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CHEMISTRY DIVISION

Section C-IV

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Waldo E. Cohn, Section Chief

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PREPARATION OF CARRIER-FREE 13.8 DAY Pr^{143}

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Bernard J. Finkle and Waldo E. Cohn

* * *

September 1, 1945

This report covers work between periods

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[redacted]

Bombardment of Ce with neutrons yields two radioisotopes by n, γ reaction: 28 day Ce^{141} and 33 hour Ce^{143}. These decay by β emission,

$$Ce^{141} \xrightarrow[28\ d]{} Pr^{141} \text{ (stable)}$$

$$Ce^{143} \xrightarrow[33\ h]{} Pr^{143} \xrightarrow[13.8\ d]{} Nd^{143} \text{ (stable)},$$

leaving 13.8 day/Pr as the only active non-Ce isotope produced. Since the irradiation of pure Ce requires only the separation of the Pr from its Ce parent to produce pure carrier-free Pr, this reaction offers a simple method of preparing Pr tracer. Ce can be separated from the other rare earths (valence: + 3) by oxidation to the + 4 valence and precipitation as iodate.

A quantity of CeO_2 was repurified from Baker and Adamson $Ce(NO_3)_3$ by several $Ce(IO_3)_4$ precipitations from ~ 4 N HNO_3. Each precipitation was made by oxidizing the Ce^{+3} with $NaBrO_3$ in 16 N HNO_3 and then adding 0.35 M HIO_3 dropwise. Following each precipitation the $Ce(IO_3)_4$ was reduced with hydroxylamine in 6 N HCl and reprecipitated as $Ce(OH)_3$. The last $Ce(OH)_3$ precipitate was dissolved in a minimum of HCl, precipitated as $Ce_2(C_2O_4)_3$, and ignited to CeO_2.

0.3 gm of the purified CeO_2 was irradiated in the Clinton pile (hole 21) for 14 days (4.95×10^4 Kwd) and set aside to allow the 33 hour Ce^{143} to decay completely to its daughter Pr^{143}. The irradiated oxide was dissolved in a small amount of hot 6 N HCl solution containing equal amounts of hydroxylamine hydrochloride and KI and was diluted with water.

An aliquot equal to 0.0005 of this solution was carried through the following procedure: To the active solution was added 20 mg Ce^{+3} carrier, 8 ml concentrated HNO_3, and ~ 2 gm $NaBrO_3$; it was then heated to the very first appearance of fumes, at which time 20 ml of 6 N HIO_3 was added dropwise with vigorous stirring, precipitating $Ce(IO_3)_4$. Approximately 5 mg of Ce^{+4} carrier (as $(NH_4)_2Ce(NO_3)_6$ dissolved in 5 ml 4-5 N HNO_3) was added dropwise with vigorous stirring over a period of

five minutes to the supernatant, again precipitating $Ce(IO_3)_4$. This last step was repeated several times to give a total of five precipitations. Following the fifth $Ce(IO_3)_4$ precipitation, ~5 mg Fe^{+3} carrier was added to the supernatant and brought down as $Fe(OH)_3$-$Pr(OH)_3$ with excess NH_4OH. The hydroxide was dissolved in 2 ml 6 N HCl, diluted to 20 ml with water, and reprecipitated with excess NH_4OH. This step was repeated (total: 3 $Fe(OH)_3$ precipitations). The last hydroxide was washed with water and dissolved in 8 N HCl. Fe^{+3} was completely extracted with isopropyl ether. The aqueous layer was evaporated to dryness several times with aqua regia to eliminate NH_4Cl, leaving solid-free Pr^{143}.

The purified Pr^{143} was taken up in water and evaporated in a capsule. An Al absorption curve was taken (Fig. 1). This curve consisted of Pr^{143} contaminated to the extent of 3% (at 0 added absorber) by a hard component, which, when subtracted, gave the typical Pr^{143} curve (cf. MUC-NS-230).

The chemical yield on the carrier-free material was 49% of the total Pr^{143} present, obtained by comparison of the carrier-free Pr activity with that of a sample (Fig. 2) whose yield was known from chemical (carrier) analysis. (Data in Table.)

Calculation of Yield of Carrier-free Pr

	Pr Act., 8/6/45	Aliquot of Total Soln.	Counted Act. per Total Soln.	Chem. Yield	Calculated Yield
Pr (Pr-carried)	4146 c/m	2.0×10^{-4}	2.073×10^{7} c/m	65.2%	-
Pr (carrier-free)	7740 "	5.0×10^{-4}	1.548×10^{7} "	-	48.6%

From the following derived equation*, the maximum activity obtainable from an infinite irradiation under the same conditions is 9.1 mC Pr^{143}/gm CeO_2:

$$A^{max}_{Pr} = A^{T,t}_{Pr} \frac{\lambda_{Pr} - \lambda_{Ce}}{\lambda_{Ce} e^{-\lambda_{Pr} t}\left(e^{-\lambda_{Pr} T} - 1\right) - \lambda_{Pr} e^{-\lambda_{Ce} t}\left(e^{-\lambda_{Ce} T} - 1\right)}$$

*Rutherford, Chadwick and Ellis, Radiations from Radioactive Substances, Chapter 1, equations (16) and (19). Also derived independently by W.E. Cohn.

where A_{Pr} = Pr^{143} activity, corrected for chemical yield, aliquot and counter geometry, (= 1.76 x 10^9 d/m), T = time of irradiation (= 14 days), and t = time after irradiation end (= 27 days).

