Cross sections for production of stable and long-lived nuclides by high energy spallation of iron; cosmic ray implications

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The cross sections for production of the stable and long-lived isotopes of Be, Sc, V, Cr, and Mn by proton spallation of Fe have been measured at 600 MeV and 21 GeV. The experimental method involves the contamination free separation of the spallation products from ultrapure iron targets and the mass spectrometric determination of their isotopic ratios. To derive absolute cross sections, radioactive isotopes are used as references. For this purpose, the cross sections of a few radioactive isotopes have been measured at 21 GeV by γ-ray spectrometry. The results are compared with the values obtained by means of semiempirical formulas. The overall agreement is good but the cross sections for formation of manganese isotopes are found to be significantly higher than predicted. The implications of the results for path-length distribution and source abundances of cosmic ray nuclei with $21 \leq Z \leq 26$ are discussed. Calculated cosmic ray isotopic distributions are presented for some elements, which illustrate the potential resources of electron capture isotopes.

[NUCLEAR REACTIONS $Fe(p,x)^{10}Be$, ^{45}Sc, $^{49,50,51}V$, $^{50,52,53,54}Cr$, $^{53,55}Mn$, $E=600$ MeV; $Fe(p,x)^{7,9,10}Be$, ^{22}Na, $^{45,46}Sc$, $^{48,49,50,51}V$, $^{50,51,52,53,54}Cr$, ^{54}Mn, ^{56}Co, $E=21$ GeV; measured σ's; deduced cosmic ray abundances; natural target.]

I. INTRODUCTION

The nuclear reactions undergone by cosmic ray nuclei during their passage through interstellar matter modify their composition in such an important way that some cosmic ray species are believed to be almost entirely "secondary" in origin, i.e., almost completely produced by such reactions. These transformations may be the major source of information on the conditions of cosmic ray propagation if, along with the cosmic ray composition near earth, one knows the cross sections of the relevant nuclear reactions.

Due to the time scale of cosmic ray propagation, cross sections need to be known for three kinds of product isotopes: (1) stable isotopes, (2) long-lived radioactive isotopes (with half-lives longer than about 10^6 years), (3) radioactive isotopes decaying only by electron capture (cosmic ray particles are generally completely stripped of their orbital electrons, so that these nuclides may have much longer lifetimes in cosmic rays than at rest).

The cross sections for production of the light secondary elements Li, Be, and B have been extensively studied for many years in this laboratory (see Ref. 1 for a review) and recently at Berkeley,[2] and they can now be considered as sufficiently well known for interpreting cosmic ray data.

This is not true for a similar group of secondary elements with $21 \leq Z \leq 25$ (Sc, Ti, V, Cr, Mn), produced by spallation of iron nuclei and for which good cosmic ray data are now available. Indeed, there was, until now, a total lack of cross-section data for the stable isotopes of these elements and partial results for isotopes of kinds (2) and (3). This situation has led us to undertake a systematic study of the production of the isotopes of the three kinds mentioned above for elements with $21 \leq Z \leq 25$ by proton spallation of iron at high energies. Because of the important role of ^{10}Be as a "cosmic ray clock" we have also studied the production of beryllium isotopes. In this article, we present the first set of results we have obtained and attempt to show what implications they may have for cosmic ray physics.

II. EXPERIMENTAL

From the experimental point of view, the problem that we had to face essentially lies in measuring the quantities of stable isotopes produced in an iron target by proton bombardment. As proton beam intensities and irradiation times are, of course, limited, these quantities are small. The use of circulating beams increases the number of protons that bombard the target, thanks to multitraversals, but the intensity received by the target is not uniform. It is maximum at the edge and rapidly decreasing towards the interior, so that in fact only a small part of the target is really useful. Finally the target must not be too thick, in

order to keep at a negligible level the contribution of low energy secondary particles (moreover, increasing the thickness of the target decreases the number of multitraversals).

Typically we have had to work with concentrations of about 10^{-8}–10^{-7} g/g for a given isotope depending on its formation cross section, and absolute quantities of the order of 1 or a few 10^{-10} g for each measurement. This poses essentially three requirements:

(1) The target must be pure enough to make sure that the impurities initially present are of negligible, or at least small, concentration compared to the spallation products we want to measure.

(2) The physical and chemical treatments of the target must not introduce more than a negligible amount of contamination by impurities compared to spallation products.

(3) We need an instrument capable of measuring these small quantities. We have used a high-sensitivity mass spectrometer. Since this kind of instrument measures isotopic ratios, it brings in a fourth requirement: absolute cross sections have to be obtained with respect to some standards, which led us to measure the cross sections of some radioactive nuclides.

We shall now describe the experimental procedure, laying stress only on the points which make the present study different from those previously carried out in this laboratory. More details have been given in Ref. 3.

A. Targets and irradiations

The choice of the targets was of course a very crucial one. The purity level needed is not normally achieved in commercially available products. We used samples of natural iron supplied by the Laboratoire de Physique du Solide, Centre d'Etudes Nucléaires de Grenoble, where they are prepared with extreme care, by the zone refining method.[4] The concentrations of impurities in the series of samples we have used for our measurements are listed in Table I for the elements we have studied. The way we obtained these values is explained below. One can see that the purity of these samples was quite sufficient for our study. The targets were approximately 15 mm × 7 mm × 0.2 mm in dimension. The irradiations were performed at CERN, three at the synchrocyclotron (600 MeV protons), and one at the synchrotron (21 GeV protons). The circulating beam was used at both accelerators. The number of protons that traversed the targets ranged from 1 to 3×10^{18}.

TABLE I. Concentration of impurities in the iron targets.

