High-Energy Proton Spallation of Titanium*

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The cross sections for the production of ^{48}V, ^{48}Sc, ^{47}Sc, ^{47}Ca, ^{46}Sc, ^{44m}Sc, ^{44}Ti, ^{43}K, ^{42}K, ^{28}Mg, ^{24}Na, ^{22}Na, and ^{7}Be from 15- to 584-MeV proton spallation of titanium are reported. The experimental production rates of the various isotopes and their ratios to ^{47}Sc are compared with theoretical spallation-yield calculations. Application of the measured excitation functions to the analysis of lunar materials and meteorites is discussed.

I. INTRODUCTION

An interpretation of the cosmogenic radionuclide concentrations in extraterrestrial materials in terms of their cosmic-ray-exposure history must necessarily depend on accurate spallation production cross sections. In the case of meteorites it is sufficient to establish the excitation functions for the spallation of the major target elements at energies above several hundred MeV or to estimate the relative production rates of a few cosmogenic radionuclide pairs.[1] This is valid, since only the spallation products produced by the high-energy galactic cosmic rays leave a discernible record in meteorites. Products of the low-energy solar component of the cosmic-ray spectrum which occur only in the first few millimeters of the body do not ordinarily survive the ablation caused by entry through the earth's atmosphere. The problem of the interaction of low-energy secondaries in a large body can be estimated from thick-target bombardment data[1] without recourse to low-energy excitation curves.

The situation for lunar material is somewhat different than that described for meteorites. The sample surface which is exposed to the intense low-energy solar component of the cosmic-ray spectrum is not removed by ablation effects prior to analysis. These solar cosmic rays can produce radionuclides on the lunar surface by low-energy nuclear reactions on minor constituents of the material in quantities which are comparable to or greater than the high-energy spallation products of major constituents. In addition, the departure in chemical composition of the lunar material from meteorites[2] can further affect the relative importance of various production modes on the lunar surface. A notable example of both these effects is the production of ^{44}Ti and ^{46}Sc. While spallation of iron is the major mode of production of these isotopes in meteorites, the relatively high concentration of titanium coupled with the intense low-energy solar component of the cosmic-ray spectrum on the moon renders titanium a major parent element in lunar material.

Because of these considerations it became evident that the excitation functions for the production of the various radionuclides measured in lunar material must be established in the important energy interval from the reaction threshold to a few hundred MeV. This work deals with the results of an investigation to establish the spallation production cross sections as a function of energy for several radionuclides from titanium targets. The results are also compared with calculated cross sections using the formulas established by Rudstam[3] in an attempt to determine if the deviation of experimental results from the theoretically predicted values noted in the cases of argon[4] and iron[5] is a function of the atomic number of the target nucleus.

II. EXPERIMENTAL PROCEDURES AND RESULTS

Titanium foils, 99.5% pure, with dimensions of 5.4 cm in diameter and 0.025 cm thick were irradiated with 15-, 30-, and 45-MeV protons at the Berkeley 88-in. cyclotron. Similar foils 0.0775 cm and 0.318 cm thick were irradiated with 118-MeV protons at the Harvard University cyclotron and with 319-, 433-, and 584-MeV protons at the Space Radiation Effects Laboratory (SREL) cyclotron. The beam currents at the Harvard and SREL cyclotrons were monitored by proportional counters and by measuring the amount of ^{24}Na activity induced in exposed aluminum foils according to the ^{27}Al$(p, 3pn)^{24}$Na reaction. The cross section for this reaction was taken to be 10.9, 11.3, 11.0, and 10.8 mb at 118, 319, 433, and 584 MeV, respectively.[6] The beam currents during the Berkeley irradiations were monitored by a calibrated Faraday cup with uncertainties of less than 5%. Integral proton doses of $1-8\times10^{13}$ protons were incident on the targets exposed at Harvard and SREL during irradiation times of 17–60 min. Proton

doses of $3-5\times10^{15}$ total protons were obtained at Berkeley during exposure periods of 5 or 10 minutes.

The radionuclides produced in the titanium foils were measured nondestructively with multidimensional anticoincidence-shielded NaI(Tl)[7,8] and Ge(Li) γ-ray spectrometers[9] after decay intervals varying from 30 h to 1 yr. Those radioisotopes measured in this work are listed in Table I along with the half-lives, γ-ray energies, and branching intensities[10] used for their identification and yield calculations. Also listed in Table I are the minimum proton kinetic energies necessary to produce each radioisotope and that particular corresponding reaction. When the Coulomb barrier is higher than the Q value, the threshold value is followed by a (V_C) to indicate that the value is based on the Coulomb barrier for the reaction listed. All thresholds are reported in the laboratory frame of reference.

