Nuclear Excitation Functions and Thick Target Yields: (Cr+d)†

PETER KAFALAS* AND JOHN W. IRVINE, JR.
Department of Chemistry and Laboratory for Nuclear Science, Massachusetts Institute of Technology, Cambridge, Massachusetts
(Received July 23, 1956)

Nuclear excitation functions and thick-target yields have been determined for several reactions which occur when chromium of natural isotopic composition is bombarded with deuterons of ~15 Mev. The reactions are $Cr^{52}(d,2n)Mn^{52}$, [$Cr^{53}(d,n)Mn^{54}$, $Cr^{54}(d,2n)Mn^{54}$], [$Cr^{50}(d,p)Cr^{51}$, $Cr^{50}(d,n)Mn^{51}\xrightarrow{\beta}Cr^{51}$, $Cr^{52}(d,t)Cr^{51}$], and $Cr^{50}(d,\alpha)V^{48}$. The excitation function for $Cr^{53}(d,n)$ was approximated by correcting for $Cr^{54}(d,2n)$ but the complex reactions yielding Cr^{51} could not be completely analyzed. Stacked chromium foils were bombarded, Mn, Cr, and V isolated chemically and the activities of relatively long-lived products measured with a calibrated scintillation counter. Absolute cross sections were calculated for several of the reactions and thick target yields as a function of deuteron energy were calculated for the production of Mn^{52}, Mn^{54}, Cr^{51}, and V^{48}. The energy loss of deuterons in chromium has been measured over the range of 3.8 to 13.6 Mev and some beam straggling data are presented.

INTRODUCTION

NUCLEAR excitation functions provide the best means of obtaining yield data for the cyclotron production of radionuclides. Deuteron bombardment of chromium has long been used for the production of carrier-free manganese and vanadium and chromium of moderate specific activity, but few quantitative data are available for the reactions of (Cr+d).[1]

This paper reports direct determination of the excitation function for the reactions $Cr^{52}(d,2n)Mn^{52}$ and $Cr^{50}(d,\alpha)V^{48}$ and an indirect determination of the excitation function for $Cr^{53}(d,n)Mn^{54}$. The formation of Cr^{51} occurs by three reactions, $Cr^{50}(d,p)$, $Cr^{50}(d,n)Mn^{51}\xrightarrow{\beta}Cr^{51}$, and $Cr^{52}(d,t)$, which cannot be resolved readily. However, by neglecting the contribution of the (d,t) above the threshold of 6.3 Mev, the cross section for the sum of (d,p) and (d,n) is obtained. Thick-target yields as a function of energy were calculated for the production of Mn^{52}, Mn^{54}, Cr^{51}, and V^{48} since these are derived directly from experimental data and do not require a knowledge of the contribution from each reaction which produces the nuclide of interest.

EXPERIMENTAL PROCEDURE

Foil Preparation

Chromium foils were prepared by the electrodeposition of chromium metal to a thickness of ~12 mg/cm^2 on sheet copper. After 20 hours annealing and outgassing at 225°C and <1μ Hg pressure, the mechanical properties of the foils were adequate for this work. It is essential that the foils be free of any visible oxide coating after this treatment since the oxide does not dissolve readily and interferes with the chemical processing. The copper was removed by long soaking (2 days at room temperature) in 1.5M nitric acid leaving foils ~1.5 cm in diameter which were mounted on aluminum rings to facilitate handling. This procedure was developed after many attempts to get foils that were strong enough to use.

Range-Energy Measurements

A low current of deuterons was obtained by scattering the primary beam from a tantalum target. Measurement of the energy of the individual deuterons was made by catching them in a NaI(Tl) crystal scintillator and making an analysis of the pulse heights obtained. The energy of the primary beam was calibrated against the protons from the reaction, $C^{12}(d,p)C^{13}$.[2] By the insertion of chromium foils in the scattered beam and measuring the energy attenuation of the deuterons the range-

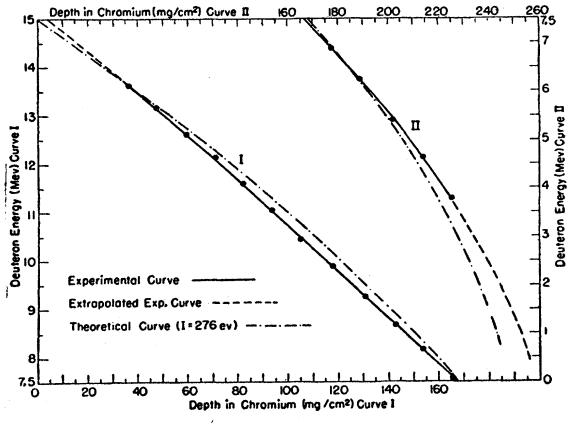

FIG. 1. Energy of deuterons at a given depth in chromium.

