PRODUCTION OF POLONIUM 208

AUGUST 1952

Final Report On Termination
Of Project

By

Robert S. Livingston
John A. Martin

OAK RIDGE NATIONAL LABORATORY
OPERATED BY
CARBIDE AND CARBON CHEMICALS COMPANY
A DIVISION OF UNION CARBIDE AND CARBON CORPORATION
OAK RIDGE, TENNESSEE

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ABSTRACT

Substantial progress has been made in the production of experimental quantities of polonium 208 in the ORNL 86-inch cyclotron. Predicted nuclear yields are now obtained at the increased energy of the cyclotron. The production rate is over 300% higher than in October 1951. Estimates of ultimate production costs in a multi-cyclotron installation now approach \$50 per curie.

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REVIEW OF PRESENT STATUS

The successful operation of the ORNL 86-inch proton cyclotron during 1951 led to the production of experimental quantities of the alpha emitter polonium 208.* In the spring of 1952 internal revisions of the cyclotron resulted in an increase in proton energy from ~ 19 Mev to ~ 23 Mev. At this higher energy the polonium yield more than doubled, increasing from about 4 mc/mah to more than 9 mc/mah. A careful re-examination of nuclear yield data then led to a complete removal of the discrepancies in yield factors previously reported; essentially full theoretical yields are now obtained. A polonium 208 production rate in excess of 0.9 curie/week has been attained.

The problems of shielding for gamma radiation and controlling airborne alpha contamination have been intensified by the higher beam energy and production rate. Exposure of personnel has been held below permissible levels by the use of remote handling equipment, gloved boxes, and controlled ventilation. Operation of the cyclotron at the very high dee-to-dee voltage, 400-450 kv, necessary for 23 Mev protons, required the installation of additional cooling and consequently delayed the return of the machine to production.

Extrapolation from careful measurements of proton beam, ion loading, and oscillator efficiency permits new and more reliable cost estimates for larger production machines. It is now believed to be entirely feasible to produce Po 208 at a cyclotron operation cost of between \$50 and \$70 per curie; a multi-cyclotron plant with a capacity of 9,000 curies per month can be constructed at

* Livingston, R. S., et. al., Production of Polonium 208 - October 1951, ORNL-1192, December 8, 1951.

a cost of 23 to 30 million dollars, depending upon the electrical efficiency. It is recommended that these new cost estimates be used in any comparison of alternative alpha emitters.

CYCLOTRON DEVELOPMENT

Operation of the 86-inch cyclotron was interrupted in mid-January, 1952, for modifications which would permit operation at higher energies, make it possible to use larger targets, and provide for better control of radiation hazards. Prior to that time a steady proton current of more than one milliamperes could be obtained at a maximum energy of ~ 19 Mev. With a change in the position and mounting of the ion source, the energy was increased to 23 Mev for a target at the 30.5" radius. The increased power input to the cyclotron then resulted in excessive rf heating in the spider and upper portion of the liner. Additional water cooling was installed in these parts, and the cyclotron now operates steadily with a current above one milliamperes at 23 Mev. The major changes in the cyclotron are described in Appendix A.

EXPERIMENTAL POLONIUM PRODUCTION

New Yield Data

Because target yields obtained during the fall of 1951 had been much lower than predicted, a careful review* of the $\text{Bi } 209(p, 2n)\text{Po } 208$ cross section data was made by E. L. Kelly of the Radiation Laboratory at Berkeley. He believes that the most reliable measurements were made with the Berkeley linear accelerator in his run of March 1950. New thick target yield curves, Figures 1 and 2, were then prepared. These new calculations reduce the expected yield about 10%.

* Private communication.