The ^{22}Na activity was below the detection limit of the Ge(Li) spectrometers, and ^{22}Na and ^{44}Ti decay schemes are so similar that they are not completely resolved with NaI detector systems. However, the ^{22}Na activity was determined by the difference between the total activity attributed to both ^{22}Na and ^{44}Ti observed in the NaI(Tl) multidimensional spectrometer analyses and the known ^{44}Ti activity as measured independently by a Ge(Li) spectrometer. No activities were observed which were attributable to the spallation of any element heavier than titanium. In addition, scandium, a potential contaminant which could have a significant effect on low-energy yields, was found to be present in the titanium foil at a level of only 38 atom parts per 10^9 by instrumental neutron activation analysis. No other elements lighter than titanium were found by this technique. Thus the presence of any impurities in the titanium foils had a negligible effect on the reported results.

The error values quoted for the experimental data summarized in Table II are consistent with the uncertainties associated with counting statistics, detector calibrations, proton fluxes, and

TABLE I. Relevant properties of radionuclides measured.

Isotope	Half-life	γ-ray energies (MeV)	Branching intensity (%)	Production threshold [a] (MeV)	Reaction
^{48}V	16.0 day	1.312	97	4.875 (V_C)	^{47}Ti(p, γ)^{48}V
		0.983	100		
		0.945	10		
^{48}Sc	1.83 day	1.040	100	11.573	^{49}Ti(p,2p)^{48}Sc
^{47}Sc	3.43 day	0.160	73	8.244 (V_C)	^{50}Ti(p, α)^{47}Sc
^{46}Sc	83.9 day	1.120	100	8.289 (V_C)	^{49}Ti(p, α)^{46}Sc
		0.889	100		
^{44m}Sc	2.44 day	1.159	98.6	8.381 (V_C)	^{47}Ti(p, α)^{44m}Sc
		0.271	86		
^{47}Ca	4.535 day	1.297	74	22.116	^{49}Ti(p,3p)^{47}Ca
^{44}Ti	48 yr	1.159	100	14.433	^{46}Ti(p,t)^{44}Ti
		0.078	98		
		0.068	90		
^{43}K	22.4 h	0.619	81	12.660	^{50}Ti(p,2α)^{43}K
		0.594	13		
		0.395	18		
		0.373	85		
^{42}K	12.36 h	1.524	18	11.327	^{49}Ti(p, 2α)^{42}K
^{28}Mg	21.3 h	1.780	100	22.896 (V_C)	^{50}Ti(p, ^{23}Na)^{28}Mg
^{24}Na	15.0 h	1.369	100	22.880 (V_C)	^{50}Ti(p, ^{27}Mg)^{24}Na
^{22}Na	2.60 yr	1.786 x 0.511 (coinc.)	90	23.069 (V_C)	^{49}Ti(p, ^{28}Mg)^{22}Na
^{7}Be	53 day	0.477	10.3	18.234	^{47}Ti(p, ^{7}Be)^{41}K

[a] Lowest possible value in the laboratory frame of reference. V_C in parentheses indicates value based on Coulomb barrier which is higher than Q value of reaction.

TABLE II. Proton spallation of titanium cross sections in mb.

