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A 2800°C CALUTRON ION SOURCE UNIT AND ITS USE IN SEPARATING THE ISOTOPES OF Pd, Pt, Ru, AND Ir

W. A. Bell, Jr.
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July 30, 1954

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STABLE ISOTOPE RESEARCH AND PRODUCTION DIVISION
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ABSTRACT

A calutron source capable of heating small (ten gram) quantities of charge material to approximately 2800°C has been developed. Design features are discussed and separations of isotopic samples (Pd, Pt, Ru, Ir) made possible by use of this source are summarized.

A 2800°C CALUTRON ION SOURCE UNIT AND ITS USE IN SEPARATING THE ISOTOPES OF Pd, Pt, Ru, AND Ir

W. A. Bell, Jr., L. O. Love, and C. E. Normand

INTRODUCTION

Electromagnetic equipment assigned to the Stable Isotope Research and Production Division at Y-12 has been used in separating relatively large quantities of isotopes of elements other than uranium, and the ultimate goal of the Stable Isotope program is the separation of each naturally occurring atomic species, not already 100 percent abundant.

Early calutron separations were made of those elements for which charge materials suitable for use in the existing ion sources were readily available. As separations progressed, the search for charge material broadened and in time the sources were modified to permit higher operating temperatures of charge containers and associated source components. An ion source capable of attaining temperatures of 1000°C was designed late in 1949. Its use permitted separation of the isotopes of the available rare earth elements, but it was inadequate for the platinum and palladium groups of metals for which no satisfactory calutron charge was known. This report describes briefly the source developed for use in vaporizing metals of the platinum and palladium groups and summarizes the results achieved in the first calutron separations of palladium, platinum, ruthenium, and iridium.

EQUIPMENT

Ion Source

Various methods by which high temperature charges could be vaporized and fed into an arc had been proposed, and a limited amount of exploratory work had been performed with three of the most feasible of these methods. These included: 1) placing a metallic charge at the anode end of the arc and allowing it to be bombarded with low energy electrons; 2) placing a metallic charge back of the arc and applying a voltage such that the metal was subjected to sputtering action; and 3) heating a charge container by high energy electron bombardment. In the first two methods, the amount of material vaporized was some function of arc conditions and the ion output was low and unstable. The third method was more direct, but initial tests[1] revealed extreme difficulties in temperature control.

A survey of the characteristics of the platinum and palladium groups of metals indicated that heating the charge container by high energy electron bombardment would probably be the most practical method of obtaining the required temperatures. The existing 1000°C source was, therefore, modified to incorporate this type of heating. In making the necessary modifications, it was recognized that the unit should be: 1) capable of operating with good temperature regulation between 1500° and 3000°C; 2) compact, with a minimum of exposed radiating surface, thus reducing the required power input; 3) capa-

---

1. Tested by W. R. Rathkamp in 1947, following a suggestion made by C. B. Graham.

ble of operating satisfactorily with a very small charge, because of the high cost of the four metals to be used in the process; 4) so designed that recharging would not present a major servicing problem as is the case with the 1000°C units now in use; and 5) so constructed as to permit the use of support gas in either or both the collimating slot and the arc chamber.

In fabrication of the new unit, the conventional charge bottle and vapor manifold were eliminated by machining into a single rectangular block of graphite an arc chamber and a charge cavity capable of holding about ten grams of charge material, as shown in Figure 1. This graphite "source block", placed in alignment with the magnetic field, was bombarded by electrons emitted from a 170-mil tantalum filament placed opposite one end of the block. A carbon grid structure (Figure 2) was located between the filament and the source block. Adjustment of the filament-grid bias gave control of the bombarding electron current, and the number and location of grid openings determined the disposition of bombarding electron beams on the source block. In a final design of the unit, adequate electron control was obtained with the following potentials: ion source block, 35 kv positive; grid, grounded; filament, 400 v positive. Filament-to-grid spacing was .020-in. Emission current varying from 50 to 300 ma could be controlled quite adequately by adjustment of the filament-grid bias. Power for heating the bombardment filament was supplied by an Alpha-2 filament transformer, with provisions made to operate the filament at any fixed voltage between 0 to 1000 volts above ground. A block diagram of this circuit is shown in Figure 3. The chief difficulty encountered