Element	Be	Sc	V	Cr	Mn
Concentration (10^{-9} g/g)	< 0.1	< 1	5	< 35	< 10

After bombardment, the targets were cut up into small pieces, 5 to 10 mg each, by means of a tungsten blade, any steel instrument being prohibited in this study. Several measurements could thus be made from one target. We made a quantitative determination of the impurity contamination in the following way. We took advantage of the fact, already mentioned, that the target is not uniformly bombarded by the proton beam. This means that the concentration of spallation products varies widely from one part of the target to another, while it is reasonable to assume that the concentration of impurities remains constant on the scale of a target. It follows that, if not completely negligible, the relative contamination will be different from a part of a target to another and thus the isotopic ratios measured in two pieces with different concentrations of spallation products (i.e., different specific radioactivity) will be different. It is easy, from this measured difference, to calculate the correction to be made and the spallation ratio. So, apart from the analysis of the most irradiated parts of the targets, we made measurements on pieces with specific activity about 10 times smaller. Under these conditions, the measured ratio of a stable isotope to a radioactive isotope would vary by almost a factor of 2 between the two series of target pieces for a 10% contamination in the most irradiated ones. The relative contamination we have deduced by this method is indicated in Table II, for the stable nuclides we have studied. As absolute quantities of radioactive isotopes can be measured by γ spectrometry, we have deduced the absolute quantities of impurities in the iron targets, the values of which have already been given in Table I.

B. Chemical separation

Before analysis by mass spectrometry, each spallation-produced element to be studied had to be chemically separated from iron and from the other spallation products. It soon became clear that this could not be achieved by means of simple physical treatments as has been successfully done for oxygen[5] and carbon[6] targets, but that we had instead to use a more sophisticated chemical technique. We chose that developed by Mérinis and Bouissières,[7] sometimes called thermochromatography. It had only been used, to our knowledge, for the separation of radionuclides, but it proved to be quite adaptable for the contamination-free

TABLE II. Relative contamination by natural impurities in the target pieces with a high specific activity. The contamination decreases with time for ^{51}V, since the concentration of this nuclide increases due to ^{51}Cr decay ($t_{1/2}=27.8$ day).

Nuclide	^{9}Be	^{45}Sc	^{50}V	^{51}V	^{50}Cr	^{52}Cr	^{53}Cr	^{55}Mn
Contamination (%)	<1	<1	0	15±5 to 3±1	<1.5	<10	<6	<10

separation of very small quantities of stable isotopes.

The experimental setup is quite similar to the one used by Mêrinis and Bouissières.[7] A temperature gradient is established along a quartz tube (1 m long, 2 mm internal diameter) by putting it inside a thick-walled copper tube, one end of which is heated to about 800°C by an electric furnace, the other one is cooled to −30°C by means of a freon compressor. The temperature varies smoothly in between.

A piece of an irradiated iron target is introduced into the hot part of the quartz tube. Chlorine is then admitted at this hot end and allowed to flow slowly inside the tube. The action of chlorine is twofold: first it chlorinates iron and the spallation products, then it carried along the resulting chlorides, which move slowly towards the colder parts and finally stop at a temperature, and thus a position, which is characteristic of each element. At the end of the separation (i.e., after about 4 h) the different elements are located in the tube by detection of characteristic γ rays of their radioisotopes. The quartz tube is then cut into pieces, about 5 cm long, on both sides of each deposit. Each element is desorbed by a drop of water or nitric acid, then evaporated on a wide tantalum or platinum filament which serves as a sample plate for the mass spectrometer. The yields of the separation need not be precisely known, nor be the same for all elements (and indeed they are not), since only isotopic ratios are directly measured. More details on this chemical technique will be published elsewhere.

It is clear that through this procedure and with a few precautions the contamination by natural impurities can be reduced to a very low level: before a separation, the whole quartz tube was heated to about 900°C, in a chlorine flow, for a few hours. Under these conditions, all the impurities that could be present on the inner wall are chlorinated and blown out of the tube, at least for the elements of interest in this study. Also the target was etched by nitric acid to remove a very likely surface contamination. Ultrapure reagents were used: water was prepared in our laboratory by several steps of ion exchange, distillations, and subboiling distillation; nitric acid, prepared by double subboiling distillation was supplied by the National Bureau of Standards. In both reagents, the impurities we are concerned with were present at the 10^{-11} g/g level, and only a drop (10 to 20 mg) was sufficient to desorb 60 to 80% of a deposit. The contamination due to the target itself was thus by far the most important one. It should be noted, however, that the method we used to determine it gives in fact the contamination introduced by the whole procedure.

C. Obtaining isotopic ratios

The mass spectrometer is an ion sputtering instrument, already used for many studies on lithium, beryllium, and boron in this laboratory. A description is given in Ref. 6 and, for its earlier version, in Ref. 5. The sample, obtained as we have indicated, is bombarded by a 6 keV Cs^+ ion beam of about 10^{-9} A. The secondary sputtered ions pass through a magnetic analyzer and an electrostatic one and are detected by an electron multiplier associated with an ion counting system including a 4000 channel multiscaler. Following Collins and McHugh[8] we have allowed the sample to be heated during the analyses at a temperature of 700°C or higher to remove the hydrocarbon peaks. 20 to 30 spectra were recorded during an analysis, each one consisting in 50 to 200 scans of 4 to 10 seconds. The standard deviation on isotopic ratios was of a few percent. A correction was applied to allow for isotopic discrimination. In the case of beryllium, we have adopted, for the discrimination factor, the value determined by Raisbeck, Lestringuez, and Yiou[9]: 1.11 ± 0.05. For the elements Sc, V, Cr, and Mn we have used the value 1.02 ± 0.02, deduced from the analysis of natural samples.

D. Obtaining absolute cross sections

In order to get absolute cross sections from isotopic ratios, we needed to know the cross sections of some radioactive isotopes. Some strong disagreements exist between published values. A study has been recently carried out at 600 MeV by three laboratories.[10] Their results are in good

TABLE III. Nuclear properties of isotopes studied (Ref. 13).