Energy (MeV)	Isotopes	σ (Expt.)	σ (Calc.)	$\sigma/\sigma(^{47}Sc)$ (Expt.)	$\sigma/\sigma(^{47}Sc)$ (Calc.)
15	^{48}V	408 ± 50		1230 ± 260	
15	^{48}Sc	<0.24		<0.66	
15	^{47}Sc	0.332 ± 0.081		1	
30	^{48}V	29.1 ± 3.9	2120	1.57 ± 0.09	15.1
30	^{48}Sc	0.420 ± 0.075	131	0.0226 ± 0.0030	0.936
30	^{47}Sc	18.5 ± 2.3	140	1	1
30	^{46}Sc	8.1 ± 1.4	110	0.435 ± 0.053	0.797
30	^{44m}Sc	12.3 ± 1.5	9.19[a]	0.6634 ± 0.0097	0.06543[a]
30	^{44}Ti	0.595 ± 0.075	0.497	0.03208 ± 0.00087	0.00354
45	^{48}V	24.7 ± 3.2	1360	1.069 ± 0.047	10.22
45	^{48}Sc	1.57 ± 0.20	84.3	0.0678 ± 0.0025	0.634
45	^{47}Sc	23.1 ± 2.8	133	1	1
45	^{46}Sc	71.1 ± 8.8	157	3.078 ± 0.038	1.179
45	^{44m}Sc	14.4 ± 1.8	28.1[a]	0.6231 ± 0.0063	0.2115[a]
45	^{44}Ti	3.68 ± 0.48	1.52	0.1593 ± 0.0066	0.01144
45	^{43}K	1.10 ± 0.25	0.900	0.0475 ± 0.0092	0.00677
118	^{48}V	5.10 ± 0.68	373	0.505 ± 0.028	5.84
118	^{48}Sc	1.50 ± 0.21	23.1	0.148 ± 0.011	0.362
118	^{47}Sc	10.1 ± 1.2	63.8	1	1
118	^{46}Sc	21.2 ± 3.6	132	2.10 ± 0.24	2.06
118	^{44m}Sc	8.8 ± 1.1	72[a]	0.870 ± 0.015	1.13[a]
118	^{43}K	1.23 ± 0.18	4.04	0.0900 ± 0.0073	0.0633
118	^{42}K	3.6 ± 0.6	10	0.36 ± 0.04	0.16
319	^{48}V	1.91 ± 0.44	70.7	0.1052 ± 0.0022	4.451
319	^{48}Sc	1.72 ± 0.40	4.38	0.0946 ± 0.0015	0.276
319	^{47}Sc	18.2 ± 4.2	15.9	1	1
319	^{46}Sc	22.2 ± 5.2	43.0	1.218 ± 0.032	2.703
319	^{44m}Sc	6.8 ± 1.9	41[a]	0.376 ± 0.062	2.55[a]
319	^{47}Ca	0.187 ± 0.055	0.349	0.0096 ± 0.0017	0.022
319	^{44}Ti	1.55 ± 0.39	2.20	0.0853 ± 0.0082	0.138
319	^{43}K	2.04 ± 0.48	2.98	0.1121 ± 0.0040	0.1875
319	^{42}K	6.4 ± 1.5	9.8	0.352 ± 0.011	0.614
319	^{28}Mg	0.023 ± 0.012	0.028	0.0012 ± 0.0006	0.0018
319	^{24}Na	0.353 ± 0.084	0.249	0.0194 ± 0.0011	0.0156
319	^{22}Na	0.196 ± 0.046	0.130	0.00981 ± 0.00036	0.00816
433	^{48}V	0.78 ± 0.15	56	0.0670 ± 0.0028	4.24
433	^{48}Sc	1.20 ± 0.23	3.48	0.1037 ± 0.0031	0.2628
433	^{47}Sc	11.6 ± 2.2	13.2	1	1
433	^{46}Sc	13.4 ± 2.6	37.6	1.156 ± 0.042	2.842
433	^{44m}Sc	3.81 ± 0.79	39.3[a]	0.328 ± 0.029	2.965[a]
433	^{44}Ti	0.89 ± 0.21	2.12	0.077 ± 0.012	0.160
433	^{43}K	1.45 ± 0.28	3.03	0.125 ± 0.005	0.229
433	^{42}K	4.15 ± 0.80	10.4	0.357 ± 0.015	0.786
433	^{24}Na	0.347 ± 0.068	0.647	0.0298 ± 0.0018	0.0489
433	^{22}Na	0.321 ± 0.062	0.373	0.0252 ± 0.0010	0.0282
584	^{48}V	1.02 ± 0.35	44.9	0.0587 ± 0.0059	4.08
584	^{48}Sc	1.62 ± 0.52	2.78	0.0931 ± 0.0014	0.253
584	^{47}Sc	17.4 ± 5.6	11	1	1
584	^{46}Sc	17.5 ± 5.6	32	1.004 ± 0.023	2.951
584	^{44m}Sc	4.6 ± 1.5	37[a]	0.262 ± 0.012	3.32[a]
584	^{44}Ti	0.62 ± 0.21	1.98	0.0356 ± 0.0032	0.180
584	^{43}K	2.11 ± 0.68	2.9	0.1211 ± 0.0022	0.2665
584	^{42}K	6.0 ± 1.9	10.5	0.3443 ± 0.0066	0.953
584	^{28}Mg	0.100 ± 0.034	0.104	0.00572 ± 0.00056	0.00940
584	^{24}Na	0.87 ± 0.28	1.3	0.04995 ± 0.00094	0.1181
584	^{22}Na	0.64 ± 0.21	0.809	0.03351 ± 0.00050	0.07344
584	^{7}Be	0.91 ± 0.32	0.0220	0.0525 ± 0.0077	0.00200

[a] Calculated for ^{44}Sc not for ^{44m}Sc.