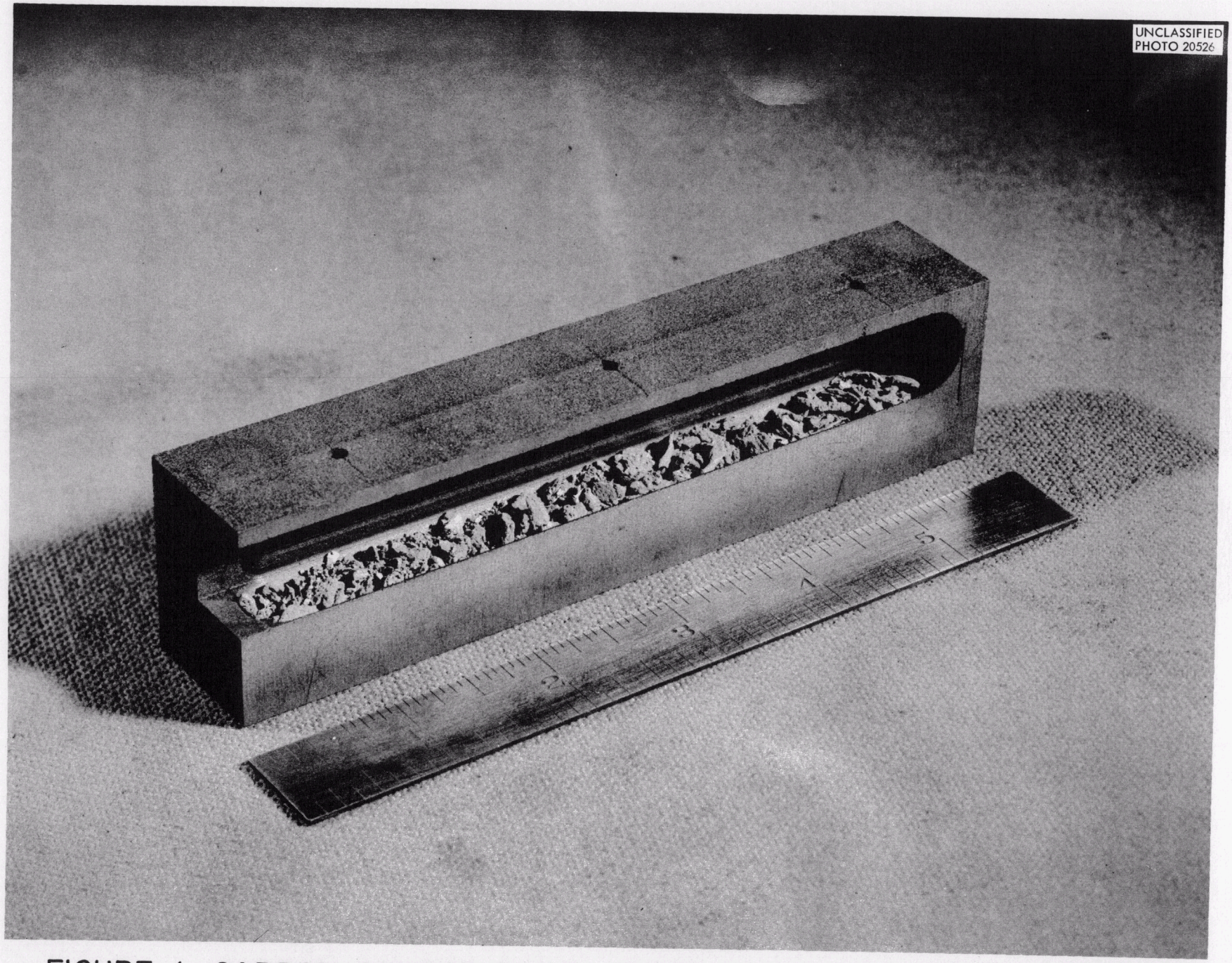

FIGURE I. CARBON SOURCE BLOCK CONTAINING PALLADIUM METAL.

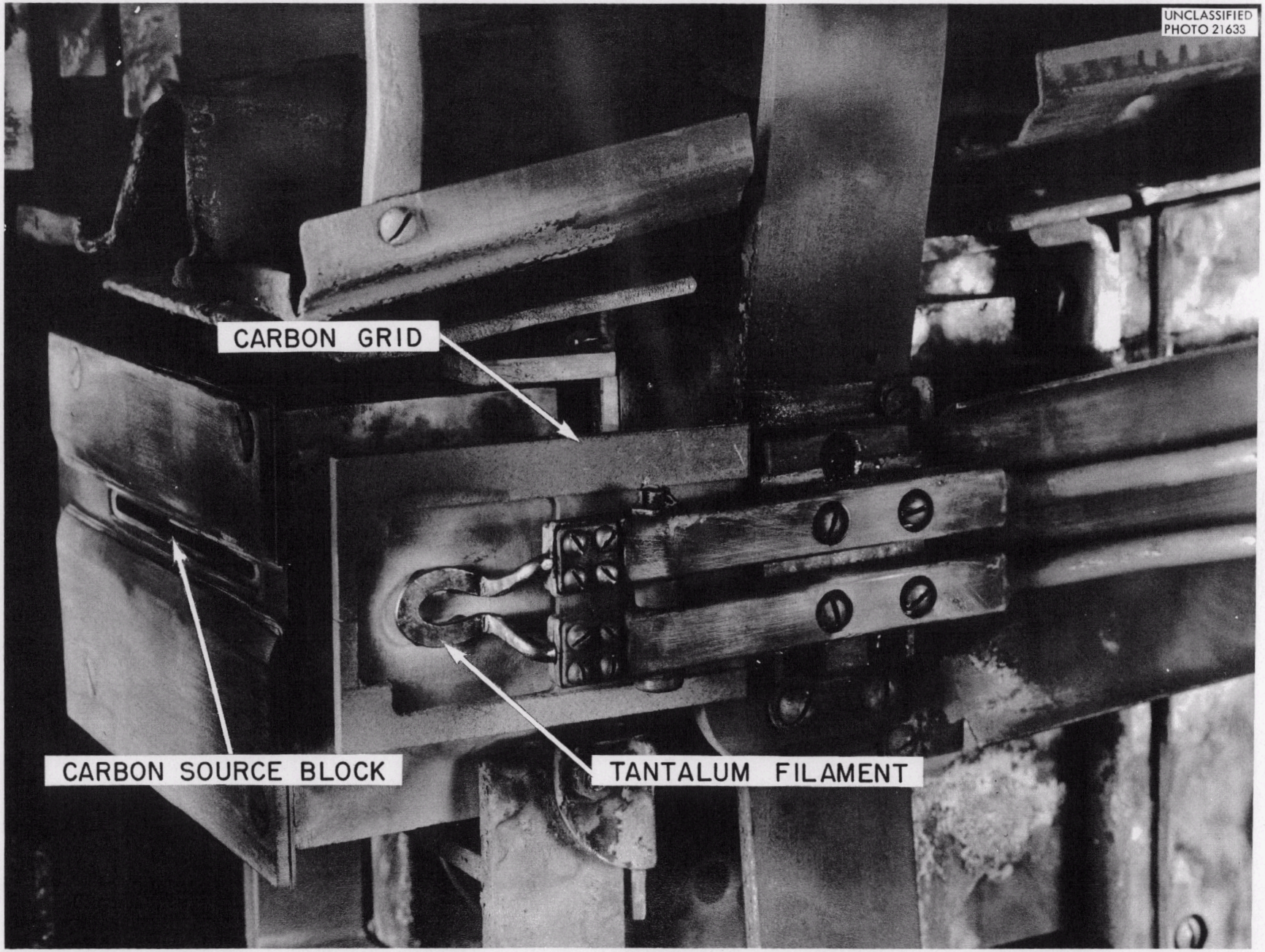

FIGURE 2. COMPONENTS OF THE BOMBARDMENT HEATER.