Nuclide	Half-life (days)	γ-ray energy (keV)	Branching ratio
^{7}Be	53.6	477	0.103
^{22}Na	949	1275	1
^{46}Sc	84	889	1
		1120	1
^{48}V	16	983	1
		1312	0.98
^{51}Cr	27.8	320	0.098
^{54}Mn	312	835	1
^{56}Co	77.3	847	1
		1240	0.66

agreement, and we have adopted the mean values of their three measurements. These values are also in general good agreement with those previously obtained at 660 MeV by Lavrukhina *et al.*[11] Around 20 GeV, only one series of cross sections was available, obtained by Estrup[12] at 24 GeV, so that we decided to make a new measurement. For this purpose, a stack of 0.05 mm thick iron foils and 0.03 mm thick aluminum foils was bombarded by the external beam of the CERN proton synchrotron (PS). The proton energy was 21 GeV. The target foils had the shape of disks, 28 mm in diameter. The irradiation was done as a "parasite experiment" with a particle physics experiment; it lasted 13 days and 1.3×10^{17}protons were received by the target. After bombardment, the trace of the beam (about 1 mm in diameter) was checked by an autoradiography of the target, and a disk of 6 mm diameter was punched out around it. The inner iron and aluminum disks were then γ counted with a Ge(Li) detector having a relative efficiency of 11% and a resolution of 3.4 keV at 1.3 MeV and previously calibrated by means of standard sources. The nuclear properties we have used are listed in Table III for the nuclides studied.

Because of the long duration of the irradiation and of the presence near our target of another thick target, which could have been a source of secondary particles, we checked our results by comparing them with those obtained for another iron target, 0.2 mm thick, previously bombarded by the circulating beam of the CERN PS, without an aluminum monitor. This latter irradiation lasted 15 h, and the target received 1.4×10^{17} protons. The proton energy was 14 GeV, but this difference with the other irradiation should not introduce any appreciable variation of cross sections.[14,15]

III. RESULTS

Isotopic ratios are listed in Table IV, as determined by mass spectrometry, after correcting for isotopic discrimination, and, when necessary, natural contamination and decay of radioactive isotopes during and after bombardment. Measurements have been made at periods of time ranging from 1 day to about 1 yr after irradiation. The number of determinations of each ratio is also shown in Table IV. The uncertainties are obtained by taking into account the standard deviation on the measured values (generally a few percent) and the uncertainties on isotopic discrimination and contamination corrections, as given above. The larger uncertainties on ^{55}Mn/^{54}Mn and chromium ratios are mainly due to the contamination corrections since only upper limits have been determined in these cases (see Table II).

The cross sections of radioactive nuclides were measured with respect to the cross section of the monitor reaction ^{27}Al(p,$3p3n$)^{22}Na, taken to be 10.0 ± 0.5 mb after Cumming *et al.*[16] As already

TABLE IV. Isotopic ratios of iron spallation products measured by mass spectrometry. They correspond to complete decay of radioactive precursors whenever applicable, except for ^{51}V/^{50}V and ^{55}Mn/^{54}Mn, which include no contribution of ^{51}Cr and ^{55}Fe.

	600 MeV	No. of determinations	21 GeV	No. of determinations
^{9}Be/^{7}Be	...	...	0.71 ± 0.08	4
^{10}Be/^{7}Be	0.23 ± 0.04	1	0.40 ± 0.06	4
^{45}Sc/^{46}Sc	$3.3\ \pm0.2$	6	$3.0\ \pm0.2$	4
^{49}V/^{50}V	2.04 ± 0.12	6	1.85 ± 0.12	4
^{51}V/^{50}V	0.38 ± 0.05	6	0.31 ± 0.04	4
^{50}Cr/^{51}Cr	0.62 ± 0.06	4	0.60 ± 0.06	3
^{52}Cr/^{51}Cr	1.59 ± 0.3	4	1.82 ± 0.4	3
^{53}Cr/^{51}Cr	0.27 ± 0.04	4	0.34 ± 0.05	3
^{54}Cr/^{53}Cr	0.29 ± 0.06	4	0.27 ± 0.07	2
^{53}Mn/^{54}Mn	1.35 ± 0.09	3	...	...
^{55}Mn/^{54}Mn	0.86 ± 0.19	3	...	...

mentioned, a comparison was made with another target, without a monitor. The values were normalized to the cross section of ^{22}Na since it was the most precisely measured nuclide. The differences between the values obtained for the two targets were less than 4% for ^{7}Be, ^{46}Sc, and ^{54}Mn, 12% for ^{51}Cr and ^{56}Co, and 20% for ^{48}V. The less good agreement for ^{48}V, and also for ^{51}Cr, can be explained by the long duration of the external irradiation, comparable to their half-lives. The correction introduced is important for these two nuclides and has a large uncertainty. We have therefore finally adopted for ^{48}V and ^{51}Cr the cross sections obtained from the second target, and for the other nuclides we took the mean of the two values. The results are given in Table V.

The uncertainties are the standard errors, including the error on the cross section of the monitor reaction, the error on the number of atoms per cm^2 (1.5%) in the target and the monitor, the statistical error on the counting rates (generally less than 2%), the error on the relative efficiency of the Ge(Li) detector (0 to 10%), and an estimated error due to decomposition of the photopeaks (1 to 5%). No error was assigned to disintegration schemes.

Our results are compared with those of previous authors. It can be seen that there are some important discrepancies with the work of Estrup, particularly for ^{54}Mn. We also show in Table V recent values obtained independently in this laboratory by Raisbeck and Yiou at 23 GeV.[19,20] The agreement is fairly good here.

From these cross sections and the isotopic ratios given in Table IV one can deduce the cross sections of the stable and long-lived isotopes. At 600 MeV, as we already mentioned, we used the radioactive isotope cross sections of Orth *et al.*[10] In the case of vanadium, ^{48}V has a too short half-life to be used as a reference with our experimental technique and we preferred to calibrate the vanadium cross sections with respect to that of ^{51}Cr, by observing the variations in time of the vanadium isotopic ratios due to decay of ^{51}Cr to ^{51}V. By plotting the ratios ^{51}V/^{50}V and ^{51}V/^{49}V versus $1-\exp(-\lambda_{51}t)$, where λ_{51} is the decay constant of ^{51}Cr and t the time, one obtains straight lines whose slopes are equal to the ratios ^{51}Cr/^{50}V and ^{51}Cr/^{49}V. The ordinates at $t=0$ are equal to the ratios ^{51}V/^{50}V and ^{51}V/^{49}V. As a test of this method, we also derived in this way the cross sections of ^{48}V; though less precise, they are in good agreement with the values obtained by direct radioactivity measurements. The cross sections are listed in Table VI.[21] Many are cumulative, including contributions from short-lived isobars, as these are of course the quantities of interest for cosmic ray physics. The other ones are independent because they refer to isotopes which either were shielded by stable isobars (e.g., ^{50}V, ^{54}Mn) or had precursors sufficiently long lived to allow corrections by using measurements made at widely different times (^{54}Cr, ^{55}Mn). Unfortunately, we have presently no data reliable enough for ^{53}Mn and ^{55}Mn at 21 GeV, due to experimental problems: the separation Mn-Fe is rather critical and we only had one irradiation at 21 GeV with a sufficiently high proton flux. Also, preliminary results on titanium have shown us that the measurement is feasible but requires still more work.