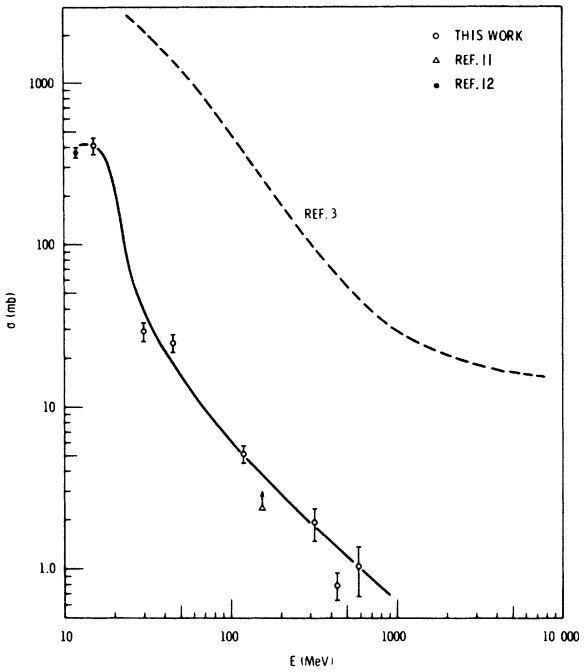

FIG. 1. Proton spallation cross sections of ^{48}V from titanium.

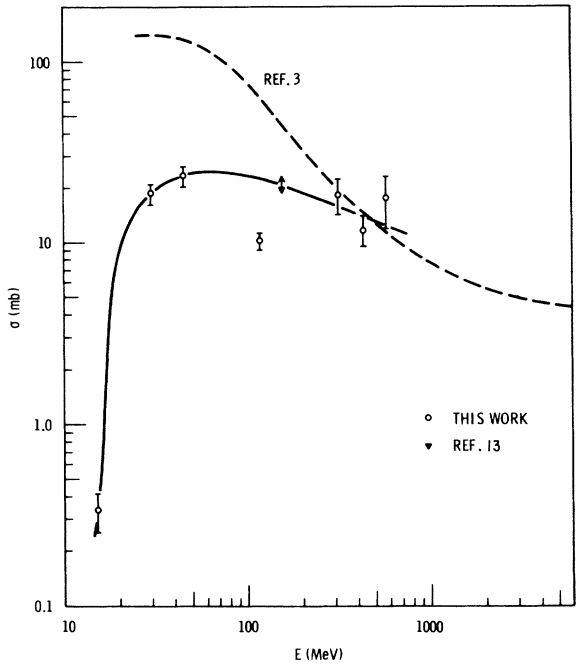

FIG. 3. Proton spallation cross sections of ^{47}Sc from titanium.

backgrounds. Since a rather large uncertainty is associated with the proton flux at the three highest energies, the ratio of the cross section for each observed isotope to that of ^{47}Sc at each energy is also given in Table II. Scandium-47 was chosen as the normalizing isotope, since it is produced over the entire energy range with a maximum in statistical accuracy. All radioisotopes were determined from each target by nondestructive γ-ray analyses. Therefore, errors normally attributable to differences in proton flux, chemical yield, or counting geometry are eliminated in the ratios of isotope yields, and only the uncertainties associated with the counting statistics remain.

III. DISCUSSION

The cross-section measurements of this work were used for a continuing evaluation of the CDMD cross-section equation of Rudstam.[3] This semiempirical, five-parameter equation corresponds to an exponential mass-yield distribution and a Gaussian charge distribution. The theoretical cross sections and ratios of cross sections calculated by this formula are presented with the experimental

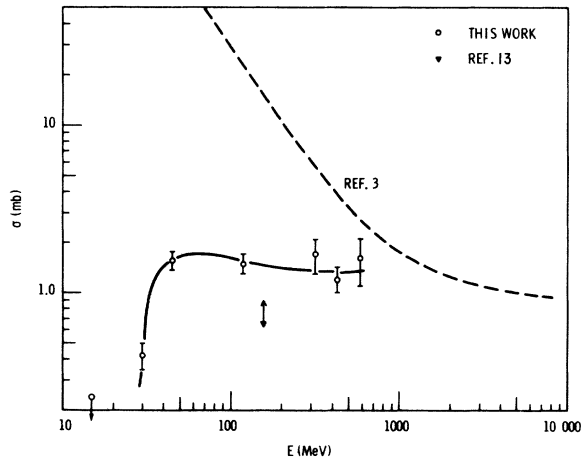

FIG. 2. Proton spallation cross sections of ^{48}Sc from titanium.