† This paper is based on a thesis submitted by Peter Kafalas to the Department of Chemistry in partial fulfillment of the requirements for the Ph.D. degree (1954). The work was supported in part by the U. S. Atomic Energy Commission.

* Present address: Chemical Engineering Division, Argonne National Laboratory, P. O. Box 299, Lemont, Illinois.

[1] Thick target yields of Mn^{52} have been reported by E. T. Clarke and J. W. Irvine, Jr. at 14 Mev, Phys. Rev. **70**, 893 (1946); W. M. Garrison and J. G. Hamilton at 19 Mev, Chem. Rev. **49**, 237 (1951), and an excitation curve for $Cr^{52}(d,2n)$ to 20 Mev was reported while the present work was in progress by Burgus, Cowan, Hadley, Hess, Shull, Stevenson, and York, Phys. Rev. **95**, 750 (1954).

[2] See Boyer, Gove, Harvey, Deutsch, and Livingston, Rev. Sci. Instr. **22**, 310 (1951) and F. A. Aschenbrenner, Phys. Rev. **98**, 657 (1955), for a description of the basic bombardment arrangements, calibration, and instrumentation used at the M.I.T. cyclotron.

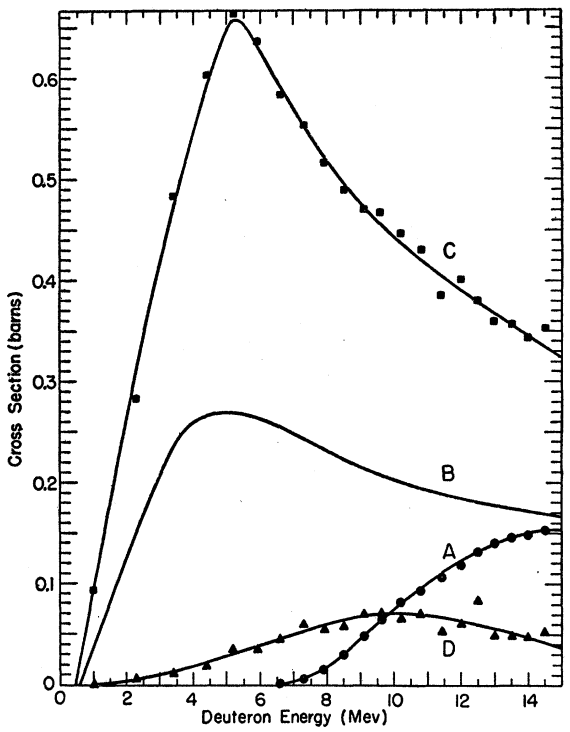

FIG. 2. Excitations functions for deuterons on chromium. A—$Cr^{52}(d,2n)Mn^{52}$; B—$Cr^{53}(d,n)Mn^{54}$; C—$\Sigma(\sigma)$ for (d,p) and $(d,n)\xrightarrow{\beta}$ for Cr^{51} formation; neglecting the (d,t) contribution above 6.3 Mev; D—$Cr^{50}(d,\alpha)V^{48}$.

energy curve was obtained. This was recalculated to give an energy *vs* depth of penetration curve for deuterons in chromium over the range 3.8 to 13.6 Mev (Fig. 1). Using a value of $I_{Cr}=276$ ev, the theoretical energy *vs* depth curve for chromium was calculated by the equation of Bethe and Ashkin.[3] Several other values of I were tested but the value of 276 ev calculated from $I=11.5Z$ determined for Al gave the best fit. For the energy values in the cross-section calculations the experimental curve was used.

Bombardment and Chemical Separations

A stack of 26 chromium foils was bombarded for 2.72 μa-hr at 14.70 Mev. Each foil was dissolved in hydrochloric acid, carrier manganese and vanadium added, and the chloride removed by boiling with nitric acid. The addition of solid potassium bromate oxidized chromium and vanadium to their highest oxidation states and precipitated manganese as MnO_2. After separating the MnO_2 the solution was neutralized to pH 1.3—1.4, hydrogen peroxide added, and chromium extracted into ethyl acetate as the peroxide, CrO_5. The aqueous phase was then made $1M$ in H_2SO_4 and the vanadium extracted into chloroform as the cupferrate. The chemical yield was determined for each separation. Table I gives a summary of the yields and decontamination factors for the above procedure.