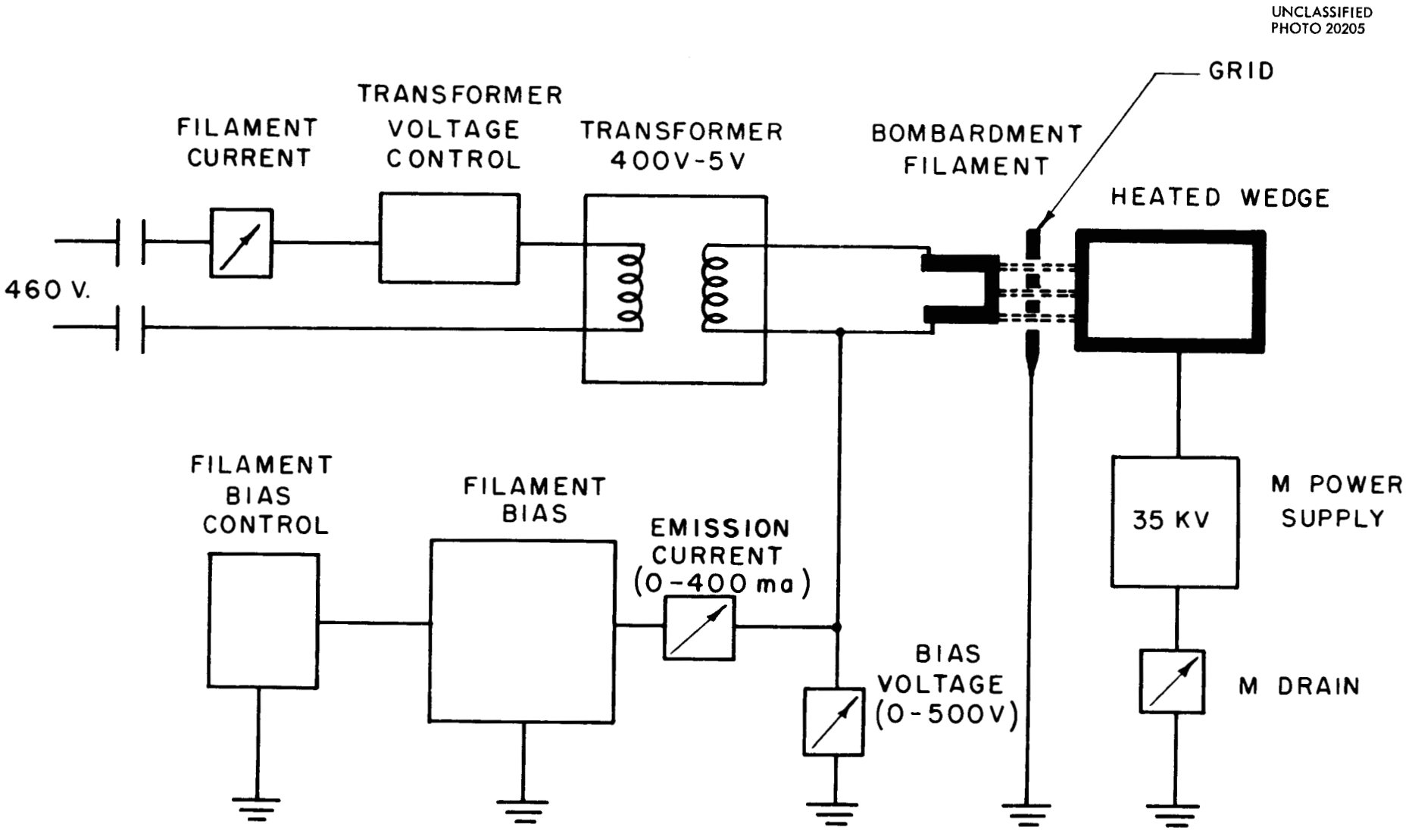

FIGURE 3.

BLOCK DIAGRAM OF HEATER SYSTEM USED IN OBTAINING CALUTRON CHARGE TEMPERATURE IN EXCESS OF 2000° C.

during operation was that of maintaining a constant grid-to-filament spacing. Increasing this spacing limited the emission current obtainable while decreasing it rendered current control difficult.

Several distinct types and arrangements of arc chamber and charge cavity were used, as well as different dispositions of bombarding electron beams. Source blocks of three types are shown in Figure 4. The evolution from Type A to Type C was dictated by the increasing temperatures required to vaporize successive elements. In Type A, the charge was placed in the arc chamber directly beneath the arc, and the block was heated by a single broad stream of electrons impinging directly back of the arc chamber and charge. Type B was the result of an effort to apply more heat to the arc chamber as well as to the charge, since in Type A heavy deposits of charge material had formed on the inner surface of the arc chamber cover, adjacent to the ion exit slit. This cover was not an integral part of the heated block in either Type A or Type B and it presented a cool surface to charge vapors. In Type C, the ion exit slit is made an integral part of the block, and the arc chamber and charge compartment are connected cylindrical cavities. The four electron bombarding beams are equally spaced around the charge compartment. With this arrangement the deposition of charge near the slit was materially reduced, the useful life of the block was increased, and because of more efficient heat transfer to the charge, shielding problems were somewhat simplified.