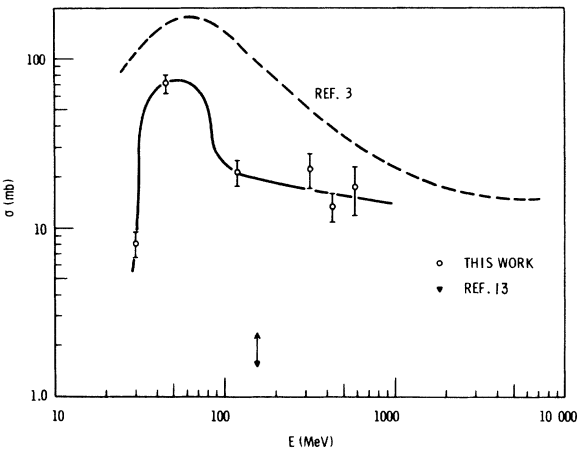

FIG. 4. Proton spallation cross sections of ^{46}Sc from titanium.

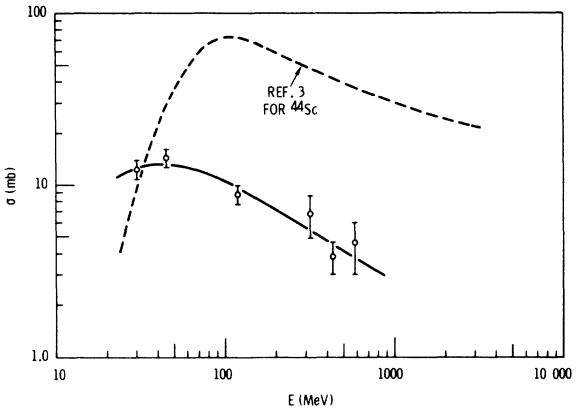

FIG. 5. Proton spallation cross sections of ^{44m}Sc from titanium.

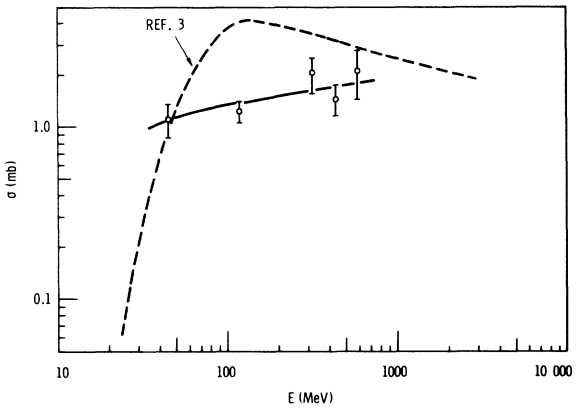

FIG. 7. Proton spallation cross sections of ^{43}K from titanium.

data in Table II. Complete excitation functions for most isotopes studied in this work have been generated by the formula, and these theoretical curves are plotted as dashed lines in Figs. 1-12 together with the experimental results of this and previously reported work.[3,11-16] The data of Cohen, Albouy, and Poffe,[13] and Valentin[11] plotted in the figures are lower limits for the yields of spallation products from natural titanium which were calculated from their reported cross sections on separated isotopes of titanium.

Several observations can be made from a comparison of the theoretical and experimental data. The best agreement between the theoretical curves and the experimental data occurs for the spallation products in the mass range 22–28. The divergence of the theoretical from the experimental excitation functions increases with product mass up to a factor of 60 at products of the same mass as the target. This disagreement at similar target and product masses is a well-recognized artifact of the Rudstam equation. The disparity in Fig. 5 is due to the fact that the experimental data are a measure of the ^{44m}Sc production, while the theoretical data apply to ^{44}Sc. The discrepancy for ^{7}Be shown in Fig. 12 stems from the semiempirical equation assumption that the target nucleus is spallated down to ^{7}Be, and discounts the more likely possibility that such a light fragment may be emitted from the spalled nucleus as an entity. In general, the theoretical excitation function maintains a shape similar to the experimental data up to product mass 46, which is 0.96 of the target mass, at which point large deviations become apparent for energies below about 300 MeV. This is in contrast to the data from the spallation of iron[5] where the deviation becomes large at about mass 48, which is only 0.85 times the target mass. This implies that the theoretical curve maintains the correct shape for a larger percentage of the products from lighter targets than from heavier targets. In almost all cases, the theoretical curve is higher than the experimental. A generally better fit would be obtained by multiplying the theoretical data for

