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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 constituent of a lunar sample, the knowledge of its exposure would be much more precise and indeed a chronological evaluation may be possible. If the excitation functions for the production of the various radionuclides in a meteorite and the elemental composition of that meteorite are known, much information can be obtained about its cosmic-ray exposure history and even perhaps about its orbital path and preatmospheric size. The excitation functions of the proton spallation of iron and titanium,[1] two principal heavy constituents of lunar soil and meteorites, have been determined in this laboratory.

Iron is perhaps the most widely studied of the major constituents of extraterrestrial material; however, additional data on many of the radioisotopes found in extraterrestrial materials were necessary. Also, some of the data in the literature seemed to be in error and an independent check was felt to be desirable. Furthermore, this comprehensive study will now allow iron foils, which are readily and inexpensively available in pure

TABLE I. Relevant properties of radionuclides measured.

Radioisotope	Half-life	γ-ray energy (MeV)	Branching intensity (%)	Production threshold [a] (MeV)	Reaction
^{57}Co	270 day	0.122	87	5.484 (V_C)	^{56}Fe(p, γ)^{57}Co
^{56}Co	77.3 day	1.239	66	5.484 (V_C)	^{56}Fe(p, n)^{56}Co
		1.038	15		
		0.847	100		
^{55}Co	18.2 h	1.409	13	5.541 (V_C)	^{54}Fe(p, γ)^{55}Co
		0.477	12		
^{54}Mn	303 day	0.835	100	9.479 (V_C)	^{57}Fe(p, α)^{54}Mn
^{52}Mn	5.60 day	1.434	100	13.345	^{56}Fe(p, αn)^{52}Mn
		0.935	84		
		0.744	82		
^{51}Cr	27.8 day	0.320	9	16.243	^{56}Fe(p, ^{6}Li)^{51}Cr
^{48}Cr	23 h	0.307	99	21.179	^{54}Fe(p, ^{7}Li)^{48}Cr
		0.111	98		
^{48}V	16 day	1.312	97	20.558	^{56}Fe(p, ^{9}Be)^{48}V
		0.984	100		
		0.944	10		
^{48}Sc	1.83 day	1.312	100	21.879 (V_C)	^{58}Fe(p, ^{11}C)^{48}Sc
		1.038	100		
		0.984	100		
^{47}Sc	3.43 day	0.158	73	18.112	^{58}Fe(p, 3α)^{47}Sc
^{46}Sc	83.9 day	1.121	100	18.728	^{57}Fe(p, 3α)^{46}Sc
		0.889	100		
^{44m}Sc	2.44 day	1.157	98.6	21.805 (V_C)	^{56}Fe(p, ^{13}C)^{44m}Sc
		0.270	86		
^{44}Ti	48 yr	1.157	100	20.336	^{54}Fe(p, ^{11}B)^{44}Ti
		0.078	98		
^{43}K	22.4 h	0.617	81	21.060	^{58}Fe(p, ^{12}Cα)^{43}K
		0.593	13		
		0.395	18		
		0.371	85		
^{42}K	12.36 h	1.525	18	20.649	^{57}Fe(p, ^{12}Cα)^{42}K
^{24}Na	15.0 h	1.369	100	26.153 (V_C)	^{57}Fe(p, ^{30}Siα)^{24}Na
^{22}Na	2.60 yr	1.275	100	26.384	^{57}Fe(p, ^{32}Siα)^{22}Na