Our results for ^{49}V can be compared with the values obtained by Lavrukhina *et al.*[11] at 660 MeV (32.3 ± 4.5 mb) and Estrup[12] at 24 GeV (22 ± 4 mb), and for ^{53}Mn with the value of Honda and Imamura[22] at 730 MeV (37 ± 15 mb). The agreement is good, within quoted uncertainties. The comparison with

TABLE V. Cross sections (mb) for production of radioactive nuclides by spallation of Fe by 21 GeV protons. The values of Estrup have been corrected for new values of the monitor reaction cross section (Ref. 16), and branching ratios (Ref. 13). The dagger (†) denotes independent cross sections.

Product	This work	Estrup [a]	Porile and Tanaka [b]	Régnier	Raisbeck and Yiou
^{7}Be†	11.4 ± 1.2	8.5		10.0 [c]	11.7 ± 1.0 [d]
^{22}Na	3.1 ± 0.2	1.7	3.4 ± 0.3	2.42 [e]	3.02 ± 0.25 [d]
^{46}Sc†	6.0 ± 0.5	4.1			6.3 [f]
^{48}V	12.1 ± 1.3	12.6			12.2 [f]
^{51}Cr	25.1 ± 3.2	20			26.8 [f]
^{54}Mn†	29.2 ± 2.7	17.6			31.6 [f]
^{56}Co†	0.78 ± 0.08				1.25 [f]

[a] Reference 12 (24 GeV).
[b] Reference 17 (29 GeV).
[c] Reference 18 (24 GeV).
[d] Reference 19 (23 GeV)
[e] Reference 18 (10 GeV)
[f] Reference 20 (23 GeV).

the cross section obtained by Honda and Lal[23] for ^{10}Be at 730 MeV is more difficult, since the cross section for formation of such a light product is expected to be very energy dependent in this energy range. We note, however, that our measured ratio ^{10}Be/^{7}Be at 600 MeV agrees well with their value of 0.22 ± 0.05, obtained after correcting for the new values of ^{7}Be branching ratio[13] and ^{10}Be half-life.[24]

It is also interesting to compare our experimental cross sections with those predicted by semiempirical formulas. This kind of formula, first derived by Rudstam,[25] to fit experimental data, and later extended by several authors, are widely used by workers in different fields, and in particular in cosmic ray physics, when spallation cross sections are needed and no experimental values are available. In Table VII, we show the ratios of cross sections calculated by means of the formulas of Silberberg and Tsao[26] to our experimental values. The overall agreement is surprisingly good, the mean value of the ratio being very close to 1, but some important discrepancies show up. In particular, there seems to be a systematic effect, at least for V and Cr, the ratio of calculated to experimental cross sections increasing with isotope mass. This is probably due to the fact that Silberberg and Tsao[26] had to use their general formula for $(p, 3pxn)$ and $(p, 4pxn)$ reactions in this mass range, rather than specific formulas for peripheral reactions, because of lack of experimental data.

TABLE VI. Production cross sections (mb) for Fe spallation. The asterisk (*) denotes values used as references for determination of the other cross sections. The dagger (†) denotes independent cross sections.

Product	600 MeV	21 GeV
Be 7†	2.01 ± 0.13* [a]	11.4 ± 1.2*
9		8.1 ± 1.2
10†	0.46 ± 0.09	4.6 ± 0.8
Sc 45	27.9 ± 1.9	18.0 ± 1.9
46†	8.45 ± 0.27* [a]	6.0 ± 0.5*
V 49	38.0 ± 3.0	18.6 ± 3.2
50†	18.0 ± 1.1	10.0 ± 1.6
51	6.8 ± 1.0	2.9 ± 0.6
Cr 50	27.2 ± 2.8	15.1 ± 2.4
51	43.8 ± 1.7* [a]	25.1 ± 3.2*
52	69.6 ± 13	45.7 ± 11.6
53	11.8 ± 1.8	8.5 ± 1.7
54†	3.4 ± 1.0	2.4 ± 1.0
Mn 53	45.0 ± 4.0	
54†	33.3 ± 1.6* [a]	29.2 ± 2.7
55†	28.6 ± 7.0	

[a] Reference 10.

Although the present results could help in refining the formulas, more data are certainly still needed.

IV. DISCUSSION

The production of cosmic ray secondary nuclides by nuclear interaction between primaries and the interstellar medium is a powerful probe of cosmic ray propagation. Indeed, most of the current ideas on propagation arise from the study of the two main groups of secondaries: Li, Be, B on one hand, Sc, Ti, V, Cr, Mn on the other hand. It has been shown (e.g., Ref. 27) that, in order to explain the abundances of both groups below a few GeV/n, one was led to assume an exponential or exponentiallike distribution of cosmic ray potential path lengths with a mean value of about 5 g/cm^2. The nuclear cross sections used to obtain these results were, to a large extent, experimental ones for the light element group, but only estimated ones for iron secondaries. It is thus interesting now to see the implications of our present measurements.

Most workers have, in their propagation calculations, used the cross sections calculated by Silberberg and Tsao[26] for iron spallation. It is therefore obvious from Table VII that no dramatic change in propagation models will be implied by our experimental results, since on the average

TABLE VII. Ratios of cross sections calculated after Silberberg and Tsao (Ref. 26) to experimental cross sections.