Along with changes in the source block and in disposition of the bombarding electron beams went changes in the extent and type of heat shielding applied. Graphite or carbon, in the form of thin sheets and, later, sheets in

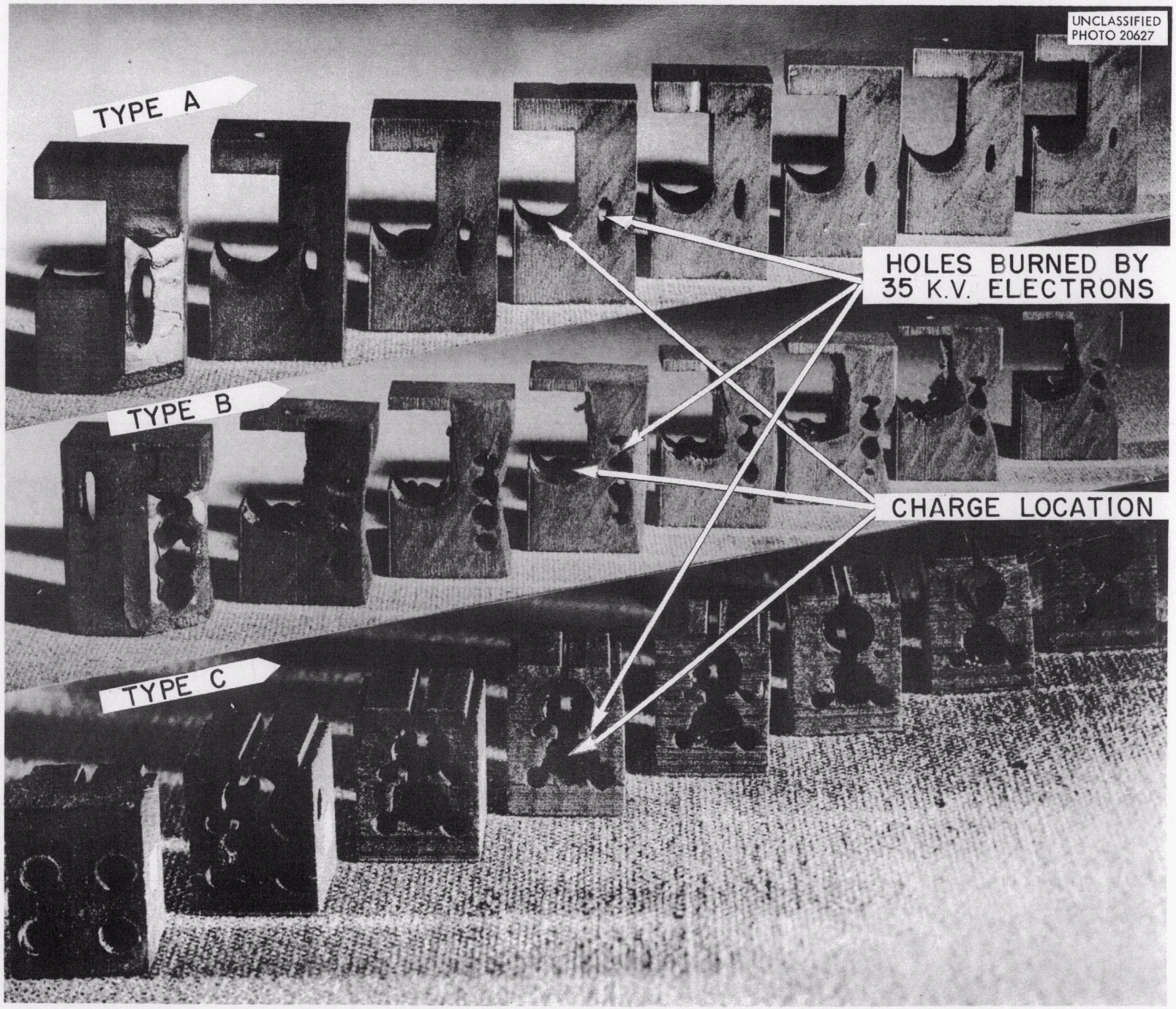

FIGURE 4. CROSS-SECTIONS OF CARBON SOURCE BLOCKS

combination with graphite powder, were used as heat barriers around the source block. Two source blocks, with shielding in place but with front covers removed, are shown in Figure 5. One of these assemblies with heat-shielded cover in place is shown in Figure 6, while Figure 7 shows details of the source block and heat shielding assembly. Figure 8 is a picture of the source showing the relative position of the ionization filament and the bombardment filament.

Receivers

The vapor pressure of the four metals processed was such that the isotopes could be collected in graphite receiving pockets placed behind 1/8" defining slots. Receivers for the two isotopes of iridium presented no problems; however, space limitations between adjacent isotope beams made the omission of certain pockets necessary in the other three collections. Thus, two types of receivers were used for each element. One palladium receiver collecte Pd-110, Pd-108, Pd-105, and Pd-102, omitting pockets for Pd-106 and Pd-104. The other receiver collected all isotopes except Pd-105. In like manner Pt-198 Pt-195, Pt-192, and Pt-190 were collected while Pt-196 and Pt-194 were rejected. Then collection of Pt-196, Pt-194, and Pt-190 was made while rejecting Pt-198, Pt-195, and Pt-192. In both platinum receivers a pocket was put in the 200 mass position for exploratory purposes. With ruthenium, one receiver collected all isotopes except Ru-100 while the other omitted pockets for Ru-101 and Ru-99. In cases where pockets were omitted, the rejected beams were stoppe by the carbon receiver faceplate.