The 3% contamination of the carrier-free material (probably originating from neutron reaction on an impurity) could possibly be eliminated by irradiating more highly purified Ce.

This reaction can be used to make high levels of Pr activity with minimum safety precautions, since the only other radiations present after 33 h Ce^{143} has been allowed to decay are the weak γ-ray (0.2 Mev) and the 0.6 Mev β^- of 28 d Ce^{141}; 14 days after the end of a 14-day bombardment the total activity of the irradiated CeO_2 was only six times that of the active Pr^{143} it contained.

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Fig. 1. Carrier-free 13.8 d Pr^{143} - Al Absorption Curve

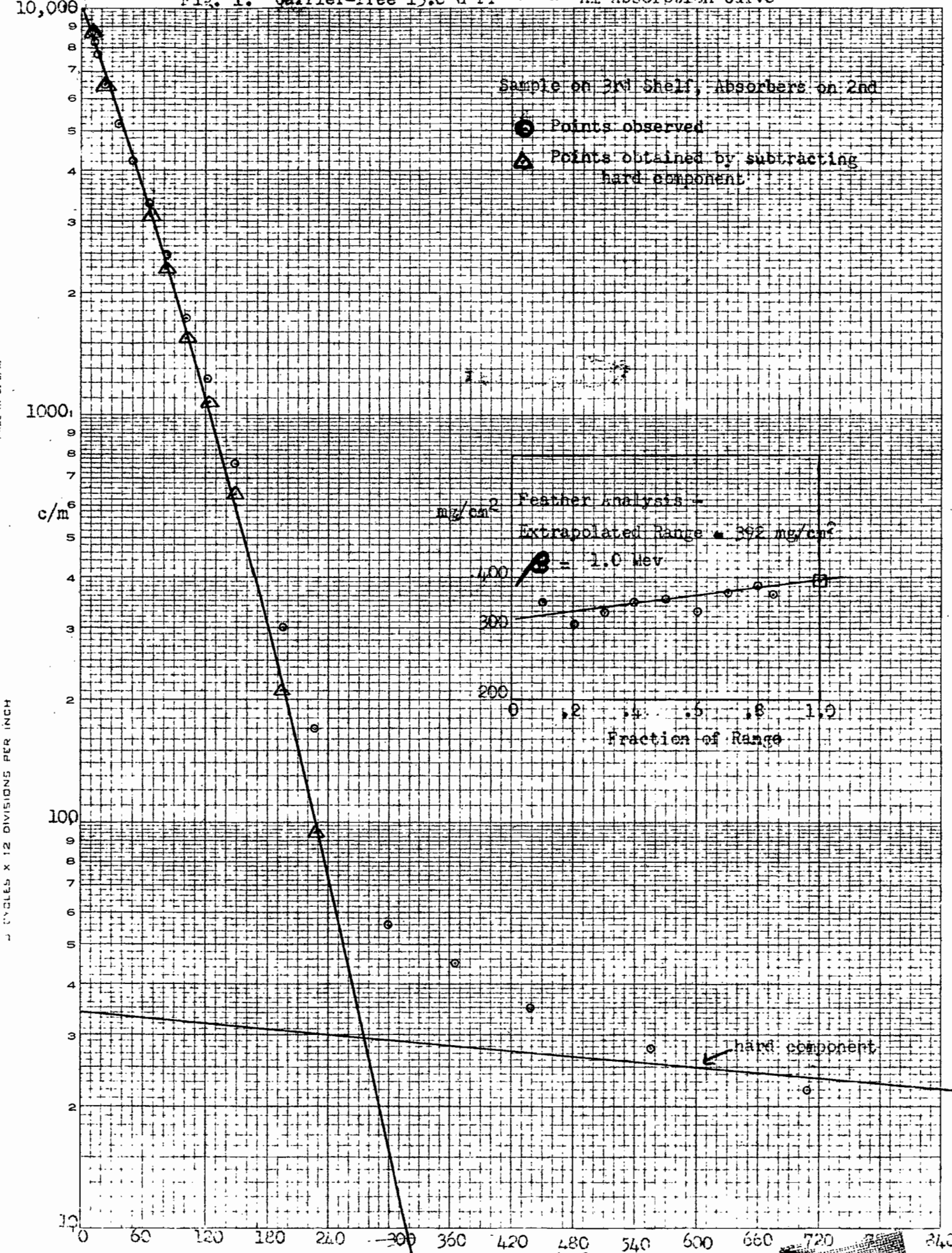

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Fig. 2. Pa-carried 1β,8 d Pr^{143} - Al Absorption Curve