	600 MeV	21 GeV
Be 7	0.89	0.75
9		0.62
10	1.70	0.96
Sc 45	0.57	0.89
46	0.65	0.83
V 48	0.66	0.83
49	1.10	1.56
50	1.44	1.60
51	1.90	2.40
Cr 50	0.73	0.93
51	1.00	1.04
52	1.15	0.92
53	1.76	1.18
54	1.18	1.67
Mn 53	0.80	
54	0.63	0.55
55	0.86	
Mean	1.06	1.12
Standard deviation	0.42	0.48

our values are very close to the previously calculated ones. However, the standard deviations in Table VII are rather large, and the differences between experiment and calculation can be important for some individual elements and, even more, individual isotopes.

We note first that the calculated ratio $^{10}Be/^{7}Be$ at 600 MeV is almost twice the experimental one and, at 21 GeV, the calculation gives a ratio $^{10}Be/Be$ which is about 30% too high. ^{10}Be, with its half-life of 1.5×10^6 years,[24] is a specially important nuclide, as it could possibly be used to determine the cosmic ray lifetime and hence the mean density of the propagation medium. Certainly, our measurement at 600 MeV plays no important role in this connection, because the cross sections at this energy are too small for iron to be compared with lighter targets, but the situation is different at higher energy. A recent measurement of the isotopic composition of cosmic ray Be near 100 MeV/n[28] seems to indicate that most of ^{10}Be has decayed. It would be interesting to measure Be isotopic abundances at much higher energy, where, due to dilation of its half-life, a larger proportion of ^{10}Be should be present. Further, cosmic ray measurements at very high energies[29-32] have shown that the abundance of iron increases with energy relative to lighter primary species like C, O, Si, while the relative abundance of B (a major progenitor of Be) decreases, even though there are some discrepancies on the magnitude of these variations. All this tends to increase the weight of Fe as a source of Be at high energy. Indeed, if one adopts for the production of ^{10}Be from oxygen the cross section measured by Yiou,[5] the contribution of Fe to the production of cosmic ray ^{10}Be above 10 GeV/n could be about as high as that of O.

We now consider the heavier secondary products of iron. The most striking discrepancy between experiment and calculation (see Table VII) arises for manganese. Even though there are no experimental values for ^{53}Mn and ^{55}Mn at high energy, it is clear that the calculated values are substantially too low. It follows that manganese production during cosmic ray propagation will be larger and the chance for manganese to be present at the source in non-negligible amounts still smaller than predicted before.

We have investigated in a more quantitative way the propagation of cosmic ray iron nuclei by means of the PCRNOR computer code. This program is the one written by Comstock[33] which allows for ionization energy losses and all possible nuclear reactions, and recently improved in order to also take into account the electron pickup and loss processes and hence the possibility of decay of electron capture isotopes.[34]

The cosmic ray source spectrum has been assumed to be a power law in total energy per nucleon of index −2.5. The propagation medium was pure hydrogen. The importance of neglecting the contribution of interstellar helium has been discussed by Raisbeck and Yiou,[35] on the basis of their measurements of the production of radionuclides by α spallation. Their results on iron spallation for products in the mass range of interest here show that the ratio of α cross sections to proton cross sections is roughly the same for all products, and close to 1 at energies about 0.6–1 GeV/n. At higher energies, this ratio seems to increase to about 1.5. On these bases, neglecting the ~10% abundance of helium in the interstellar gas will only lead to underestimation by roughly 20% of the path length traversed by cosmic rays, with no appreciable change in the calculated relative abundances.

From our experimental cross sections, we extrapolated excitation functions down to 100 MeV,[3] by analogy with known similar reactions and Silberberg and Tsao's calculations. We feel quite confident in these extrapolations for the higher energies, where cross sections vary slowly. Our excitation functions are more questionable at energies lower than about 300 MeV where cross sections often start to vary more rapidly, to form a peak near or below 100 MeV. This is however not too critical for propagation calculations, as ionization energy losses are important in this energy region and most of the products are in fact formed at a significantly higher energy.

For all reactions for which we had no experimental data, we used the formulas of Silberberg and Tsao,[26] i.e., for formation of tertiary and higher order products and also of titanium from iron. These reactions contribute to the production of ~6% of Mn, ~17% of Cr, ~38% of Sc (including those which transform a nuclide into an isotope of the same element). In other words, the larger part of each species is produced through one reaction, whose cross section is measured, while the rest is produced through several or many reactions whose cross sections are estimated. It is clear, therefore, that the calculated abundances will rely on essentially experimental nuclear data.

We have computed the interstellar abundances of all stable and electron capture isotopes with $21 \leq Z \leq 26$, as a function of energy, assuming different distributions of path length. We have grouped isotopes to derive elemental abundances, as there are presently no reliable cosmic ray data on isotopes in the mass region under study here. The results are compared with experimental data in Figs. 1–4. Assuming the cosmic ray sources only emit iron, with its solar system isotopic composi-

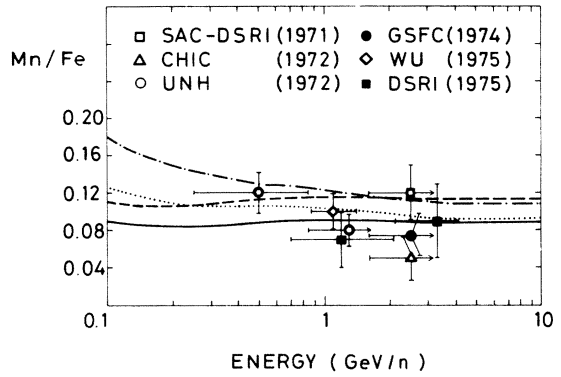

FIG. 1. Cosmic ray abundance of Mn, relative to Fe. The curves give the interstellar abundance calculated for an exponential distribution of path length with an escape length of 5 g/cm^2 (full line) and 10 g/cm^2 (dashed line), a slab of 2 g/cm^2 (dash and dot line), and a Gaussian centered about 2 g/cm^2 (dotted line). The density of the propagation medium is 1 atom/cm^3. Experimental points (not corrected for solar modulation) are from: SAC-DSRI, 1971 (Ref. 36), CHIC, 1972 (Ref. 37), UNH, 1972 (Ref. 38), GSFC, 1974 (Ref. 40), WU, 1975 (Ref. 42), and DSRI, 1975 (Ref. 43).

tion (i.e., the isotopic composition of our iron targets), we find that the experimental abundances of Sc, V, Cr, Mn are reasonably accounted for by propagation through a slab of ~2 g/cm^2 of hydrogen, while it has been shown by several authors (e.g., Ref. 27) that the formation of the light elements Li, Be, B requires about 4 g/cm^2. A Gaussian distribution of path length centered about 2 g/cm^2 gives similar results. The calculated abundances are less sensitive to the mean value (the escape length) of an exponential distribution of potential path length, at least above a certain value, due to the small interaction mean free path of iron.