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

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

Energy (MeV)	Radioisotope	σ (Expt.)	σ (Calc.)	σ/σ (^{54}Mn) (Expt.)	σ/σ (^{54}Mn) (Calc.)
14.1	^{57}Co	9.3 ± 1.3			
14.1	^{56}Co	398 ± 50			
15	^{57}Co	5.59 ± 0.77			
15	^{56}Co	212 ± 26			
29.5	^{56}Co	36.6 ± 4.9	898	0.938 ± 0.083	3.30
29.5	^{55}Co	65.8 ± 8.3	49.1	1.69 ± 0.13	0.180
29.5	^{54}Mn	39.0 ± 5.5	272	1	1
29.5	^{52}Mn	39.9 ± 4.9	4.39	1.023 ± 0.073	0.0161
30	^{56}Co	33.6 ± 8.0	882	0.881 ± 0.077	3.17
30	^{55}Co	57.5 ± 7.3	49.2	1.44 ± 0.11	0.177
30	^{54}Mn	40.0 ± 5.6	278	1	1
30	^{52}Mn	40.2 ± 4.9	4.65	1.006 ± 0.071	0.0167
44.6	^{56}Co	20.8 ± 2.9	567	0.112 ± 0.008	1.47
44.6	^{55}Co	10.1 ± 2.2	46.4	0.054 ± 0.010	0.12
44.6	^{54}Mn	186 ± 23	385	1	1
44.6	^{52}Mn	21.2 ± 2.6	13.9	0.1138 ± 0.0023	0.03602
44.6	^{51}Cr	142 ± 17	15.3	0.762 ± 0.015	0.0398
44.6	^{48}V	3.91 ± 0.54	0.292	0.0210 ± 0.0014	0.000759
44.6	^{48}Sc	0.0216 ± 0.0067	0.0181	0.000116 ± 0.000033	0.000047
45	^{57}Co	0.690 ± 0.086	4970	0.00368 ± 0.00011	12.9
45	^{56}Co	20.4 ± 2.6	561	0.109 ± 0.004	1.45
45	^{55}Co	11.5 ± 1.7	46.2	0.0614 ± 0.0052	0.120
45	^{54}Mn	187 ± 23	386	1	1
45	^{52}Mn	20.1 ± 2.5	14.1	0.1074 ± 0.0027	0.03656
45	^{51}Cr	142 ± 18	15.7	0.759 ± 0.021	0.0407
45	^{48}V	3.96 ± 0.55	0.306	0.0212 ± 0.0014	0.000793
120	^{56}Co	8.2 ± 1.5	150	0.249 ± 0.045	0.469
120	^{54}Mn	32.9 ± 5.7	316	1	1
120	^{52}Mn	12.9 ± 1.6	35.8	0.393 ± 0.050	0.113
120	^{51}Cr	43.9 ± 6.1	70.0	1.34 ± 0.19	0.222
120	^{48}Cr	0.345 ± 0.045	0.326	0.0105 ± 0.0014	0.00103
120	^{48}V	9.4 ± 1.2	7.45	0.288 ± 0.0013	0.0236
120	^{47}Sc	0.277 ± 0.044	1.28	0.0084 ± 0.0013	0.0041
120	^{44m}Sc	0.89 ± 0.12	1.48 [a]	0.0272 ± 0.0036	0.00469 [a]
320	^{54}Mn	22.9 ± 4.8	104	1	1
320	^{46}Sc	3.74 ± 0.88	732	0.163 ± 0.018	0.0707
320	^{44}Ti	≤0.14	0.5	≤0.0063	0.0036
320	^{22}Na	≤0.050	0.0224	≤0.0022	0.000216
434	^{54}Mn	16.0 ± 3.0	90.6	1	1
434	^{52}Mn	6.00 ± 0.99	19	0.375 ± 0.039	0.213
434	^{51}Cr	29.3 ± 5.2	51.7	1.83 ± 0.22	0.571
434	^{48}Cr	0.454 ± 0.079	0.619	0.0284 ± 0.0033	0.00683
434	^{48}V	10.8 ± 0.21	14.2	0.676 ± 0.068	0.156
434	^{47}Sc	1.31 ± 0.21	3.34	0.0819 ± 0.0079	0.0369
434	^{46}Sc	5.4 ± 1.4	9.5	0.33 ± 0.07	0.10
434	^{44m}Sc	3.59 ± 0.59	9.91 [a]	0.224 ± 0.022	0.109 [a]
434	^{44}Ti	≤1.8	0.54	≤0.11	0.0059
434	^{43}K	0.309 ± 0.096	0.76	0.0193 ± 0.0055	0.00844
434	^{42}K	1.7 ± 0.4	2.6	0.11 ± 0.02	0.029
434	^{22}Na	≤0.17	0.095	≤0.011	0.00105
585	^{56}Co	3.3 ± 1.1	18	0.160 ± 0.028	0.23
585	^{54}Mn	20.8 ± 6.7	78	1	1
585	^{52}Mn	6.5 ± 2.0	17.9	0.311 ± 0.031	0.230
585	^{51}Cr	29.6 ± 9.2	50	1.43 ± 0.15	0.640
585	^{48}Cr	0.55 ± 0.17	0.67	0.0263 ± 0.0026	0.00859
585	^{48}V	13.7 ± 4.3	15.3	0.659 ± 0.078	0.197
585	^{48}Sc	0.236 ± 0.076	0.95	0.0114 ± 0.0015	0.0122
585	^{47}Sc	1.90 ± 0.58	3.8	0.096 ± 0.0088	0.0482
585	^{46}Sc	5.4 ± 1.8	11	0.262 ± 0.036	0.142

TABLE II (*Continued*)

