 day As^{74} isotopes might be found directly in fission with a very low yield, the investigation was initiated.

B - Chemical Operations for Isolating Arsenic

Arsenic carrier (along with other carriers as indicated in Table 1) was added to 3 ml. of 120gt UNH solution. A CeF_3 precipitation was made, then boric and perchloric acids were added and the ruthenium and rhenium carriers distilled out. Sulfuric acid was then added and distillation was continued, while passing in $HCl-HBr_2$ mixture, in an effort to distill out the Sb, As and Sn. Most of the tin appeared to distill over in this operation, but very little antimony and arsenic were found in the distillate. The residue from the distillation was diluted and neutralized to about 1N H_2SO_4 with NaOH. Alpha-benzoin oxime was then added to precipitate molybdenum and palladium. After removing the precipitate, the residual solution was treated with H_2S , which precipitated a mixture of sulfides. The sulfide precipitate was dissolved in 6N NaOH and the insoluble portion removed. The material soluble in NaOH was precipitated with HCl, then redissolved in concentrated HCl. This solution was treated with concentrated H_2SO_4 and SO_2 , then heated to $180^\circ C$ to distill off the As, Sb, and Sn. Arsenic was separated from the other two elements as As_2S_3 by passing H_2S into the strongly acid distillate. It was further purified by one distillation from concentrated HCl at about 110° in the presence of antimony, tin and tellurium hold back carriers. The arsenic was finally isolated as As_2S_3 . A chemical yield of 6.7% was obtained.

C - Experimental Results and Discussion

A total of 240 c/m (1st shelf on standard counter assembly) was observed 76 days after the end of bombardment. The activity was not identified. However, from this observed counting rate an upper limit for the fission yield could be calculated.

Assuming that the observed activity is due to arsenic and that absorption of the beta radiations can be neglected in the rough calculations, one finds a total of $\frac{204}{5.08 \times 10^{-2} \times 205} = 12,500$ disintegrations/minute at the time of measurement. From this value, fission yields were calculated, using the different half-life values given in Table 2. In calculating these results, corrections were made for growth and decay during bombardment and 76 days of decay after bombardment.

DECLASSIFIED

HW-3-3360

-5-

1-3-48

Table 1

Procedure Used for Isolating Arsenic From 150gt Uranyl Nitrate Solution

Source: 3ml of 40% UREN 120gt. pushed from pile 3/8/45. Add Mo, Sn, Sb, Re, Pd, Co, Ru and As carriers. Dilute. Add HF

Ppt. CoF ₃ (Set aside)	Solution Add boric acid and HClO ₄ . Distill										
	Distillate Ru and Re (Set aside)	Residue Add H ₂ SO ₄ Distill at 180°C while passing in HCl-HBr mixture									
		Distillate Sn and some Sb	Residue Neutralize to 1N H ₂ SO ₄ with NaOH. Add o-benzoic-acid								
			Ppt. Mo & Pd (Set aside)	Solution Warm and pass in H ₂ S							
				Sol'n. Discard	Precipitate Sulfides of Sb, As, also some Sn and Re. Add NaOH						
					Ppt. (f) Dis- card	Solution Add HCl until slightly acid					
						Sol'n. Discard	Ppt. Dissolve conc. HCl. Add H ₂ SO ₄ . Distill.				
							Residue Discard	Distillate As, Sn, Sb In conc. HCl. Add H ₂ S			
								Sol'n. Sn, Sb (Set aside)	Ppt. As ₂ S ₃ . Dis- solve HNO ₃ . Evap- orate to dryness. Add HCl, SO ₂ . Distill		
									Residue Dis- card	Distillate Add H ₂ S	
										Ppt. As ₂ S ₃ Mount for counting	

DECLASSIFIED

HW-3-3366

-6-

1-8-48

Although the assumptions made initially for the calculations summarized in Table 2 may or may not be valid, the results suffice to show that it is quite unlikely that a long-lived arsenic activity is formed in fission, especially as the result of chain decay from fission fragments most likely to be formed initially. If any such long-lived arsenic activity is formed, it must have a very long half-life and/or very soft radiations.

For the known 16 day As^{74} and the 90 day $As^{72,73}$ isotopes, which must be formed directly in fission because their potential Ge parent isotopes are stable, the $10^{-6}\%$ fission yield is an upper limit, since the chemical identification of the activity as arsenic was not absolutely rigorous.

M.F. ACKEN, CHIEF SUPERVISOR
200 AREAS TECHNICAL DIVISION

W.H. SULLIVAN-SENIOR SUPERVISOR

Gordon R. Leader / WHS
Gordon R. Leader - Pres. Chem.

GRL:j4

DECLASSIFIED 3-3366

INV.

9-47

Figure 1

Decay Curve for the Iodine Activity
Obtained from the Stock Cases

DATE

FILMED

6 / 9 / 94

END