Activity Measurements

All activities were measured on solution aliquots with a well-type NaI(Tl) scintillation counter. The counter efficiency was determined by measuring standard solutions of Mn^{52} (50.1%), Mn^{54} (16.3%), Cr^{51} (4.38%), and V^{48} (46.1%).

The Mn^{52} and V^{48} were standardized by 4π proportional counting and the disintegration rate calculated using the β^+/electron capture ratios of Good, Peaslee, and Deutsch.[4] Measurement of standard Mn^{54} in the same counter gave a correction factor of 1% for x-rays and Auger electrons from the electron capture process. Mn^{54} and Cr^{51} were standardized by K-γ coincidence measurements.[5]

RESULTS

Cross Sections

The excitation functions which were determined are shown in Fig. 2. For $Cr^{52}(d,2n)$ no effort was made to measure the isomer Mn^{52m} (21.3 min). Since this decays less than 1% to Mn^{52} it has no measurable effect on these results. The threshold of 6.6 Mev is substantially lower than the calculated value of 8.06 Mev which is ascribed to straggling of the beam.

Burgus *et al.*[1] have recently determined the same excitation function to 20 Mev by bombarding stainless steel foils. Although they made a correction for straggling their threshold is also low, by about 0.8 Mev. Their cross-section values are approximately 20 millbarns lower than ours in the range 8–12 Mev and their curve crosses ours at 13.5 Mev.

It should be noted that the work of these authors makes no mention of the contribution of the reaction $Fe^{54}(d,\alpha)Mn^{52}$ to the measured Mn^{52} activity. Since stainless steel foils were used in their Mn^{52} bombardment neglect of the (d,α) reaction is probably not justified.

Mn^{54} is produced by two reactions, $Cr^{53}(d,n)Mn^{54}$ and $Cr^{54}(d,2n)Mn^{54}$. Above the 4.6-Mev threshold for $Cr^{54}(d,2n)$ the observed activity includes contributions from both reactions. An estimate of the $Cr^{54}(d,2n)$ contribution was made by assuming this reaction has

TABLE I. Separation efficiency.

Element separated	Chemical yield (%)	Decontamination factor Mn	Cr	V
Mn	80	...	$>10^3$	10^2
Cr	75	$>10^4$	...	$>10^3$
V	60	$>10^4$	10^3	...

[3] H. A. Bethe and J. Ashkin, *Experimental Nuclear Physics*, edited by E. Segrè (John Wiley and Sons, Inc., New York, 1953), Vol. 1, p. 167.

[4] Good, Peaslee, and Deutsch, Phys. Rev. **69**, 313 (1946).

[5] These nuclides were standardized for us by W. S. Lyon, Jr. of the Oak Ridge National Laboratory, Oak Ridge, Tennessee.

twice the cross section of $Cr^{52}(d,2n)$ at equal energies in excess of threshold. The factor of two involves the assumption of equal cross sections for the formation of Mn^{52} and Mn^{52m}. The difference between the calculated curve for $Cr^{54}(d,2n)$ and experimental curve for total Mn^{54} production is plotted as Curve *B*, Fig. 2.

Cr^{51} is formed by the three reactions: $Cr^{50}(d,p)Cr^{51}$, $Cr^{50}(d,n)Mn^{51}\xrightarrow{\beta^+}Cr^{51}$, and $Cr^{52}(d,t)Cr^{51}$. Curve *C*, Fig. 2, is the sum of the cross sections for the (d,p) and (d,n) reactions, as the Mn^{51} (44 min) was allowed to decay before chemical separations were started. Below 6.3 Mev, the threshold for $Cr^{52}(d,t)$, the curve should be correct. Above this value there will be a small systematic error due to (d,t). On the basis of some unpublished data of E. T. Clarke on the cross section of $Co^{59}(d,t)Co^{58}$, the (d,t) contribution is estimated to be less than 5% of the (d,p).

V^{48} is formed by the reaction $Cr^{50}(d,\alpha)V^{48}$. The excitation function for this reaction is shown as Curve *D*, Fig. 2. The analytical data on the chemical yield were poor and are responsible for the rather large scatter in the experimental points.