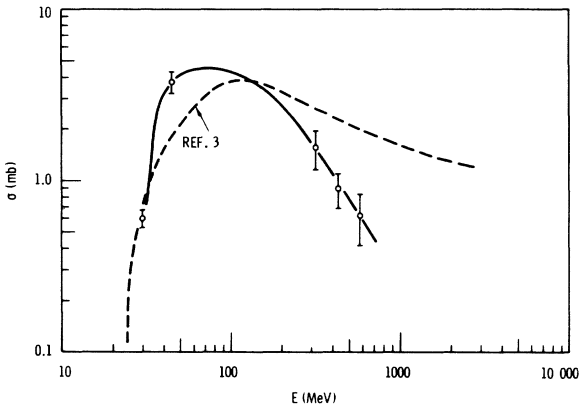

FIG. 6. Proton spallation cross sections of ^{44}Ti from titanium.

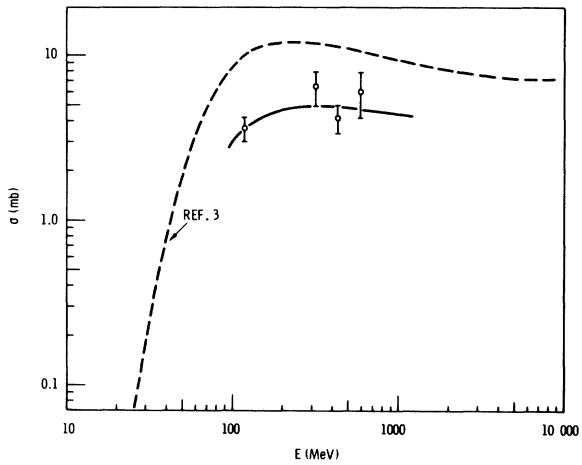

FIG. 8. Proton spallation cross sections of ^{42}K from titanium.

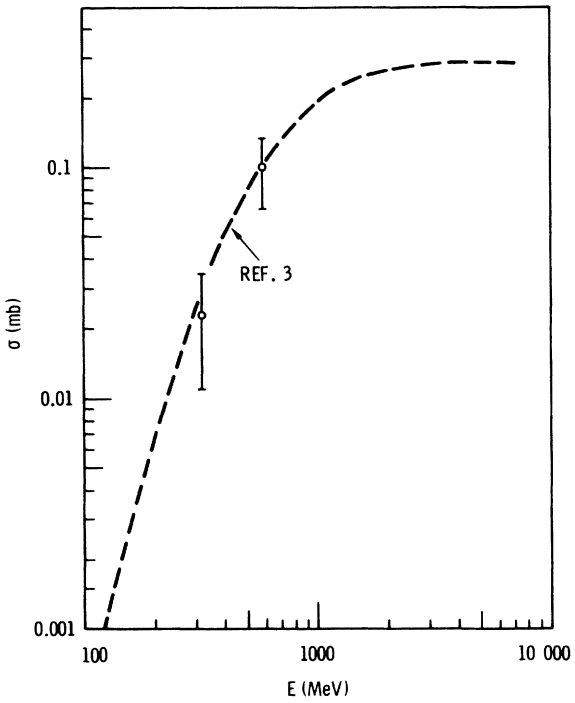

FIG. 9. Proton spallation cross sections of ^{28}Mg from titanium.

the spallation of titanium by 0.79. Comparison of this value to the "better fit" multiplier of 0.81 obtained for the spallation of argon[4] and the "better fit" multiplier of 0.66 obtained for iron[5] suggests

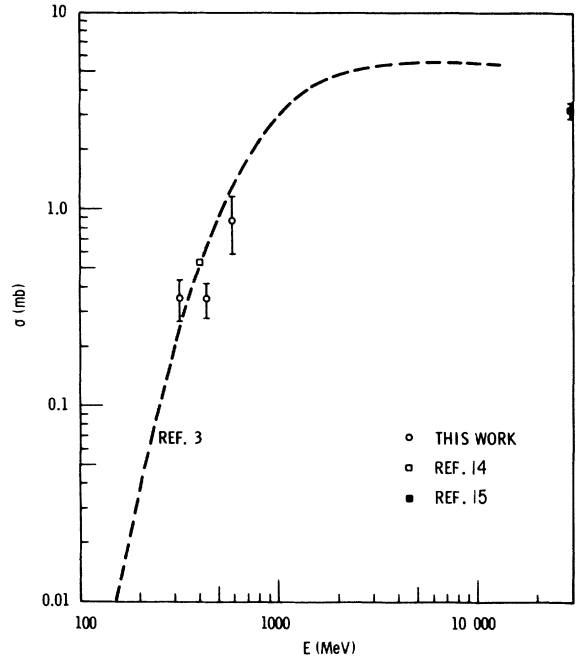

FIG. 10. Proton spallation cross sections of ^{24}Na from titanium.