FIGURE 1. POLONIUM 208 YIELD (THICK TARGET)

FIGURE 2. EFFICIENCY OF USE OF ELECTRICAL POWER

Beam Integration

The accuracy of yield prediction depends necessarily on the validity of measurement of current to the target. The system of beam integration described in the October Report has in recent months been augmented by a calorimetric system based on the temperature rise in the target-cooling water. Integration of a chart of this temperature change provides a measure of the kilowatt hours to the target. This method has proved to be highly reliable and can be used with the more recent yield data to give accurate predictions of assayed yields.

Production Summary

The first runs after the shutdown for alterations were for the purpose of establishing the yield, the beam distribution, and the operating characteristics of the cyclotron. Subsequent runs served to demonstrate the increased production rates and to test the cyclotron for reliability. A summary of all runs made since October 1951 is given in Appendix B. The large increase in yield in April 1952 is due, of course, to the increase in beam energy.

The polonium production rate has increased steadily since the first production runs in May 1951, as shown in Figure 3. The present weekly production rate is over 300% greater than that reported in October 1951.

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PRODUCTION COST ESTIMATES

The latest experimental information on nuclear yields and electrical efficiency have been combined to bring up to date the tables previously prepared* on expected Po 208 costs. The revised figures, Table I, show that at higher accelerator currents the cost of isotope production becomes increasingly dependent on the efficiency of conversion of electrical power to beam power. In this revision, costs are estimated for three different electrical efficiencies, Cases A, B, and C as described in Table II. Considerable experimental information indicates that these efficiencies are possible but it should be emphasized that the practical achievement of the higher efficiencies cannot be demonstrated until a cyclotron is actually constructed to deliver currents in the range between ten and fifty milliamperes.

Gross Electrical Efficiency

Cases A and B are both based on a new measurement** of the ratio of cyclotron-beam power to dee-ion-loading power which was recently obtained in the 86-inch cyclotron. This study showed that somewhat more than 40% of the total ion loading rf power is in the beam intercepted by the target, and somewhat less than 60% in the stray ions which are wasted into the dees. If present overall efficiency of the oscillator (about 60%) and the power for rf

* ORNL-1192, p. 48, and Letter from R. W. Cook to C. E. Larson, May 9, 1952, containing Minutes of the Meeting in Washington, D. C. on April 2, 1952.

** Livingston, R. S. and Boch, A. L., The Oak Ridge 86-Inch Cyclotron, June 1952, ORNL-1196, p. 26.

TABLE I: REVISED COST OF PRODUCING POLONIUM 208Costs for Production with the 86-Inch Cyclotron

	<u>Energy</u>	<u>Current</u>	<u>Yield</u>		<u>Cyclotron Operation Costs (\$/curie)</u>
	<u>(Mev)</u>	<u>(ma)</u>	<u>mc/mah</u>	<u>%</u>	<u>86" Cyclotron</u>
October 15, 1951	19	0.680	3.1	61	18,800
January 15, 1952	19.7	0.820	4.4	75	10,800
July 20, 1952	23	1.0	9.4	97	4,750
Anticipated	23	1.85	9.4	97	2,450

Projected Costs for Production with Phase-Compensated Cyclotron (PCC)
and with Multi-Cyclotron Plant (MCP)

<u>Current*</u> <u>(ma)</u>	<u>PCC Operation Costs (\$/curie)</u>			<u>MCP Operation Costs (\$/curie)</u>			<u>MCP Production</u> <u>(curies/mo)</u>
	<u>Case A</u>	<u>Case B</u>	<u>Case C</u>	<u>Case A</u>	<u>Case B</u>	<u>Case C</u>	
7	497	483	465	288	274	256	1280
15	259	246	228	161	148	130	2740
50	114	100	83	84	71	53	9120

* Cyclotrons to operate at 27 Mev with 95% innage and 97% yield.