Energy (MeV)	Radioisotope	σ (Expt.)	σ (Calc.)	σ/σ (^{54}Mn) (Expt.)	σ/σ (^{54}Mn) (Calc.)
585	^{44m}Sc	5.2 ± 1.6	12.5 [a]	0.248 ± 0.024	0.160 [a]
585	^{44}Ti	0.25 ± 0.18	0.68	0.0118 ± 0.0077	0.0087
585	^{43}K	0.70 ± 0.23	1.00	0.0336 ± 0.0056	0.0129
585	^{42}K	2.49 ± 0.78	3.6	0.120 ± 0.014	0.0460
585	^{24}Na	0.266 ± 0.085	0.45	0.0128 ± 0.0017	0.00572
585	^{22}Na	0.260 ± 0.097	0.28	0.0114 ± 0.0022	0.00356

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

forms, to be used as flux monitor foils for reactions from 10 to 10's of thousands MeV, and for determination of both the spectra and flux of cosmic particles in extraterrestrial experiments.

A rather thorough comparison of the cross sections obtained from a theoretical equation with the experimental values for most of the useful spallation products is now possible. The yields for each observed radionuclide were calculated as a function of energy according to the semiempirical method of Rudstam,[2] and these were compared with the experimental values obtained by γ-ray analysis of spallation-product radionuclides in iron foils irradiated with protons at 10 different energies between 14.1 and 585 MeV.

II. EXPERIMENTAL PROCEDURES AND RESULTS

Pure iron foils 6.35 cm square and 0.005 or 0.051 cm thick were proton irradiated at 15, 30, and 45 MeV at the Berkeley 88-in. cyclotron, at 120 MeV at the Harvard University cyclotron, and at 320, 434, and 585 MeV at the Space Radiation Effects Laboratory (SREL) cyclotron. The number of protons incident on each target was monitored at the Harvard and SREL cyclotrons by proportional counters and by measuring the amount of ^{24}Na activity produced in aluminum monitor foils according to the reaction ^{27}Al(p, 3pn)^{24}Na. The cross section for this reaction was taken to be 10.8, 11.3, 11.0, and 10.8 mb at 120, 320, 434, and 585 MeV, respectively.[3] The beam currents during the Berkeley irradiations were integrated with uncertainties of less than 5%, by a calibrated Faraday cup. Integral proton doses ranging from 1 to 8×10^{13} protons were incident on the targets exposed at Harvard and SREL during irradiation times of 17 to 60 min. Proton doses of ~3×10^{15} total protons were obtained at Berkeley during exposure periods of 10 min or less.

The radionuclides produced in the iron foils were measured nondestructively with multidimensional anticoincidence shielded NaI(Tl)[4,5] and Ge(Li) γ-ray spectrometers[6] following decay periods varying from 40 hours to several months. Those radio-

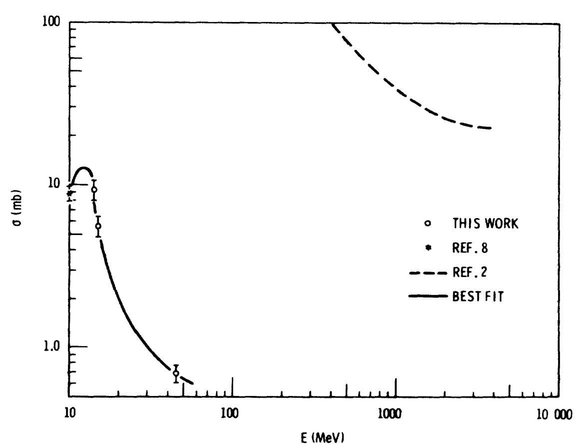

FIG. 1. Proton spallation cross sections for production of ^{57}Co from iron.

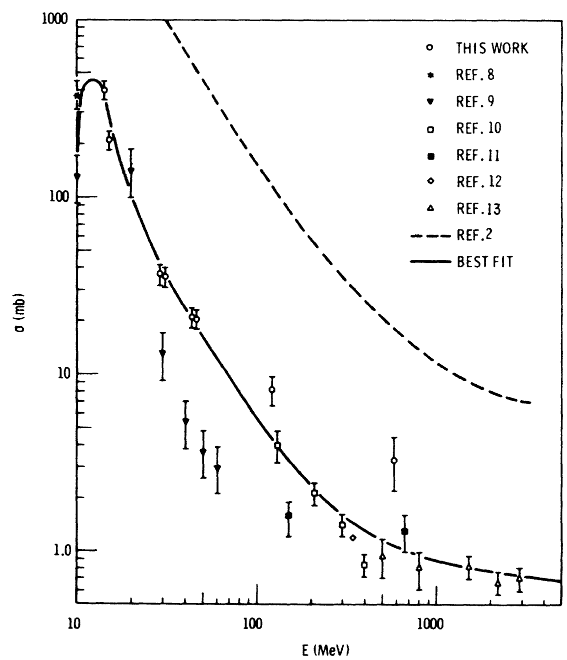

FIG. 2. Proton spallation cross sections for production of ^{56}Co from iron.

