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The Thick Target Yield and Excitation Function for the Reaction $Rh^{103}(pn)Pd^{103}$

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The low yield of carrier-free Pd^{103}, produced by the bombardment of a rhodium target with the internal beam of the ORNL 86-Inch Cyclotron, led to an investigation of the excitation function and thick target yield produced by bombardment with the external beam. The thick target yield for 16.7-Mev protons is 219 mc/ma-hr, a value which is high enough to encourage the further exploration of target development. The excitation function of the reaction is given.

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I. INTRODUCTION

The isotope Pd^{103} has been used with success as a source of interstitial irradiation in the treatment of various advanced tumors.[1] In the initial studies the Pd^{103} employed was obtained by neutron bombardment of palladium metal. One of the difficulties with the isotope so produced is that it contains large amounts of inert or carrier-palladium; therefore the specific activity of the end product is low. In an effort to obtain a product of higher specific activity, the ORNL 86-Inch Cyclotron was used to irradiate rhodium metal. By the $Rh^{103}(p,n)Pd$ reaction, Pd^{103} is produced carrier-free and the precursor isotope is 100% abundant. In addition, neutron activation of impurities such as iridium is avoided.

II. EXPERIMENTAL PROCEDURES

For the initial experiments, rhodium foils 3-1/2 x 1-5/8 x 0.029 in. were attached to the standard 6 x 5-5/8-in. copper target plate. One foil was soft soldered with a 50-50 lead-tin solder and the other was furnace brazed with silver solder. This foil size was chosen as a compromise to minimize the rhodium cost per bombardment while keeping the area large enough to intercept a large fraction of the incident beam. The beam has a width of some 2-3/4 in. but

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[1] P. V. Harper, K. A. Lathrop, L. Baldwin, Y. Oda, and L. Kryshtal, Ann. Surg. 148, 606 (1958).

there is a 1-1/2-in. core that contains 90% of the total current. The beam strikes the target at a grazing incidence and thus spreads over a length of 4 inches. The maximum angle of incidence of the protons is 7.6° so that the minimum effective thickness of the 2.9-mil rhodium foils is 735 mg-cm^{-2}, which is sufficient to stop an 18.7-Mev proton. The average energy of the beam incident on the target was 16.7 Mev, as determined by measuring simultaneously both the current and the power to the target.

The soft-soldered target failed at 900 μa after 10 min of bombardment. A hole the size of a half-dollar was melted in the rhodium foil. This target was not processed.

The silver-soldered target gave indications of melting at 1100 microamperes (sparking to target and pressure rise) but by adjusting the cyclotron operating condition it was possible to operate at 1600 microamperes. The total integrated charge was 4444 μa-hr for an average current of 1500 μa in a bombardment of 2.95 hours. A small hole 1/4 x 3/8 in. was melted in the rhodium foil near one corner.

For the external beam measurement, ten numbered foils of rhodium 0.033-in. thick were stacked together and bombarded with the 22.4-Mev external proton beam of the cyclotron. The foils were held in a target cup in the rear of a Faraday Cup assembly 7-1/2-in. long with a 3-1/2-in. diameter. The integrated current was measured with an integrator which produces an output pulse each time a capacitor is charged to the trigger voltage. The integrator was

calibrated with an internal current source. The energy of the incident beam was measured by range absorption in aluminum. The range-energy relation for rhodium was constructed by interpolation from the tables of Aron, Hoffman, and Williams.[2] A total of 0.329 μa-hr of protons was collected.

The foil from the internal bombardment was removed from the copper target plate by dripping concentrated nitric acid along the edges of the foil. It was then dissolved in $KHSO_4$ at a temperature of 500°C. Aliquots of both the $KHSO_4$ melt dissolved in water and of the HNO_3 solution were counted under conditions of negligible self-absorption (0.2 mg-cm^{-2} rhodium) in a defined geometry. A 2-mm NaI(Tl) crystal with a 5-mil Be window was used with a 256-channel analyzer to observe the 20.2-kev K-X-ray peak of Rh^{103}. A fluorescence yield of 0.80 was assumed. The accompanying 40-kev X-ray is converted almost completely in the L-shell and does not interfere.