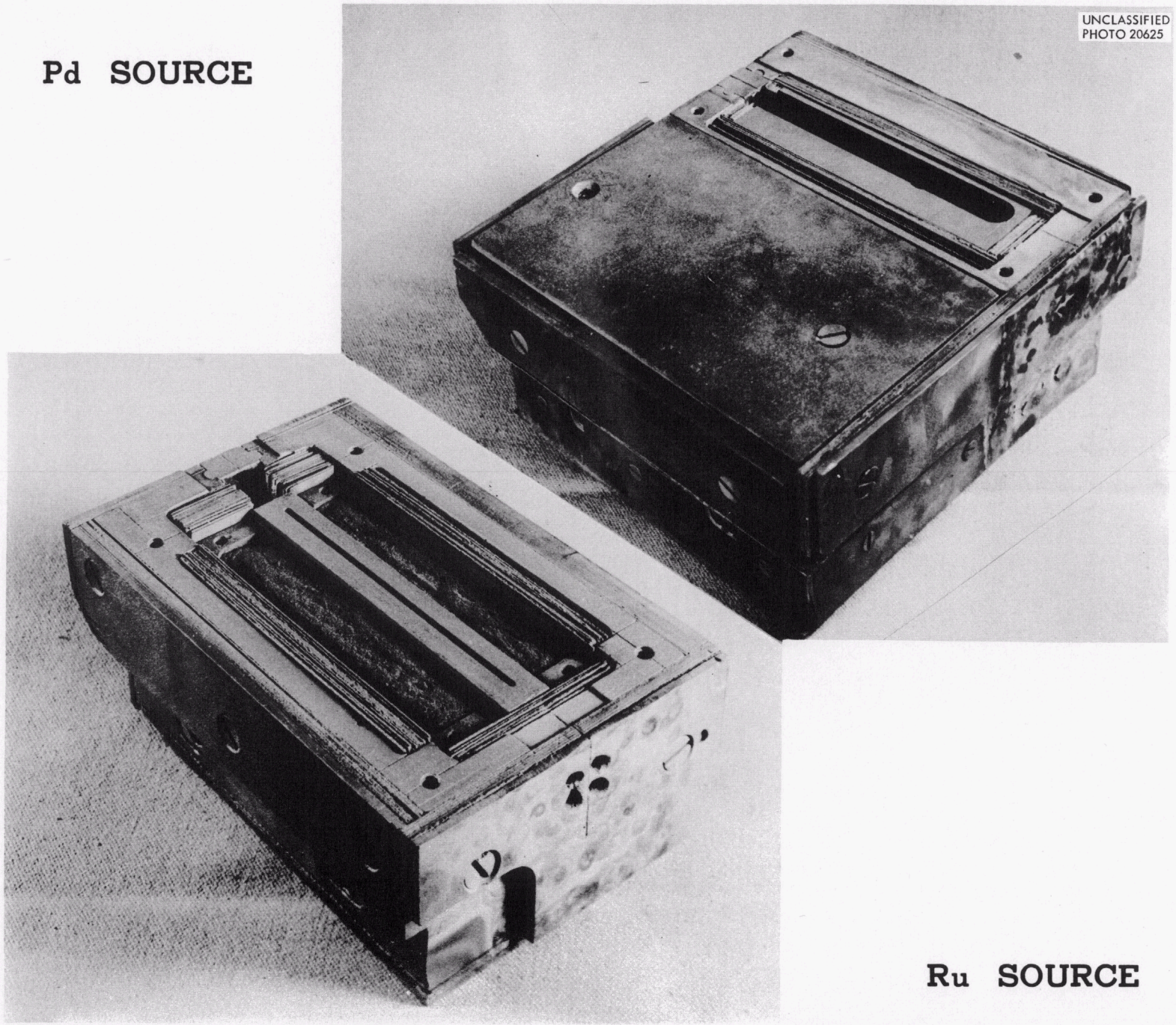

FIGURE 5. SOURCE BLOCKS SHOWING HEAT SHIELDING.

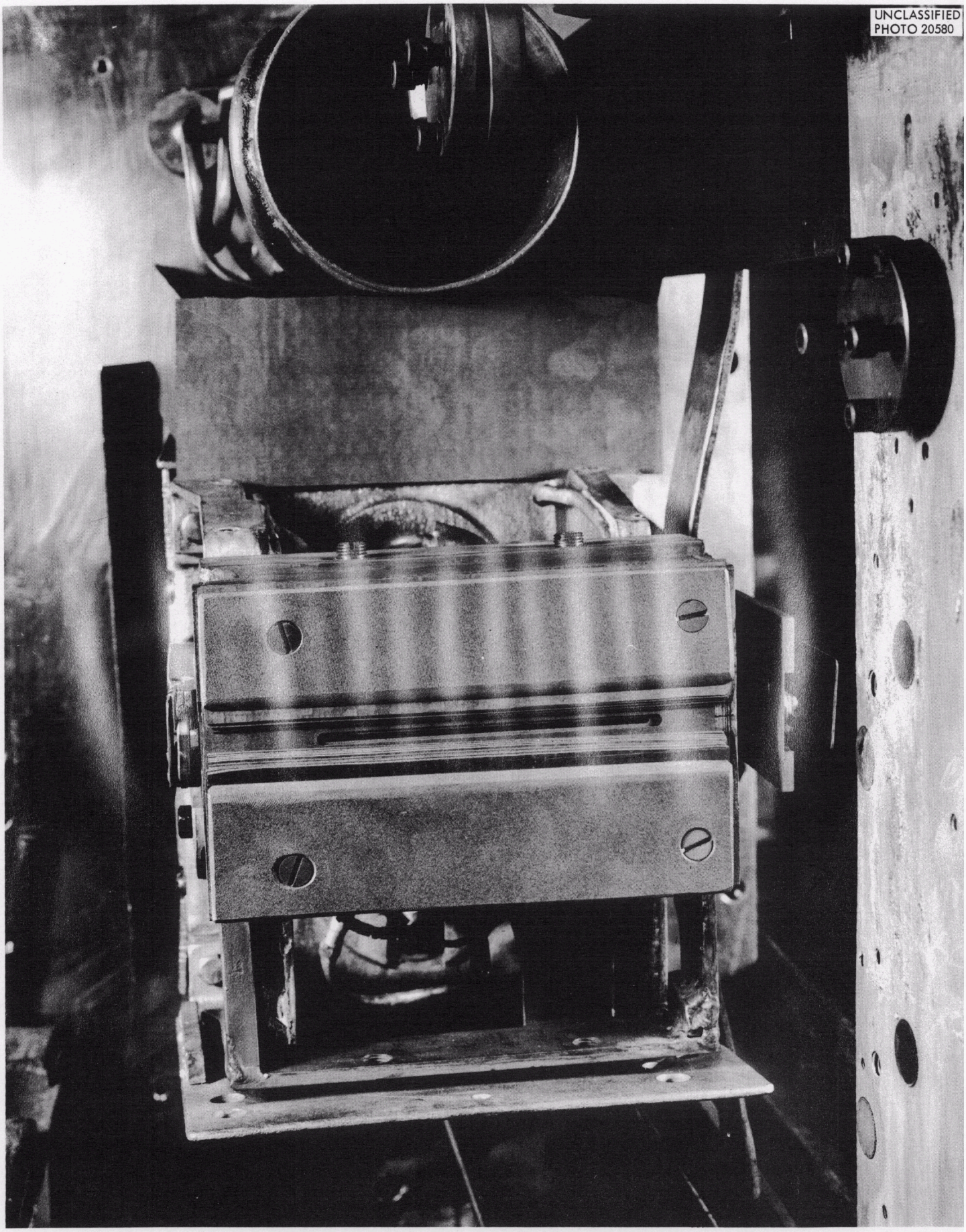

FIGURE 6. COMPLETED SOURCE BLOCK MOUNTED ON THE SOURCE ASSEMBLY.