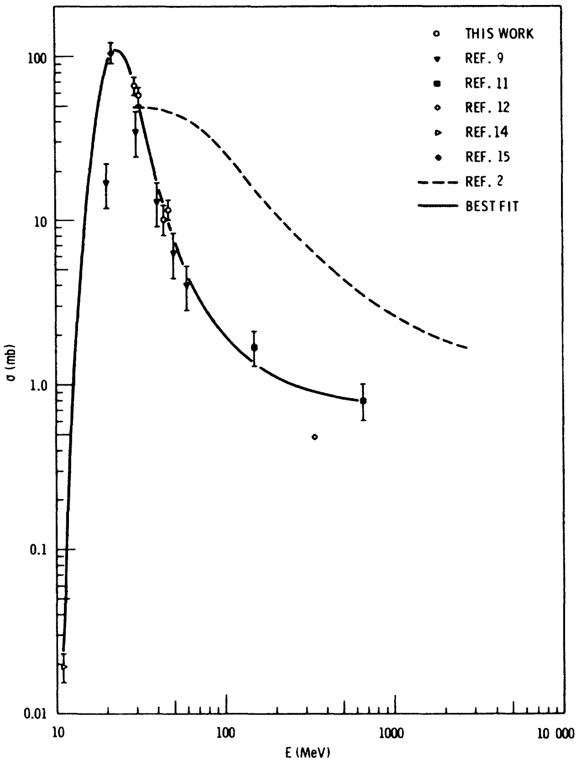

FIG. 3. Proton spallation cross sections for production of ^{55}Co from iron.

isotopes measured in this work are listed in Table I along with the half-lives, γ-ray energies, and branching intensities[7] used for their identification and yield calculations. Also listed in Table I are the minimum proton kinetic energies necessary to produce each radioisotope and the corresponding reaction. When the Coulomb barrier is higher than the Q value, the threshold value is followed by (V_C) to indicate that the value is based on the Coulomb barrier for the reaction listed. All

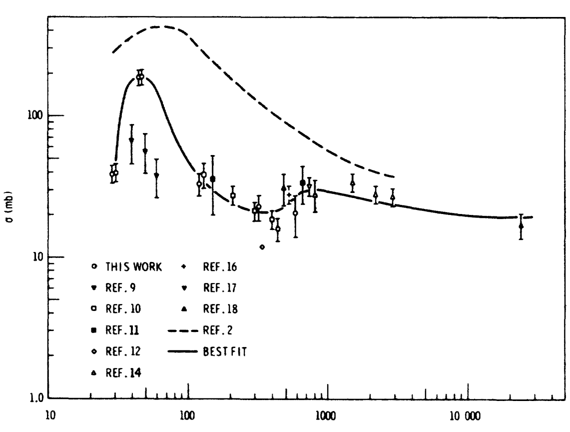

FIG. 4. Proton spallation cross sections for production of ^{54}Mn from iron.

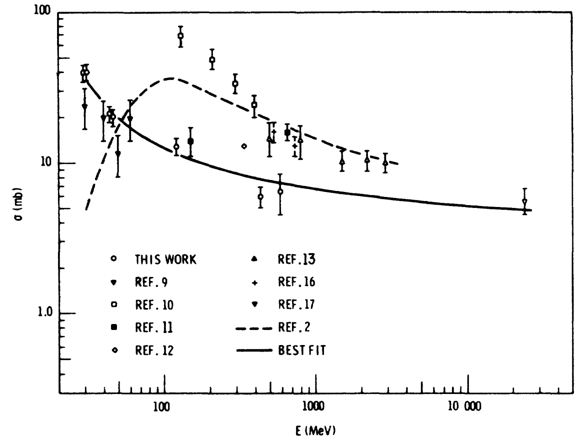

FIG. 5. Proton spallation cross sections for production of ^{52}Mn from iron.

thresholds are reported in the laboratory frame of reference. No activities were observed which were attributable to the spallation of any element heavier than iron. In addition, cobalt and sodium were the only light impurities which could be found in the target material by instrumental neutron-activation analysis and these at levels of only 1 and 4 atom ppm, respectively. Thus, the effects of impurities present in the iron are negligible.