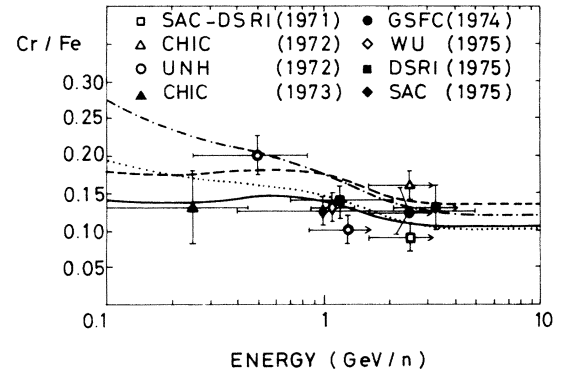

FIG. 2. Same as Fig. 1, for Cr. CHIC, 1973 (Ref. 39); SAC, 1975 (Ref. 41). For Ref. 39, a value of 0.1 has been assumed for Mn/Fe in deriving Fe abundance from the measured Mn+Fe abundance.

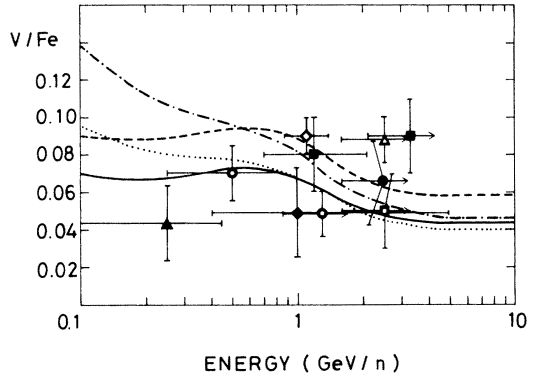

FIG. 3. Same as Fig. 2, for V.

Indeed, experimental abundances are consistent with an exponential distribution having an escape length between 4 and **10** g/cm^2.

Thus we also come to the conclusion that (1) an exponential distribution of path length with an escape length of ~5 g/cm^2 of hydrogen (i.e., ~6 g/cm^2 of 90% hydrogen+10% helium) which explains the formation of the L group[27] is also consistent with the measured abundances of the Sc-Mn group (very recently Fontes[44] on the basis of his latest experimental cross sections found that 5.5 g/cm^2 of H gives the best agreement with the measured abundances of the L elements at 1 GeV/n); (2) the source abundances of all elements Sc to Mn are zero, as for the L group, or at least close to zero, since the experimental uncertainties do not allow us to exclude small source abundances for these elements, of the order of their solar system abundances. Although several Sc points clearly lie above our calculated curves, it seems to us premature to assume a nonzero source abundance, which would be quite strange, for this very rare element.

As expected, these results are in agreement with previous findings, but we believe it is an im-

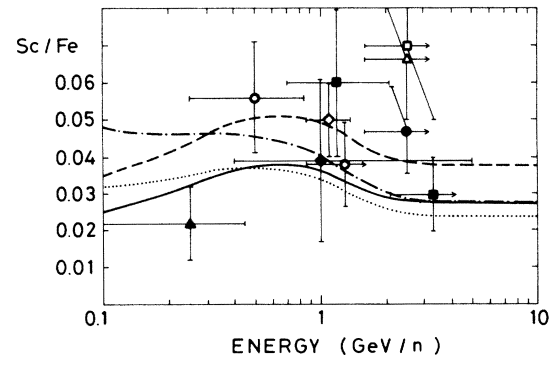

FIG. 4. Same as Fig. 2, for Sc.

portant improvement to have these conclusions based now on experimental nuclear cross sections. One may ask whether these results are biased by the adopted isotopic composition of Fe, as some indications of a high ^{54}Fe isotopic abundance have been reported.[45] Calculations done with a pure ^{54}Fe source and using Silberberg and Tsao's cross sections for the spallation of ^{54}Fe give abundances less than 20% lower than in the case of a "normal" iron source except for Mn whose abundance relative to iron at 1 GeV/n drops from 9.4% to 5.1% since only ^{53}Mn is formed in this case. We shall come back to this point later.

Several groups have reported[30,31] variations of the Fe secondaries to Fe ratio as a function of energy above a few GeV/n, which they interpret in terms of a decrease of the escape length with increasing energy. Using a constant escape length of 5 g/cm^2, we find that due to cross-section variations, the ratio of Sc-Mn to Fe decreases by some 20–25% between ~800 MeV/n and a few GeV/n (from ~0.5 to ~0.4). At higher energies, this ratio remains constant. A comparison with experimental data is difficult, since most authors give relative abundances of groups including elements like Ca or S which are partly primary. We note however that this decrease due to cross section is insufficient to explain the variations of the Sc-Mn/Fe ratio reported by Benegas *et al.*[42] between 0.5 and 5 GeV/n or deduced by Juliusson *et al.*[46] from the measurements of Juliusson[31] above 27 GeV/n. A study of this problem is presently in progress.

We now proceed to the results connected with electron capture nuclides. Several of the nuclides we have studied are radioactive only by electron capture: ^{49}V, ^{51}Cr, ^{53}Mn, ^{54}Mn, to which one should add ^{55}Fe. When completely stripped of their orbital electrons, as high energy cosmic rays normally are, these nuclei are stable. Their decay becomes possible only if they first pick up an interstellar electron on an atomic level. The potential use of these nuclides to probe some propagation conditions has been discussed by Yiou and Raisbeck,[47] Reames,[48] and Raisbeck *et al.*[49] The knowledge of the relevant nuclear cross sections allowed us to make a more quantitative approach by means of the PCRNOR code which, as mentioned above, allows electron capture nuclei to pick up an electron from the propagation medium and then either lose this electron or decay. The cross sections for pickup and loss of electrons have been calculated in the way indicated by Raisbeck and Yiou,[50] who found a reasonable agreement with their experimental results on charge exchange of protons. There is, however, at present no experimental confirmation of the validity of this treatment in the charge and energy ranges relevant to the present study, and this may be the main uncertainty of our results.