Yields

Thick-target yiedls *versus* energy for the production of these four nuclides are shown in Fig. 3. These are obtained by graphical integration of the area under the experimental curves for total nuclide production.

The yields at 15 Mev for Mn^{52} are about 1.5–2 times higher than those obtained routinely with the M.I.T. cyclotron by Clarke and Irvine[1] and Backofen.[6] Since the normal bombardments are made on a chromium plated probe in the vacuum chamber, loss due to volatilization of manganese is expected. The ratio of activity of Mn^{54}/Mn^{52} of $\sim$0.01 is in agreement with 0.011 reported by Backofen and Herber.[7]

Half-Lives

The decay was observed over a period of at least four half-lives of twenty-one samples each of Cr^{51} and V^{48} and twelve samples with a high Mn^{52}/Mn^{54} ratio. The twenty-one Mn^{54} samples were followed only for about four months. The observed values of Mn^{52} ($5.69\pm0.03d$), Mn^{54} ($290\pm6d$), Cr^{51} ($27.9\pm0.2d$) and V^{48} ($15.99\pm0.08d$) are in good agreement with the best values summarized by Way *et al.*[8]

TABLE II. Beam straggling.

Av energy (Mev)	$-\Delta E$ (%)	$+\Delta E$ (%)	Av energy (Mev)	$-\Delta E$ (%)	$+\Delta E$ (%)
13.1	4.5	4.5	7.3	11	17
12.6	5.0	6.3	6.6	12	18
12.0	5.0	6.6	5.9	14	21
11.5	5.2	7.0	5.2	22	25
10.9	5.5	7.4	4.2	30	35
10.3	6.8	7.9	3.3	40	50
9.7	7.3	9.4	2.1	80	100
9.1	8.0	10			
8.5	8.5	11			
7.8	10.5	13			

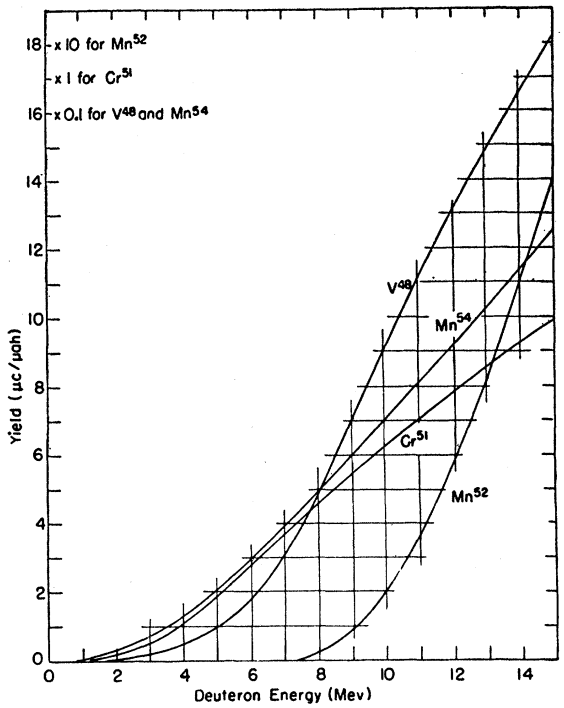

FIG. 3. Thick target yields of Mn^{52}, Mn^{54}, Cr^{51}, and V^{48} for deuteron bombardment of chromium of normal isotopic composition.

Straggling of the Deuteron Beam

In the course of getting the stopping power of chromium a set of data on straggling was obtained. After passing through the chromium foils the deuteron beam was caught in a NaI(Tl) crystal and the pulses fed into pulse analyzer. The average deuteron energy and limits (>95%) are shown in Table II for an initial deuteron energy of 13.6 Mev. A pronounced forward asymmetry was observed.

ACKNOWLEDGMENTS

The authors wish to thank Dr. Frank A. Aschenbrenner for his supervision of and assistance with the cyclotron bombardments and for numerous helpful discussions of the problems involved. They are also indebted to the cyclotron crew, especially E. F. White and F. J. Fay, for their help and cooperation.

[6] E. W. Backofen (private communication, 1956).
[7] E. W. Backofen and R. H. Herber, Phys. Rev. **97**, 743 (1955).
[8] Way, King, McGinnis, and van Lieshout, Atomic Energy Commission Report USAEC-TID-5300, June 1955.