The error values quoted for the experimental data, which are summarized in Table II, are consistent with the errors associated with counting statistics, counter calibrations, proton fluxes, and backgrounds. Since a rather large uncertainty is associated with the proton flux at the four highest energies, the ratio of the cross section for each observed radioisotope to that of ^{54}Mn at each energy is also given in the table. ^{54}Mn was chosen as the normalizing isotope since it is produced over the largest energy range with a maximum statistical accuracy. All radioisotopes were

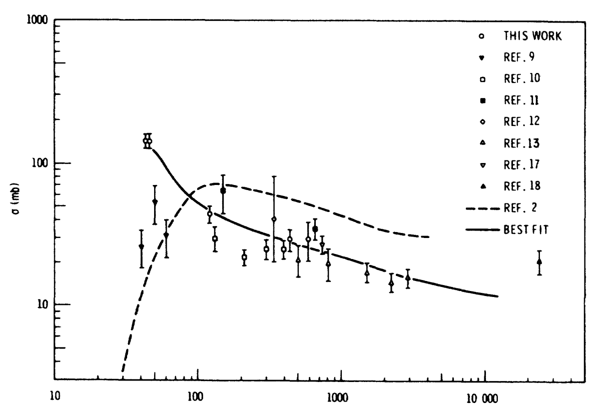

FIG. 6. Proton spallation cross sections for production of ^{51}Cr from iron.

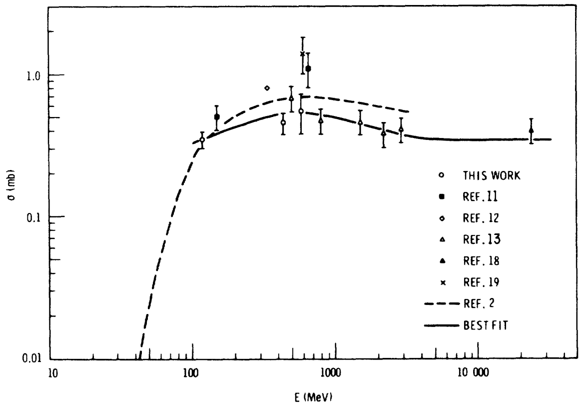

FIG. 7. Proton spallation cross sections for production of ^{48}Cr from iron.

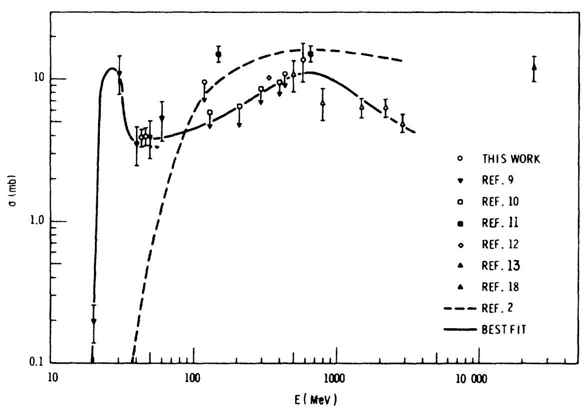

FIG. 8. Proton spallation cross sections for production of ^{48}V from iron.

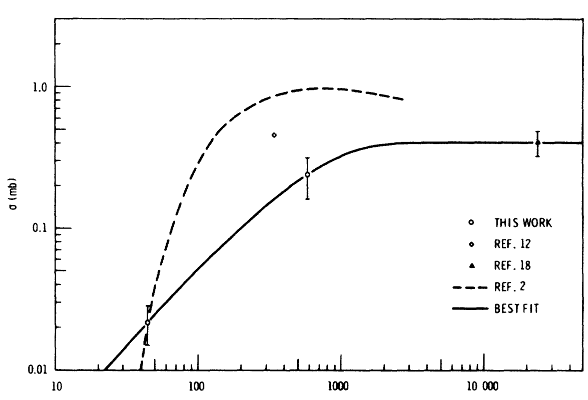

FIG. 9. Proton spallation cross sections for production of ^{48}Sc from iron.

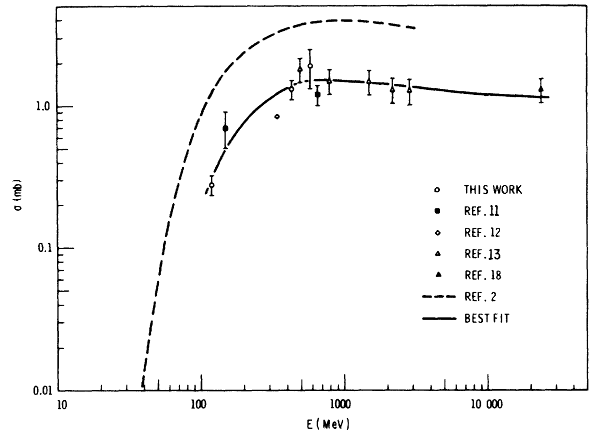

FIG. 10. Proton spallation cross sections for production of ^{47}Sc from iron.