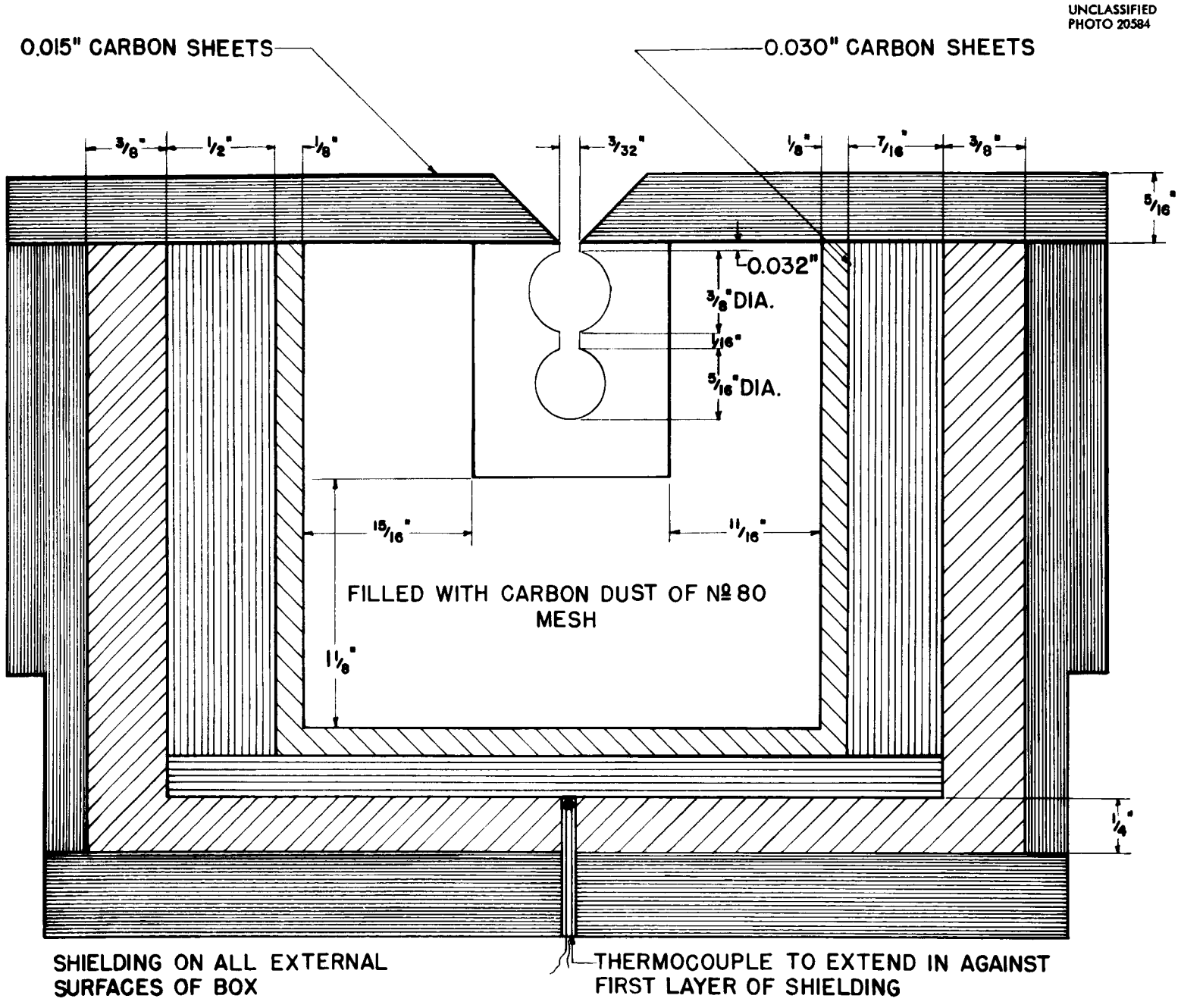

FIGURE 7. DETAILS OF SOURCE BLOCK AND HEAT SHIELDING.