For the external beam target foils, the rhodium outside the window opening of the target cup was trimmed off and the foils dissolved individually in ~20 gm of hot potassium bisulphate. (Radioautographs showed that there was no activity on the foils outside of the window opening.) The melt was dissolved in water and transferred to volumetric flasks after filtering. Any residue was digested further with $KHSO_4$ and added to the rest of the sample. For the total yield, aliquots (1/10) of each sample were combined and made up to volume in a separate flask.

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[2] W. A. Aron, B. G. Hoffman, and F. C. Williams, UCRL-121 (unpublished).

Aliquots (1/10 to 1/25) were transferred to a separatory funnel and diluted to ~30 ml with water. Palladium carrier in the amount of 200 μg was added in 0.5 ml con H_2SO_4. Then, 3 ml of saturated dimethyl glyoxime in 50% ethanol was added and, after mixing, KOH was added dropwise until the palladium glyoxime precipitate appeared. This was extracted in two 15-ml portions of chloroform. Repetition of the precipitation and extraction steps yielded 1% additional activity. After evaporation of the chloroform the palladium glyoxime was digested to dryness with a few drops of con HNO_3 and H_2SO_4 and then dissolved in 4.00 ml of 5N HNO_3. Three ml of this was transferred to a 10-ml Lusteroid tube for counting in a NaI well crystal.

The well crystal was calibrated with Pd^{103} sources of known activity as determined by calibration with the thin NaI(Tl) crystal. Duplicate samples were prepared by pipetting with the same pipette onto the paper disc for the standardization count, and into 3.00 ml of 5N HNO_3 in a 10-ml Lusteroid tube for the well counter. With two counts on each of two samples for both counters the efficiency of the well counter was determined to be 0.300 ± 0.007.

III. RESULTS

The yield for the internal run corrected to date of bombardment was 430 mc or 96.8 mc/ma-hr. The yield for 22-Mev protons calculated by Martin et al.[3] is 520 mc/ma-hr.

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[3] J. A. Martin, R. S. Livingston, R. L. Murray, and M. Rankin, Nucleonics 13, No. 3, 28 (1955).

Figure 1 shows the cross section plotted as a function of energy as measured in the external beam experiment. The relative precision can be judged from the scatter in the duplicate points. The absolute values are good to ±15% considering errors in beam monitoring, chemical yields, and counting normalization. The energy is the proton energy at the center of the foil.

The total yield, determined from four aliquots of the combined sample, was 77.5 μc. This gives 236 mc/ma-hr for 22.4-Mev protons in contrast to a calculated value of 520 mc/ma-hr. From the excitation function the yield is 219 mc/ma-hr for 16.7-Mev protons (the average energy for the internal bombardment). This value is to be compared with the 96.8 mc/ma-hr obtained from the internal bombardment.

The low yield obtained by bombardment with the internal beam may be accounted for in two ways: 1) Active material may have evaporated when part of the target melted, 2) The change in cyclotron operating conditions noted may have moved the beam core off the rhodium foil for most of the bombardment. It is difficult to explain this discrepancy between the full energy yield and the calculated value; better agreement is usually obtained.

The limit of the current that can be safely put on a target is determined by the power dissipation of the target. Thus, 200 μa at 20 Mev, or 400 μa at 10 Mev could both be handled safely by a given target since the depth distribution of the input power is not altered drastically by the change in energy. Figure 2 shows the relative yield as a function of the energy for conditions of constant input power.

It can be seen that 11.5 Mev is the optimum choice of bombarding energy for this reaction, being a 22% improvement over the yield at 16.7 Mev. Unfortunately, it is unlikely that the modifications of the probe system for the 86-inch cyclotron necessary to permit bombardment at energies below 16.7 Mev will be made; however the value of 219 mc/ma-hr for the production of Pd^{103} by 16.7-Mev protons is large enough to warrant further work on improved target construction.