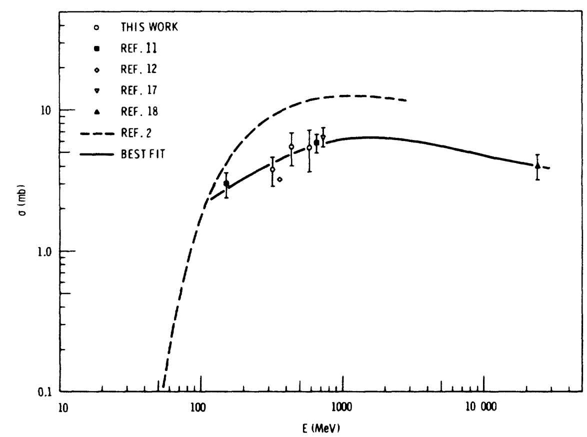

FIG. 11. Proton spallation cross sections for production of ^{46}Sc from iron.

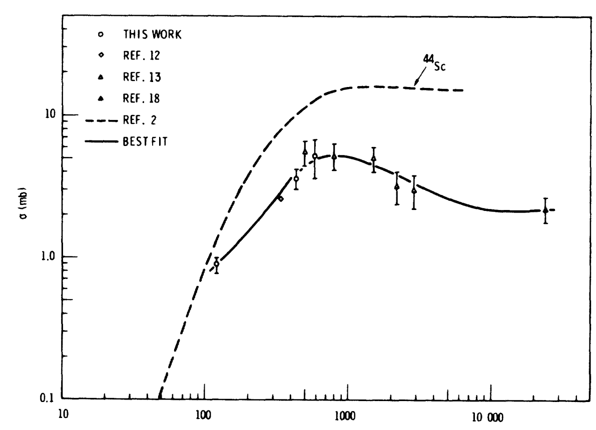

FIG. 12. Proton spallation cross sections for production of ^{44m}Sc from iron.

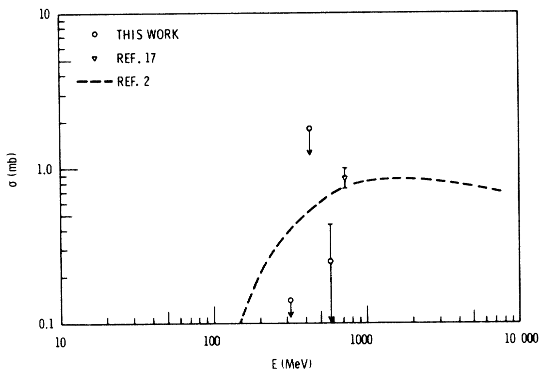

FIG. 13. Proton spallation cross sections for production of ^{44}Ti from iron.

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 these ratios of isotope yields, and only the uncertainties associated with the counting statistics remain.

III. DISCUSSION

Probably as much work has been done on the proton-induced spallation of iron as on any other element. With the data reported herein added to the existing work, a rather complete study is obtained. This large quantity of experimental data affords an opportunity to evaluate the semiempirical cross-section formulas of Rudstam[2] with regard to the masses of spallation products and the bombarding energies at which the equation will most accurately generate cross sections for the spallation products from a medium-weight element. Rudstam's theoretical formula which best fits the data is his CDMD cross-section equation, a five-parameter equation corresponding to an exponential yield-mass distribution and a Gaussian charge distribution, obtained by fitting it to the experimental data available at that time. The calculated cross sections, ratios of cross sections, and the experimental data are listed in Table II for comparison. Excitation functions were generated for all observed radioisotopes and are illustrated in Figs. 1-17. Appropriate data from the published literature are also plotted in each figure,[3, 8-22] and the best smooth curve is drawn through the data for each radioisotope as a solid line. The agreement between the data from the present work and the "best fit" curves for those spallation products which have been well studied substantiates the reliability of the procedures used in this work. The data of Tanaka and Furukawa,[8] based on enriched isotope irradiations, have been revised to represent the cross section expected from a natural iron target. The data points of Williams and Fullmer[9] have been read from their graphs, and although their data for ^{51}Cr, ^{52}Mn, ^{54}Mn, ^{55}Co, and ^{56}Co are systematically low, their general shape of the excitation functions appears to be good and might possibly be used to extrapolate the results reported herein to lower energies after appropriate adjustment of the curves has been made. The ^{56}Co data of Rayudu[10] has been multiplied by 0.9418 before plotting it in Fig. 2 so that the cross section would be that expected from natural iron. This paper shows that the reported cross sections are based on 96.18% of the weight of iron, but this percentage was meant to be 94.18%,[23] which is the sum of the isotopic abundances of ^{56}Fe, ^{57}Fe, and ^{58}Fe. The ^{56}Co data of Rudstam, Stevenson, and Folger[12] plotted in Fig. 2 has been multiplied by 5 to account for the positron branching ratio of only 20%; they quote very few error values except to say that some of their data are "uncertain." The