TABLE II: EXAMPLES OF RF POWER DISTRIBUTION
FOR EACH OF THREE CASES

	<u>Cases</u>		
	<u>A</u>	<u>B</u>	<u>C</u>
<u>Description of Cases</u>			
Oscillator Efficiency, %	60	80	80
Beam Power/Ion Load, %	40	40	75
<u>Examples</u>			
Ion Current (at 27 Mev), ma	15	50	50
<u>Power Distribution</u>			
DC Power Input, kw	1750	4250	2300
Oscillator Losses, kw	700	850	450
RF Power Output, kw	1050	3400	1850
Dee Excitation Power, kw	50	50	50
Total Ion Load, kw	1000	3350	1800
Stray Ion Power, kw	600	2000	450
Beam Power, kw	400	1350	1350
Gross Electrical Efficiency, %	20	30	60

losses are taken into account, an overall efficiency of about 20% results. This coincides approximately with the case of 15 ma considered in the October Report.*

In Case B the oscillator efficiency has been revised upward to take into account an oscillator circuit where attention has been given to improving efficiency. An oscillator efficiency of 80%, which is well established by present rf engineering practice for high Q circuits, is anticipated. The gross efficiency is then about 30%.

Case C takes advantage of the decreased stray-ion losses resulting from the use of improved dc-injection-type ion sources and the use of specially shaped focusing electrodes, both of which are now under development. It is expected that 75% of the total ion loading power will then be used in accelerating protons to the target; the gross efficiency would be approximately 60%. The distribution of rf power load for samples of these three cases is given in Table II.

Beam Current

Studies on cyclotron performance and beam attenuation in the ORNL 22-inch cyclotron** and analysis of the established characteristics of high intensity ion sources has led to the conclusion that cyclotron currents much higher than those estimated in the October Report are practicable. A cyclotron equipped with a dc-injection-type ion source, and with suitable power supplies but of otherwise conventional design, should deliver 50 to 80 ma of useful beam. Such an injector system would make it possible to (1) obtain greater ion output from the higher arc currents obtainable in this type of ion source, (2) maintain greater output because of the constant dc accelerating potential applied, and (3) reduce beam attenuation by the introduction of improved focusing electrodes.

* ORNL-1192, p. 15.

** ORNL-1339, pp. 15, 16.

A demonstration of a 50 ma cyclotron current is now being planned for the small test machine at this laboratory. A new type of ion source, yielding more than one ampere of ions per square inch of arc aperture, has been developed.* Although adaptation of this ion source to cyclotron operation has not been considered it would undoubtedly make still larger beam currents possible.

New Construction Costs

The new construction costs, Table III, were obtained by modifying the cost estimates of the October Report to provide for the greater capacity of the rf system required for the higher beam currents. The costs are tabulated for three different cases of which Case C shows that the higher electrical efficiency would result in a large reduction in the cost of power equipment.

* ORNL-1380.

TABLE III: COST ESTIMATESCost of Power Supplies

<u>Design Capacity</u>	<u>RF Supplies</u>		<u>DC Supplies</u>	
	<u>kw</u>	<u>\$/cyclotron</u>	<u>kw</u>	<u>\$/cyclotron</u>
Case A at 15 ma	1050	100,000	1750	91,700
Case B at 50 ma	3400	425,000	4250	200,000
Case C at 50 ma	1850	225,000	2300	110,000

Total Construction Costs

<u>Design Capacity</u>	<u>PCC</u> [*]	<u>MCP</u> ^{**}
Case A at 15 ma	\$1,766,600	\$19,237,100
Case B at 50 ma	2,201,700	30,716,700
Case C at 50 ma	1,882,700	23,060,700

* Phase-Compensated Cyclotron Facility, ORNL-1192, p. 63.

** Multi-Cyclotron Production Plant, ORNL-1192, p. 75.

APPENDIX A: CYCLOTRON DEVELOPMENTIncreased Energy

The maximum energy that could be obtained through January 1952 on a target 30.5" from the geometric center of the cyclotron was ~ 19 Mev. As calculated from H_p the energy at this radius should be ~ 22 Mev. When the target was withdrawn to a larger radius to obtain higher energy the proton beam struck the periphery of the south dee. It thus appeared that the effective center of beam rotation was displaced several inches to the south of the center line. Careful measurement of the positioning of grazing incidence targets indicated that the apparent center was about 3" south and $1\frac{1}{2}$ " below the geometric center.