For each electron capture nucleus which has picked up an electron, there is a range of densities of the propagation medium within which radioactive decay and loss of the electron can compete and the abundance of this species is therefore dependent on the density.[47-49] For all isotopes studied here, except ^{53}Mn which has a much longer half-life, this happens above a few times 10^2 atoms/cm^3. These nuclides could thus serve as probes of propagation through such a dense medium.[51] For a less dense medium, like the interstellar gas, decay occurs essentially each time an orbital electron has been picked up, so that the abundance does not depend any more on the density.[47,49] It still reflects, however, the strong energy dependence of the electron pickup cross section and Raisbeck *et al.*[49,52] have proposed to take advantage of this to study solar modulation. As an illustration of this potentiality, Figs. 5 and 6 show the calculated interstellar isotopic composition of vanadium and chromium for different values of the energy. The calculations have been done using an exponential distribution of path length and a leakage length of 5 g/cm^2, but the results are only slightly model dependent. They essentially depend on electron pickup cross sections and nuclear cross sections or rather, nuclear cross-section ratios, which are precisely the quantities we measured.

There is a drastic change in isotopic composi-

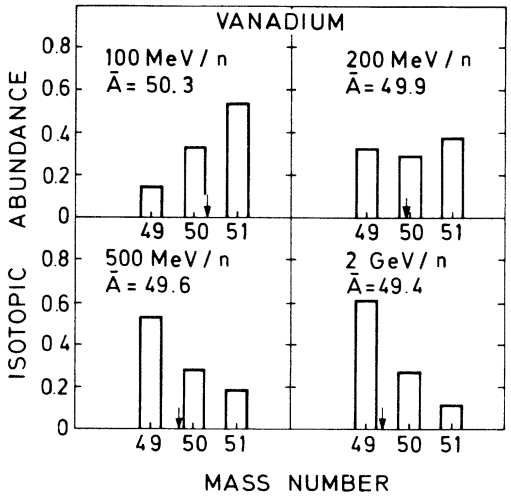

FIG. 5. Interstellar cosmic ray isotopic composition of V at four energies, calculated with a 5 g/cm^2 exponential distribution of path length and a density $\rho < 10^2$ atoms/cm^3. The arrows indicate the mean mass $\bar{A}$. The elemental abundance is chosen equal to 1 for normalization.

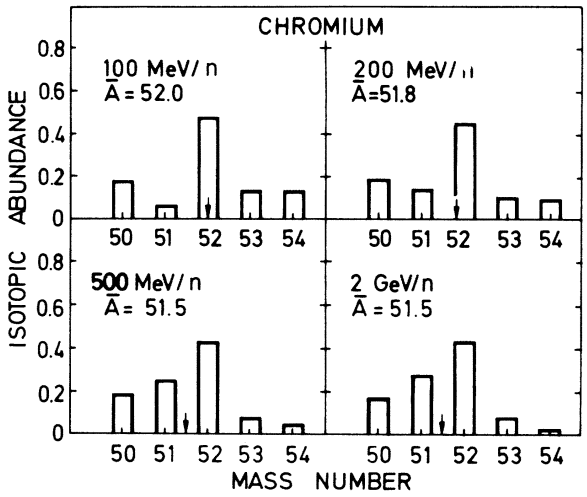

FIG. 6. Same as Fig. 5, for Cr.

tion between 100 MeV/n and ~1 GeV/n, above which energy ^{49}V and ^{51}Cr behave as stable nuclides. However, this change has almost no effect on elemental abundances, since the decay of an isotope is compensated by the growth of another one (the cross sections involved being roughly equal). Hence, one needs a measurement of isotopic abundances. As suggested by Raisbeck *et al.*[49,52] a comparison between calculated and measured isotopic compositions would yield the mean energy loss suffered by cosmic ray particles when penetrating the solar system and its time dependence within a solar cycle. These measurements are by no means easy ones, but several satellite experiments are planned in the near future, which could achieve them in the proper energy range. In the same way, such measurements made in an out-of-ecliptic mission should allow a test of the dependence of adiabatic energy loss on the spatial coordinates. In the case of vanadium, a precise determination of the mean mass could already be of interest since its interstellar value is expected to vary by almost 1 amu between 100 MeV/n and 2 GeV/n, as can be seen in Fig. 5.

We know of two other suggestions to determine the energy loss due to solar modulation. Waddington[53] and Rao, Young, and Fukui[54] argued that large nuclear cross-section variations below ~1 GeV/n should be reflected in the interstellar abundances of Mn and Cr, so that the measured abundances should in turn vary between solar maximum and solar minimum, depending on the amount of interplanetary energy loss. It is clear, from the curves in Figs. 1 and 2 that this is only true for the slab model and that no appreciable change in Mn and Cr abundances is to be expected below ~1 GeV/n, if one believes in an exponential distribution of path length. The other possibility, worked out by Meyer,[55] similarly relies on variations of nuclear cross sections for the production of ^{2}H, and is supported by existing measurements of ^{2}H/^{4}He. This method implies some assumptions on the shape of the interstellar energy spectrum, and gives rise to some difficulties with existing modulation theories. In any case, it would be interesting to compare indications obtained from both the ^{2}H/^{4}He ratio and the isotopic composition of iron secondaries.