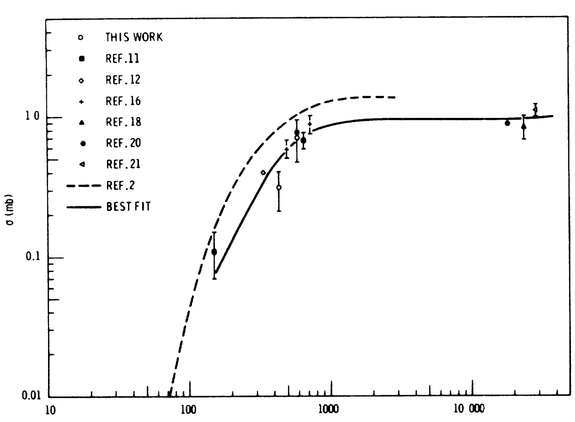

FIG. 14. Proton spallation cross sections for production of ^{43}K from iron.

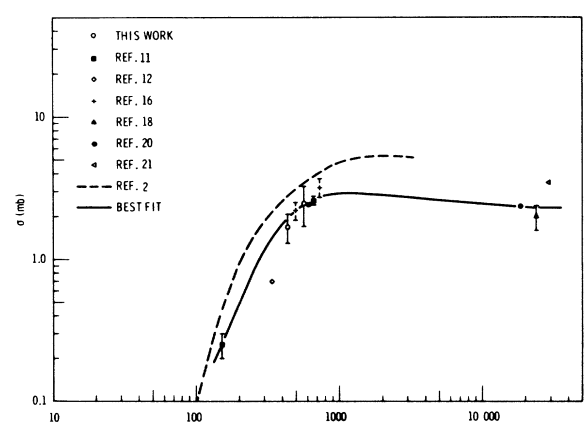

FIG. 15. Proton spallation cross sections for production of ^{42}K from iron.

^{55}Co data of Cohen[14] plotted in Fig. 3 have been corrected by the percentage of ^{54}Fe in natural iron to again have the data be representative of what would be expected from a natural iron target. The ^{52}Mn data of Rayudu[10, 13] plotted in Fig. 5 are too high, probably due to a positron-emitting impurity in his sample, since he measured only the positron annihilation radiation and not the 0.744-, 0.935-, or 1.434-MeV γ rays emitted in the decay of ^{52}Mn. Rayudu[10] considers his ^{48}V data to be lower limits for cumulative yields; therefore, they must be upper limits for independent yields of ^{48}V and are plotted in Fig. 8 as such. The ^{44}Ti data of Honda and Lal[17] plotted in Fig. 13 have been corrected to correspond to the more recently accepted value of 48 years for the half-life of this isotope; their assumed value was 200 years.

A cursory examination of the figures demonstrates the extent of agreement between the theoretical excitation functions as generated by Rudstam's CDMD equation and the experimental excitation functions. The differences between them as a function of energy or mass of the product nuclide are also evident. The theoretical curve plotted in Fig. 12 is for ^{44}Sc and therefore would not be expected to agree with the experimental data, which are for ^{44m}Sc. The agreement over the entire energy range is best for the lightest fragments (almost identical curves for ^{24}Na) and steadily degenerates as the fragments get heavier until at products of similar mass as that of the target, agreement is nonexistent. The disagreement at similar target and product masses is a well-known shortcoming of the Rudstam equation. The theoretical excitation function maintains a shape similar to the experimental curve until the product gets to be as heavy as about mass 48. At this mass and heavier, the theoretical curve parallels the experimental one at high energies but not at low energies. In almost all cases of disagreement, the theoretical curve is higher than the experimental. Indeed, in the high-energy proton-induced spallation of argon,[24] the Rudstam theoretical curves fit the experimental data better if multiplied by 0.81. It appears that the theoretical data for iron would fit the high-energy experimental data better if multiplied by 0.66; however, this would not help to adjust the shape of the theoretical curve at low energies and high product masses. The high-energy spallation cross sections for 12 isotopes from titanium, the element midway between argon and iron, have been measured,[1] and a constant multiplier of 0.79 was necessary to optimize the agreement between the theoretical and the experimental data. This suggests that a reduction factor which is not linearly dependent on the target mass should be introduced into the semiempirical equation.