It has been established that the position of the center of rotation depends upon the location of the ion source. In the 86-inch cyclotron the ion source is mounted at the end of a 15-foot stem which is inserted through the top of the tank and downward between the dee stems. An accelerating-slit electrode is attached to the dee facing the ion source. Both the ion source and this accelerating electrode were moved upward 1.5". To insure proper positioning and grounding of the lower end of the stem, a bracket was attached to the west side of the liner, positioning the ion source one inch below the geometric center. This arrangement insures that the placement of the ion source will be the same from run to run and also eliminates sparking from the stem to ground due to rf pickup. In addition, the dees were rotated from the support plate, displacing them southward 0.5", as measured at the magnetic center.

Beam measurements now indicate that the effective center of rotation is approximately $1\frac{1}{2}$ " above the geometric center. Since these changes, the pattern of the beam on the target is only $3\frac{1}{4}$ " by $\frac{3}{16}$ "; it was formerly 4" by $\frac{1}{4}$ ".

Larger Targets

A larger vacuum lock, 24" by 24", was attached to the under side of the vacuum tank to permit the installation of larger targets. Target size is now limited by the width of the magnet gap and the dimensions of the opening between the dees; a grazing incidence target of approximately 100 square inches can now be accommodated. The target dolly and water lines have been modified to provide increased water cooling to the larger targets.

Slotted-Plate Target. A new design of grazing-incidence type target successfully dissipates over 40 kw of beam power, on aluminum. These targets are made from plates of aluminum in which small slots are milled and then partially filled by welding to provide many small water passages close to the surface, Figure 4. Headers are welded to the ends and a shallow depression in the face is filled with bismuth which is then machined to a uniform thickness. The final operation is to bend the target to the proper curvature and fit it to the target support stem. A 6" by 14" target of this construction has been operated steadily with 1.2 ma at ~ 23 Mev for an extended period without evidence of surface damage.

Rotating Target. The 6" by 14" grazing-incidence target is the largest that can be conveniently placed in the cyclotron at present. Further increases in target area can be obtained by use of a rotating target. The advantages of combined rotation and grazing incidence are evident in a 15" rotating target now being tested. This target is similar to the slotted-plate target in construction, Figure 5. The beam will strike a $3\frac{1}{2}$ " by 4" area of the disc and rotation will increase the area of bombardment to ~ 140 square inches. At a loading of 0.5 kw/in^2 a total heat load of 70 kw can be absorbed, corresponding to nearly 3 ma of current.

SLOTTED PLATE

WELDED AND MACHINED

COMPLETED

FIGURE 4. SLOTTED-PLATE TARGET,

SLOTTED PLATE

"HELIARC" WELDED

ASSEMBLED

FIGURE 5 15" ROTATING TARGET

Appendix A (cont.)

Target Handling Facilities

At the higher energy of 23 Mev now obtained, larger amounts of gamma activity are produced in bismuth targets, in addition to the desired alpha activity. The principal gamma activity is due to the 5.7 hour decay (1.3 Mev gamma rays) of Po 207 produced from the (p,3n) reaction on bismuth. At 19 Mev the production of Po 207 was negligible; however, at 23 Mev a target contains about 50 curies/ma at equilibrium. At a distance of five feet from such a target the radiation level is 15 R/hr. To provide adequate protection, a 42" concrete radiation shield, with remote handling equipment and viewing window, has been constructed in the cyclotron pit directly opposite the cyclotron tank. From behind this shield the operator is able to remove a radioactive target, transfer it to a lead-shielded container, and seal this storage container. With this arrangement it is possible to keep the gamma exposure of operating personnel well below tolerance and also to limit the airborne dispersal of alpha active material. The airborne contamination level in the pit is presently 10-15 dis/min-m³, compared to 100-120 dis/min-m³ just prior to the January shutdown. At the entrance to the pit and outside the enclosed areas the level is negligible.