IV. ACKNOWLEDGEMENTS

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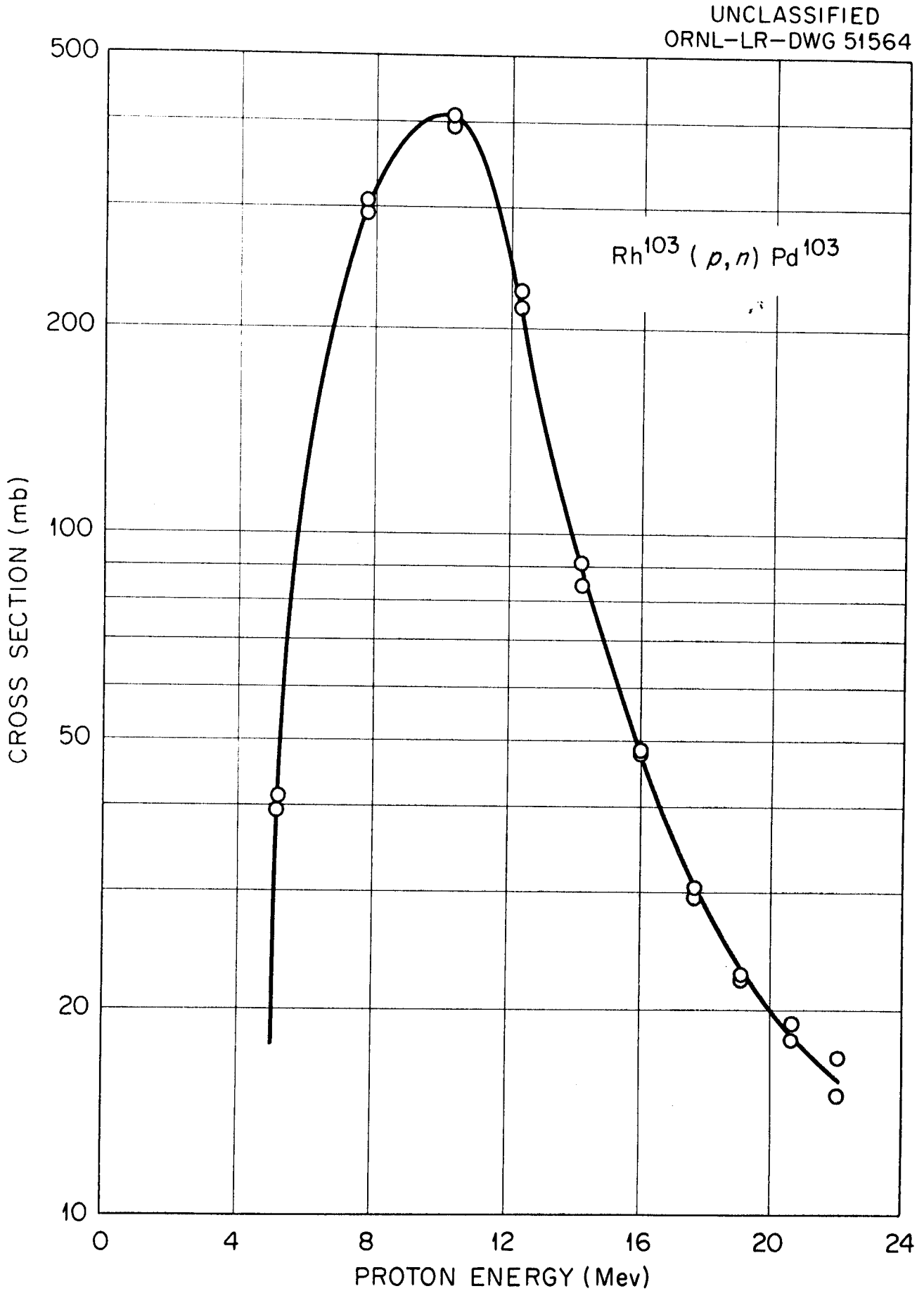

Fig. 1. Excitation function for the reaction $Rh^{103}(p,n)Pd^{103}$.