Care should be exercised in attempting to extrapolate the excitation functions in the figures to the threshold energies given in Table I, particularly for spallation products more than a few mass units removed from the target. Although the products can theoretically be formed by the given reactions at the threshold energy, the cross section is likely to be so infinitesimal as to be zero for practical purposes. Effective thresholds, energies at which the spallation products are produced in sufficient abundance to be detectable, may be much higher, as in the case of the sodium isotopes where the effective thresholds are over 200 MeV.

These rather complete excitation functions can be usefully applied to the analysis of the cosmogenic radionuclide content of extraterrestrial materials and can perhaps elucidate some information regarding the cosmic-ray proton flux and spectrum incident on the materials when used in conjunction with an assay of the amount of iron present. For example, the isotopes of chromium,

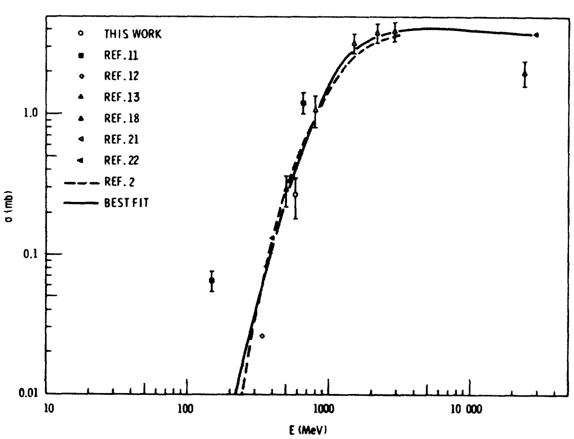

FIG. 16. Proton spallation cross sections for production of ^{24}Na from iron.

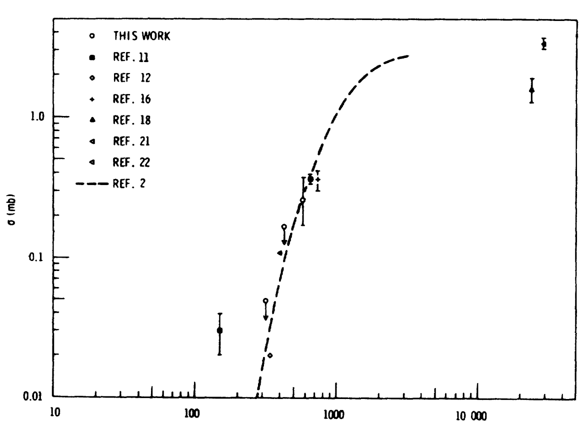

FIG. 17. Proton spallation cross sections for production of ^{22}Na from iron.

manganese, and cobalt are produced in meteorites and lunar surface samples principally by cosmic-ray spallation of iron. Due to the differences in the peak energy of some of these excitation functions, such as 10 to 20 MeV for ^{57}Co and ^{56}Co, 20 to 35 MeV for ^{55}Co, and 35 to 80 MeV for ^{54}Mn, an energy spectrum of incident particles can be determined from a measurement of the relative concentrations of these radioisotopes. In addition, since these isotopes have half-lives ranging from 18.2 h to 303 days, their relative concentrations can be employed to determine variations in the cosmic-ray flux over the last several years.

These excitation functions also provide for the use of iron foils as monitors for high-energy proton beams. For example, measuring the quantity of an isotope such as ^{24}Na produced in an exposed iron foil can be done with a very high degree of sensitivity. Large-crystal multidimensional analyzers[4,5] such as those at our laboratory can measure this isotope with an efficiency of 6.8% in a background field of 0.018 counts/min with no (<1%) Compton interference from the presence of as much as 10^7 dis/min of other radioisotopes. From the known cross section of a measured radionuclide, the number of protons incident on an iron foil at any energy can be determined. Similarly, by measuring more than one radionuclide, the energy of an unknown beam can be determined, and, in the last step, by measuring several radionuclides, a particle spectrum can be obtained. Once a particle spectrum and flux are known, other parameters, such as radiation dose from exposure, can be calculated by integration of the energy deposition in a body. Such applications are particularly adaptable to the cosmic environment of space.

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