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APPENDIX B: BISMUTH BOMBARDMENT SUMMARY

<u>Target Number</u>	<u>Date</u>	<u>Average Beam (μa)</u>	<u>Bombardment Time (μah)</u>	<u>Assayed Yield (mc)</u>	<u>Observed Yield (mc/mah)</u>	<u>Cumulative Activity (mc)</u>
	10/15/51		(Corrected total from ORNL-1192)			2088
78	10/18/51	510	15,990	75.6	4.73	2164
79	10/22/51	459	32,798	145.8	4.44	2309
80	10/25/51	665	29,602	131.6	4.44	2441
81	10/29/51	632	36,130	141.1	3.90	2582
82	11/1/51	595	21,180	78.3	3.70	2660
83	11/5/51	551	28,540	149.5	5.24	2810
84	11/8/51	303	4,840	28.2	5.83	2838
86	11/12/51	544	29,170	170.2	5.83	3008
85	11/15/51	473	5,150	27.8	5.40	3036
88	11/18/51	431	10,290	32.2	3.13	3068
89	11/23/51	441	23,100	48.0	2.08	3116
91	11/25/51	506	12,720	64.0	5.03	3180
87	11/28/51	490	47,130	199.5	4.23	3380
92	12/4/51	691	60,890	231.6	3.79	3611
93	12/5/51	540	8,100	50.1	6.18	3661
94	12/12/51	620	41,497	199.5	4.81	3864
95	12/14/51	501	12,430	61.5	4.95	3922
96	12/16/51	615	26,820	91.9	3.43	4014
97	12/19/51	513	18,250	71.6	3.92	4086
99	12/26/51	408	12,610	51.3	4.07	4137
98	1/2/52	426	55,350	251.1	4.54	4388
101	1/5/52	467	21,460	108.5	5.06	4497
103	1/9/52	887	65,879	282.0	4.28	4779
100	1/11/52	654	15,624	93.0	5.95	4872
104	1/13/52	772	17,914	84.5	4.72	4956

Appendix B (cont.)

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<u>Target Number</u>	<u>Date</u>	<u>Average Beam (μa)</u>	<u>Bombardment Time (μah)</u>	<u>Assayed Yield (mc)</u>	<u>Observed Yield (mc/mah)</u>	<u>Cumulative Activity (mc)</u>
106	4/11/52	168	160	1.4	8.62	4958
107	4/24/52	156	148	2.0	13.72	4960
108	4/25/52	177	235	1.4	6.1	4961
109	5/3/52	269	1,319	12.0	9.1	4973
110	5/4/52	286	1,459	12.8	8.8	4986
111	5/10/52	226	2,060	17.3	8.4	5003
112	5/12/52	214	4,238	26.2	6.2	5030
113	5/18/52	244	3,045	35.7	11.7	5065
115	6/3/52	574	37,742	333	8.54	5388
114	6/9/52	661	51,075	446	8.73	5834
116	6/14/52	666	27,829	256	9.19	6090
117	6/18/52	614	15,714	101	6.43	6191
119	6/23/52	549	19,972	184	9.21	6375
120	7/2/52	604	41,280	388	9.40	6763
118	7/8/52	747	52,192	405	7.75	7168
121	7/15/52	641	55,600	623	11.20	7791
123	7/27/52	771	147,459	1452	9.84	9243

APPENDIX C: DISPOSITION OF CYCLOTRON PRODUCED Po 208

Total Produced	9.243 curies
Total On Hand (adjusted to September 1, 1952)	6.836
Present Location	
Los Alamos Scientific Laboratory	0.827
Mound Laboratory	
Half-Life Study	0.903
Other Experiments	0.286
Stockpile	4.820

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