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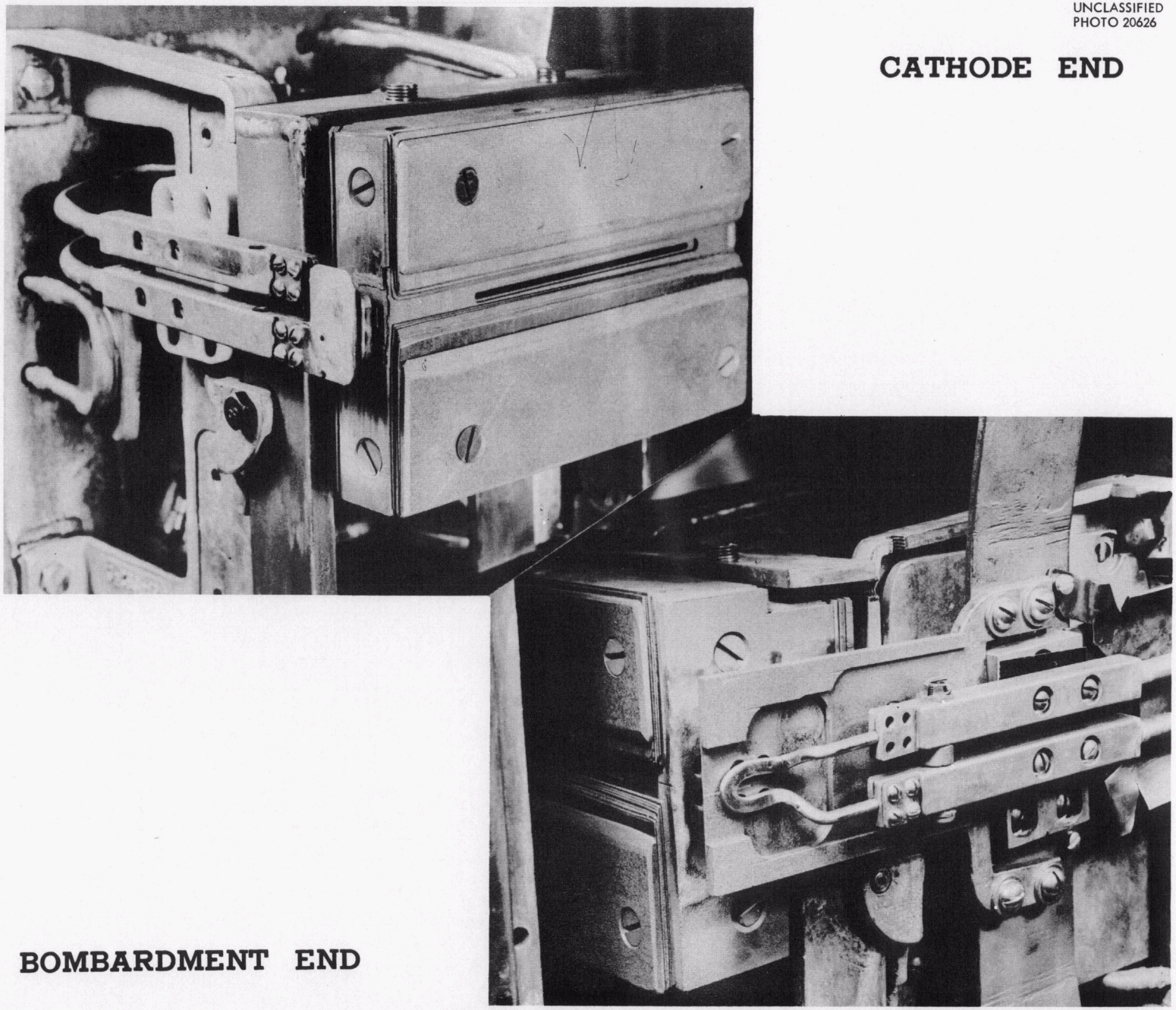

FIGURE 8. BOMBARDMENT HEATED SOURCE UNIT SHOWING FILAMENT LOCATIONS.

Liner

The usual Beta stainless steel liner was used in these collections. It was water-cooled, condensing out charge vapors before they reached the vacuum pumps, thus aiding in recovery of valuable charge materials. Sufficient baffling was provided to protect equipment from sideband beams produced by support gases.

OPERATION

The bombardment-heated sources required close attention during operation. Whenever accelerating voltages tripped out as the result of high drains or sparking, the unit cooled very rapidly until all output was lost. Likewise, slight variations in bombardment filament current or filament-to-grid bias were reflected immediately, either as cooling or overheating of the source block. Periodic increases in bombardment current were required to maintain production as the charge became depleted.

Heat conductivity of the graphite charge container was such that a localized hot spot appeared at the point of electron impact on the source block. Charge vaporization occurred chiefly at this hot spot and progressed slowly along the length of the arc chamber as erosion proceeded. This localized vaporization produced an arc of uneven density along its length. This effect was noticeable at the receiver as the position and intensity of the received beam varied with run time.

In operation, the arc was supported at all times by nitrogen introduced at the collimating slot. A second gas line was used initially to supply chlorine to the arc chamber, with the thought that chlorine pick-up might permit operation at a lower temperature. No such effect was observed. However, the introduction of either nitrogen or chlorine into the arc chamber did improve beam focus.

The four elements (Pd, Pt, Ru, and Ir) were processed in order of increasing melting point. Operation summaries for the elements arranged in this same order are given in Table I. Total collection of isotopes of each of the elements indicated from monitored currents are given in Table II. Tables III through VI give recovery, receiver retention, and mass assay data for the isotopes collected.

TABLE I

SUMMARY OF OPERATIONS

Element	Total Runs	Runs Attaining Innage[1]	Maximum Innage Per Run (Hrs.)	Average Innage Per Run (Hrs.)	Peak Output (Ma)	Avg. Output per Inn. Hr. (Ma)	CCR[2] (Gm/Hr)	PE[3] (%)	Est. Oper. Temp. °C
Pd	92	75	18	6.0	37.2	9.2	0.9	4.2	1750
Pt	160	129	22	8.5	12.8	3.7	1.2	2.4	2250
Ru	95	89	29	9.1	11.0	2.4	0.7	1.4	2600
Ir	115	97	26	9.7	7.8	0.5	0.6	0.6	2800

1 Innage means ion reception.
2 Charge Consumption Rate. (Grams of element per hour of innage).
3 Process Efficiency. (Est. collection/wt. of element lost from charge container).

Note: Due to the erosion of the source block by bombarding electrons the Charge Comsumption Rate and Process Efficiency were calculated by assuming that all the charge installed was used.

TABLE II

ESTIMATED COLLECTION OF ISOTOPES

Palladium		Platinum		Ruthenium		Iridium	
Isotope	Est. Wt.[1] (gm)	Isotope	Est. Wt. (gm)	Isotope	Est. Wt. (gm)	Isotope	Est. Wt. (gm)
110	2.286	200[2]	0.251	104	1.613	193	2.124
108	5.712	198	0.728	102	3.003	191	1.343
106	4.277	196	11.357	101	1.082	Total	3.467
105	1.704	195	4.299	100	0.445		
104	2.264	194	12.779	99	0.740		
102	0.432	192	0.296	98	0.234		
Total	16.675	190	0.258	96	0.416		
		Total	29.968	Total	7.533		

1 Est. Weight - calculated on the basis of monitored currents only.
2 Isotope not known to exist. Pocket installed for exploratory purposes.