Manganese has two electron capture isotopes: ^{53}Mn has a long half-life (3.7×10^6 yr)[22] and its decay will only be possible at low densities; ^{54}Mn has a "short" half-life (312 day[13]) and should normally behave like ^{49}V or ^{51}Cr. Its case is, however, more complicated since β decay, although never experimentally identified, is energetically possible. Cassé[56] has discussed this problem and calculated a partial half-life for β decay of 2×10^6yr with a factor of 2 uncertainty. We have used this latter value in our calculations. The results for manganese, obtained with a 5 g/cm^2 exponential distribution of path length, are illustrated, as previously, by isotopic distributions shown in Figs. 7–9 for several densities of the propagation medium. At 1 atom/cm^3 (i.e., about the mean density in the galactic disk) one notes the same decay feature of ^{54}Mn at low energies as for ^{49}V and ^{51}Cr, accompanied by an increase of ^{55}Mn due to decay of ^{55}Fe. Only a small proportion decays by β emission (~15% at 1 GeV/n), while at 10^{-2} atom/cm^3, decay is almost complete. This is easily under-

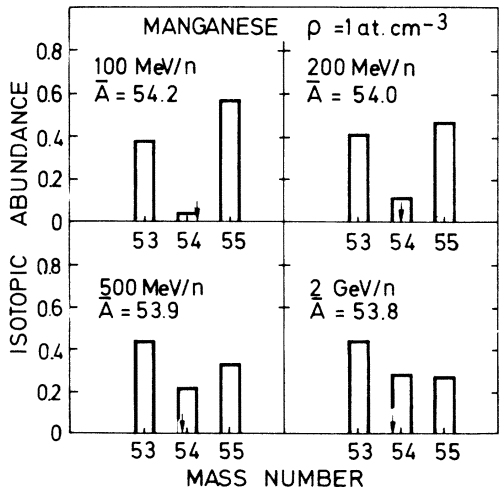

FIG. 7. Interstellar cosmic ray isotopic composition of Mn at four energies, calculated with a 5 g/cm^2 exponential distribution of path length, and a density $\rho = 1$ atom/cm^3.

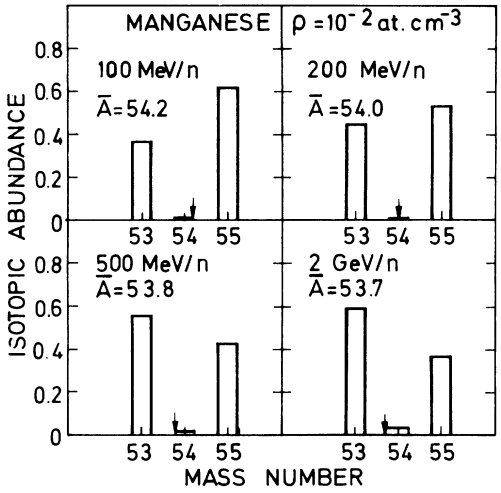

FIG. 8. Same as Fig. 7, for $\rho = 10^{-2}$ atom/cm^3.

standable since, for a given path length to be traversed, the lower the density, the longer the confinement time. ^{54}Mn is thus well suited to probe densities in the range between 1 and 10^{-2} atom/cm^3, within which lies the density that can be inferred from recent ^{10}Be measurements (see for instance the review of Meyer[57] and references therein). It must be emphasized that this obviously relies upon the β half-life calculated by Casse,[56] an experimental verification of which would be highly desirable.

At densities higher than about 10^{-2} atom/cm^3, ^{53}Mn does not decay at all since, when it has picked up an electron, the probability of losing it is much larger than the probability of radioactive decay. Below ~10^{-2} atom/cm^3, decay becomes noticeable at low energies, so that the role of ^{53}Mn could be that of fixing a lower value for the density of the propagation medium.

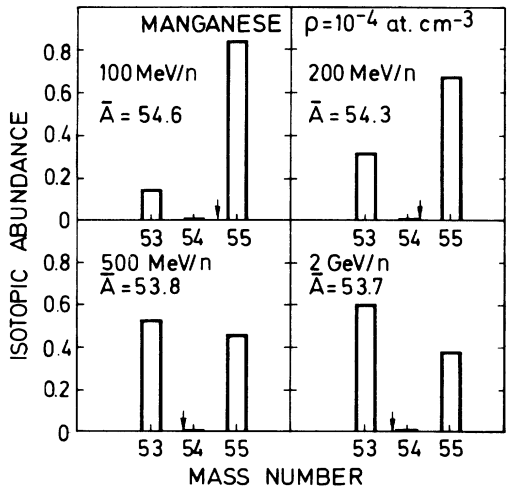

FIG. 9. Same as Fig. 7, for $\rho = 10^{-4}$ atom/cm^3.

What is the influence of these isotopic variations on the elemental abundance of Mn? We find that complete decay of ^{54}Mn makes the Mn/Fe ratio at 1 GeV/n drop from 9.4% to 6.7%, but we have already noted that such a low Mn abundance can also be obtained by assuming a high abundance of ^{54}Fe. Therefore we believe that, also in this case, isotopic measurements are necessary. Only a high Mn abundance would be an indication that ^{54}Mn has not decayed and that ^{54}Fe has a low abundance, although even this case is not unambiguous, since one can still imagine a nonzero source abundance of ^{55}Mn. As present measurements of Mn/Fe range from 6% to 12%, it is difficult to draw a conclusion. In any case, it should be realized that, in order to test for eventual decay of ^{54}Mn by β emission, measurements must be made at a sufficiently high energy, corresponding to an interstellar energy larger than about 500 MeV/n, where decay by electron capture is no longer important.

V. CONCLUSION

We have presented here the first experimental cross sections for production of stable isotopes of Be, Sc, V, Cr, and Mn by high energy spallation of Fe, along with some measurements on radioactive isotopes. These cross sections have allowed us to confirm and complement some previous ideas on cosmic ray propagation, and to quantitatively illustrate the possibilities offered by electron capture nuclides.

The results of our calculations are still subject to some uncertainties and some measurements are clearly needed. Our measurements should be extended to lower energies, to make more precise the excitation functions we have used, and electron pickup cross sections should be measured for heavy ions in the energy range from 0.1 to 1 GeV/n. Such experimental data will probably be available in the near future. Of great importance, but extremely difficult, would be a determination of the β branching ratio of ^{54}Mn.

The calculated isotope distributions we have presented may thus have to be adjusted for new experimental data. We do not expect their main characteristics to be deeply altered, and we believe that the potential information they contain is a strong motivation, among others, for cosmic ray isotope measurements.

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