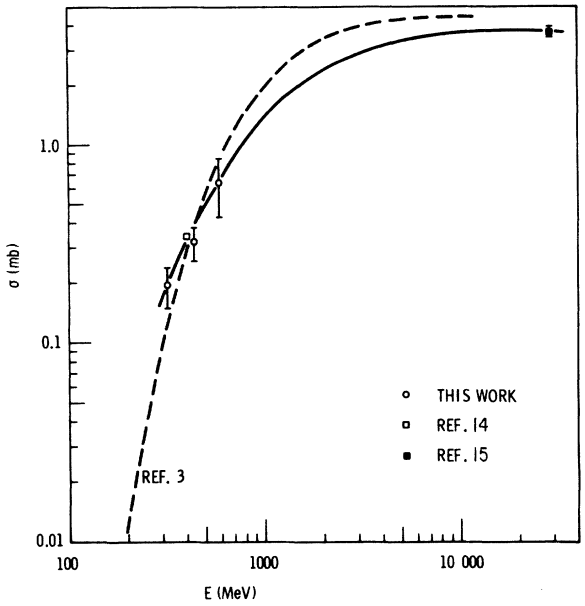

FIG. 11. Proton spallation cross sections of ^{22}Na from titanium.

that a reduction factor which is dependent on the target mass should be introduced into the semiempirical equation. Since the agreement between the theoretical and experimental excitation curves is better for light fragments than for heavy, a dependence on the mass of the fragment should also be incorporated. Finally, since the low-energy portion of the theoretical excitation function deviates

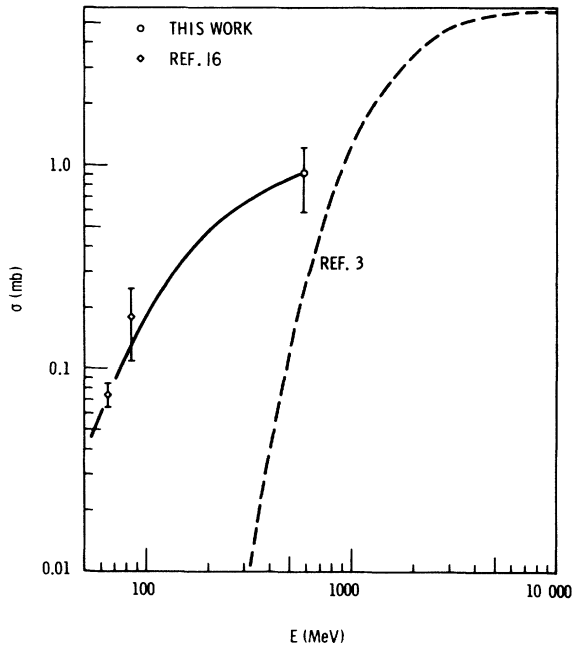

FIG. 12. Proton spallation cross sections of ^{7}Be from titanium.

widely for heavy fragments from heavy targets, a dependence on the energy of the incident protons must also be included. When these modifications to the semiempirical yield equation are completed[6] it will be able to more accurately predict the yields of all spallation fragments, including very short-lived and stable isotopes, from a wide variety of targets, over a large energy range.

In order to draw accurate conclusions regarding the cosmic-ray exposure history from measurements of the cosmogenic radionuclide content of meteorites and lunar surface samples, a thorough knowledge of the cross section of each measured radionuclide from all possible parents is necessary. The high titanium content of Apollo 11 and 12 lunar samples illustrates the need for the spallation excitation functions of titanium.[17]