TABLE III

SAMPLES PRODUCED AND MASS ASSAY
Pd, Series GG-XBX

Sample	Isotope	Est. Wt. Collected (gm)	Wt. Chemically Recovered (gm)	Receiver Retention (%)	Natural Abundance[1] (%)	Mass Assay of Sample (%)[2] 110	108	106	105	104	102
775a	Pd^{110}	2.286	1.7484	76.5	13.5	91.4	7.3	0.9	0.4	0.1	-
774a	Pd^{108}	5.712	4.0269	70.5	26.8	2.1	94.2	2.8	0.6	0.2	0.03
773a	Pd^{106}	4.277	2.9673	69.4	27.2	0.4	4.6	82.3	11.3	1.2	0.06
772a	Pd^{105}	1.704	1.1475	67.3	22.6	0.2	1.8	16.0	78.2	3.8	0.04
771a	Pd^{104}	2.264	1.8327	81.0	9.3	1.4	2.8	9.5	23.2	63.2	-
770a	Pd^{102}	0.432	0.3583	82.9	0.8	0.7	3.9	11.9	22.0	26.3	35.2
	Total	16.675	12.0811	72.5	100.2						

1 M. B. Sampson and W. Bleakney, Phys. Rev. 50, 732 (1936).
2 ORNL Mass Spectrometry Laboratory.

TABLE IV

SAMPLES PRODUCED AND MASS ASSAY
Pt, Series GH-XBX

Sample	Isotope	Est. Wt. Collected (gm)	Wt. Chemically Recovered (gm)	Receiver Retention (%)	Natural Abundance[2] (%)	Mass Assay of Sample (%)[4] 200	198	196	195	194	192	190
782a	Pt^{200}[1]	0.251	0.2798	111.5	-	-	-	-	-	-	-	-
781a	Pt^{198}	0.728	0.4977	68.4	7.23	-	61.0	26.5	9.0	3.6	0.04	-
780a	Pt^{196}	11.357	6.9895	61.5	25.4	-	1.1	65.9	26.3	6.7	<0.2	-
779a	Pt^{195}	4.299	3.2157	74.8	33.7	-	0.5	11.6	60.1	27.8	<0.05	-
778a	Pt^{194}	12.779	12.0060	94.0	32.8	-	0.4	5.8	28.6	65.1	0.2	-
777a	Pt^{192}	0.296	0.2789	94.2	0.78	-	0.4	10.3	22.6	52.8	13.9	-
776a	Pt^{190}	0.258	0.3446	133.6	0.012[3]	-	2.9	15.6	29.6	45.3	5.8	0.76
	Total	29.968	23.6122	78.8	99.922							

1 Isotope not known to exist. Pocket installed for exploratory purposes.
2 M. G. Ingram, D. C. Hess, Jr., and R. J. Hayden, ANL-4012 (August 1947).
3 W. T. Leland, Phys. Rev. 76, 992 (1949).
4 ORNL Mass Spectrometry Laboratory.

TABLE V

SAMPLES PRODUCED AND MASS ASSAY
Ru, Series GJ-XBX

Sample	Isotope	Est. Wt. Collected (gm)	Wt. Chemically Recovered[1] (gm)	Receiver Retention[1] (%)	Natural Abundance[2] (%)	Mass Assay of Sample (%)[1,3] 104	102	101	100	99	98	96
796a	Ru^{104}	1.613	-	-	18.3	-						
795a	Ru^{102}	3.003	-	-	31.3		-					
794a	Ru^{101}	1.082	-	-	17.0			-				
793a	Ru^{100}	0.445	-	-	12.7				-			
792a	Ru^{99}	0.740	-	-	12.8					-		
791a	Ru^{98}	0.234	-	-	2.2						-	
790a	Ru^{96}	0.416	0.3220	77.3	5.7	0.5	1.1	0.7	0.7	1.1	0.4	95.5
	Total	7.533			100.0							

1 Incomplete.
2 H. Ewald, private communication to S. Flugge and J. Mattauch, Ber. deut. chem. Ges. 76A, 1(1943); Z. physik 122, 487 (1944).
3 ORNL Mass Spectrometry Laboratory.

TABLE VI

SAMPLES PRODUCED AND MASS ASSAY
Ir, Series GL-XBX

Sample	Isotope	Est. Wt. Collected (gm)	Wt. Chemically Recovered (gm)	Receiver Retention (%)	Natural Abundance[1] (%)	Mass Assay of Sample (%)[2,3] 193	191
811a	Ir^{193}	2.124	1.1888	60.0	61.5	-	
810a	Ir^{191}	1.343	1.0053	74.8	38.5	14.1	85.9
	Total	3.467	2.1941	63.3	100.0		

1 M. B. Sampson and W. Bleakney, Phys. Rev. 50, 732 (1936).
2 Incomplete
3 ORNL Mass Spectrometry Laboratory.

CONCLUSION

This high temperature source definitely has usefulness in an isotope separation program, its value being established to some extent by the separations already accomplished through its use. Since both palladium and platinum separations were made during a stage of source development, it is believed the results described herein certainly could be improved upon. Both ruthenium and iridium operation, however, are believed to be about representative of that to be expected later. Here, charge temperatures were such that the vapor pressure of the graphite source block became a limiting factor in operation, creating high pressure areas in the electrode and bombardment filament regions. At these points, two effects were noted: 1) electrode drains were excessive and sparking was encountered, thus disrupting temperature control, and 2) deposits of graphite formed on the colder surfaces changing critical filament-to-grid spacings. Were it possible to substitute some lower vapor pressure material for the graphite being used, considerably higher temperatures and increased ion output could be obtained.

ACKNOWLEDGMENTS

The authors wish to acknowledge the efforts of W. K. Prater who designed and supervised the fabrication of the high temperature source. The production operations were under the supervision of C. V. Ketron, R. L. Caldwell, George Fisher and K. A. Spainhour; the electrical modifications and maintenance were under the supervision of George Banic.

The recovery of the unused charge material and the purification of the collected samples were done by W. C. Davis and Cliff Sheridan of the Stable Isotope Chemistry Department. The mass assays reported in this report were made by the ORNL Mass Spectrometry Laboratory under the supervision of Russell Baldock. The statistical data were compiled by Sue English.