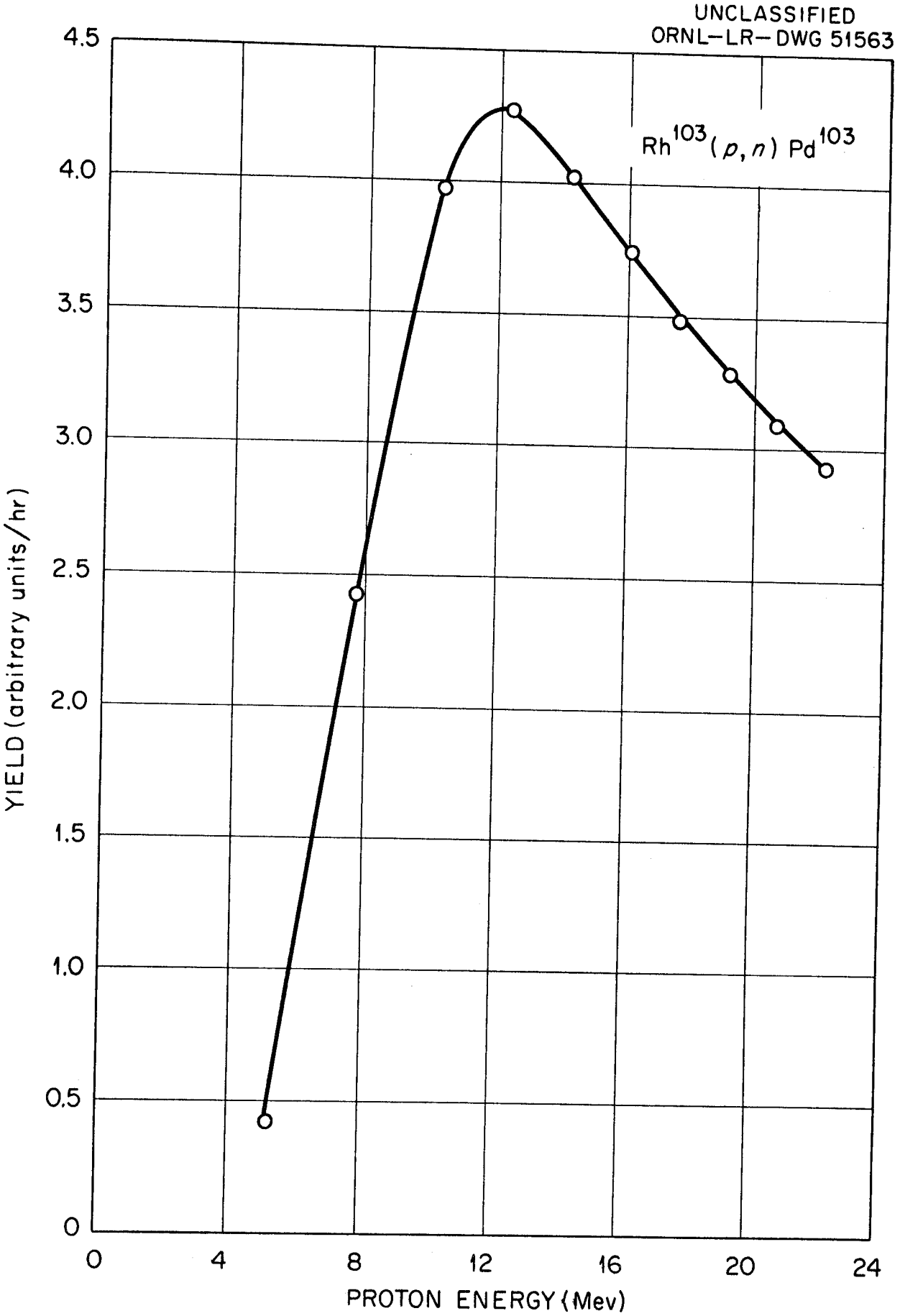

Fig. 2. Yield of Pd^{103} in arbitrary units per hour vs beam energy for constant power input to the target.