Several of the radionuclides measured in lunar material can be produced from both iron and titanium with the relative contributions from each highly dependent on energy. For example, the production cross section of ^{48}V is at least 100 times higher from titanium than from iron at 15 MeV, and the production of ^{46}Sc ranges from 10 to 70 times higher from titanium than from iron at energies up to and beyond 150 MeV. The intensity of the low-energy particles from the solar flare of April 12, 1969 is an additional factor enhancing the production of the ^{48}Sc and ^{49}V in the lunar samples from the spallation of titanium even though the chemical composition favors their production from iron by a factor of 3 to 7. The quantities of the various radioisotopes present in a specimen with titanium content comparable to that of the lunar samples could yield important information regarding the incident cosmic particles if the samples are analyzed immediately upon return to earth. For example, ^{48}V, ^{48}Sc, ^{47}Sc, ^{46}Sc, ^{44m}Sc, and ^{44}Ti are produced copiously from titanium by protons of less than 50 MeV. The relative concentrations of these isotopes of scandium, which have half-lives ranging from 1.83 to 83.8 days, could be employed to determine the variations in the solar activity over time scales ranging from a few days to a few months. In addition, the measurement of ^{44}Ti would permit the temporal variation in solar activity to be defined over the last few solar cycles. A comparison of the concentration of ^{48}V, which is produced from titanium primarily by particles with energies of 10 to 30 MeV, with the concentrations of the scandium and titanium isotopes, which are produced primarily by particles of 30 to 50 MeV, would help to establish the shape of the energy spectrum of incident particles from 10 to 50 MeV. The radionuclides ^{28}Mg, ^{24}Na, ^{22}Na, and ^{7}Be which require about 150 MeV to be produced in appreciable quantities from titanium can be produced prolifically by lower-energy particles on lighter target elements. These effective energy barriers thus differentiate the radionuclides as being produced principally from the solar (low-energy) or the galactic (high-energy) components of the cosmic-ray spectrum. These effective thresholds also provide information on the production rate of a radionuclide with depth in a meteorite or a lunar specimen since the radionuclides produced by the solar (low-energy) protons are always found near the surface of the material while the radionuclides found deep within the specimen must be a product of the galactic (high-energy) component of the cosmic-ray spectrum. Such a radionuclide profile could be calculated from the known excitation functions according to a method employed by Arnold, Honda, and Lal.[18]

With excitation functions extended to low-energy regions a reasonably accurate cosmic-ray exposure history can be determined from an analysis of the radionuclides in a mixture of exposed elements such as a lunar surface sample, a meteorite, or a part from a spacecraft recovered from the moon. In fact, because of the difference in the half-lives of the different products, chronological fluctuations of the cosmic-ray spectrum may be evident. Ideally, pure elemental samples could be exposed to the cosmic flux for known periods of time to eliminate any uncertainty in the parentage of the observed isotopes and their production cross sections. The flux and energy spectrum of the incident protons would then be accurately obtainable from the various quantities of radionuclides present. Such measurements would be possible on the lunar surface if pure elemental targets were exposed at the initiation of extravehicular activity (EVA) and recovered prior to takeoff or during a later landing at the same site.

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High-Energy Proton Spallation of Iron*

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The cross sections for the production of ^{57}Co, ^{56}Co, ^{55}Co, ^{54}Mn, ^{52}Mn, ^{51}Cr, ^{48}Cr, ^{48}V, ^{48}Sc, ^{47}Sc, ^{46}Sc, ^{44m}Sc, ^{44}Ti, ^{43}K, ^{42}K, ^{24}Na, and ^{22}Na from 14.1 to 585-MeV proton spallation of iron are reported. The experimental production rates of the various radioisotopes and their ratios to ^{54}Mn are compared to theoretical spallation-yield calculations. These data and previously published data are combined to develop the excitation functions for these isotopes from proton spallation of natural iron. Applications of these excitation functions to beam monitoring and to the studies of meteorites and lunar surface material are discussed.

I. INTRODUCTION

The spallation cross sections of the constituent elements of any specific material are required to unravel the prior irradiation history of that material from measurements of its induced spallation products. For example, proton spallation cross sections are needed to interpret cosmogenic radionuclide concentrations in terms of the cosmic-ray exposure history of spacecraft materials, meteorites, material from the earth's moon, or future samples from the planets. Only the high-energy cross sections are necessary for the interpretation of the cosmogenic radionuclide concentrations in meteorites, since any spallation products formed at energies below about 100 MeV are near the surface and would be ablated away on entry through the earth's atmosphere. Analysis of lunar surface materials which are carried back to earth in a spacecraft can provide information on the energy, flux, and intensity down to about 10 MeV if excitation functions throughout this energy region are known. The lunar samples which have been returned to earth have been extensively analyzed for primordial and cosmogenic radionuclides. The quantities of the cosmogenic radionuclides in a lunar sample provide a basis for calculating the cosmic-ray spectra, exposure time, and flux incident on that particular sample if the radionuclides can be related to spallation production from specific elements. Several of the cosmogenic radionuclides which are generated in extraterrestrial materials result from the cosmic-ray proton spallation of iron.

If the complete excitation functions were known for each spallation product of each elemental con-