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Cross sections for the spallation production of ^{10}Be in targets of N, Mg, and Si and their astrophysical applications

G. M. Raisbeck and F. Yiou
Laboratoire René Bernas du Centre de Spectrométrie Nucléaire et de Spectrométrie de Masse, 91406 Orsay, France
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Using an isotope-separator technique, cross sections have been measured for the spallation production of ^{10}Be in targets of N, Si, and Mg by protons of 600 MeV and in N by protons of 150 MeV. For 600-MeV protons $\sigma(^{10}\text{Be}) = 1.3 \pm 0.6$, 0.7 ± 0.3, and 1.3 ± 0.4 mb in N, Si, and Mg respectively. For 150-MeV protons $\sigma(^{10}\text{Be}) = 0.6 \pm 0.2$ mb in N. The results are discussed in terms of the cosmogenic production of ^{10}Be in meteorites, lunar surface, cosmic dust, the earth's atmosphere, and the interstellar gas. It is shown that the Si and Mg components of chondritic material will be important sources of ^{10}Be, at least as far as the primary galactic flux is concerned. It is also pointed out that the quantitative evaluation of the ^{10}Be from lighter nuclei such as oxygen in meteorites and nitrogen in the earth's atmosphere depends strongly on the magnitude of the (unmeasured) secondary-neutron-induced reactions. ^{10}Be is shown to be a small fraction of the total Be formed in the spallation of cosmic rays over the whole range of important primary nuclei. Using standard radiochemical techniques cross sections have also been measured for ^{7}Be and ^{22}Na production in targets of Si and Mg irradiated by 600-MeV protons. The results differ significantly from previously published values.

[NUCLEAR REACTIONS $^{14}\text{N}(p, x)^{10}\text{Be}$, $E = 150$, 600 MeV, ^{24}Mg, $^{28}\text{Si}(p, x)^{10}\text{Be}$, ^{7}Be, ^{22}Na, $E = 600$ MeV, measured σ's. Discuss implications for cosmogenic production of ^{10}Be.]

INTRODUCTION

The flux of high-energy particles that comprises the nuclear component of galactic cosmic rays is constantly undergoing nuclear reactions with a variety of astrophysical and geophysical targets. Such interactions take place in meteorites, lunar soil, the earth's atmosphere, cosmic dust, and the interstellar gas itself. A record of these nuclear transformations, and often the most easily identifiable one, is preserved by the long lived radionuclides which are formed in such reactions. An important example of such a cosmogenic product is the nuclide ^{10}Be [half-life $= (1.5 \pm 0.3) \times 10^6$ yr],[1] having important applications in such diverse areas as cosmic ray physics, geophysics, and cosmochronology.

^{10}Be is the only long-lived isotope that can be formed from the two most abundant components in the earth's atmosphere, oxygen and nitrogen. After it is swept out of the atmosphere by precipitation, the ^{10}Be is then distributed in geophysical sinks such as marine sediments or polar ice.[2] If these deposits are left undisturbed the ^{10}Be activity can provide an unbroken record of geophysical activity for periods up to several millions of years. In a complementary fashion, if an alternate technique can be found to date such deposits, the ^{10}Be activity could be used to probe the constancy of the cosmic-ray flux over similar periods (and perhaps even relatively short-term fluctuations such as might be caused by a local supernova, etc.).

In meteorite studies ^{10}Be is again an important product, being, along with ^{26}Al, the only long-lived activity to be formed in oxygen, silicon, and magnesium, the three most abundant elements in chondritic material. Because it is generally accepted to be produced by high-energy particles ^{10}Be has sometimes been used to try to distinguish between galactic and solar components of cosmic rays, or to investigate depth effects.[3,4] Also, because the relative rates of production of ^{10}Be and ^{26}Al are expected to be much different in the earth's atmosphere than in material of chondritic-like composition, the ratio of these two species in terrestrial reservoirs has been suggested as a way of estimating the influx of extraterrestrial cosmic dust.[5]

In cosmic-ray studies themselves ^{10}Be has perhaps been the object of the most intense interest of any single nuclide ever since it was realized that it might offer the possibility of acting as a "horloge," thus providing temporal (and by implication spacial) information regarding propagation conditions.[6,7]

In order to fully utilize the possibilities offered by studies such as those described above, it is of course necessary to know the production cross sections for the relevant nuclear reactions. Un-

fortunately such cross sections, to date, are very sparse. On one hand, the very property which makes ^{10}Be interesting as a dating tool, i.e., its long half-life, also makes its determination by standard radiochemical techniques extremely laborious. In addition, the interference caused by the simultaneous production of ^{7}Be (half-life = 53.2 days) in any nuclear irradiation has generally meant that such experiments required a cooling time of several years before the ^{10}Be could be measured.[8,9] Recently we have developed a technique for ^{10}Be determinations which avoids (or at least minimizes) such difficulties.[10] In this paper we report the results of applying this technique to the high-energy proton fragmentation of three target nuclei; nitrogen, magnesium, and silicon, which are important in the context of the problems mentioned above.

EXPERIMENTAL

Irradiations were carried out using the internal beams of the CERN (600-MeV) and Orsay (150-MeV) synchrocyclotrons. Although our technique is very clean, it has a relatively low over-all efficiency (~1%), and thus long bombardments were required. Each of the targets was irradiated 30–40 h, giving a total integrated flux (including multiple traversals) of $\sim 5 \times 10^{18}$ protons. The Mg target was made up of 10 pieces of 99.9% pure metal foil, each 0.5 mm thick and 4 cm^2 in area. The Si was irradiated in the form of ~500 mg of 99.99% pure powder wrapped in thin gold foil. Each of the "nitrogen" targets consisted of ~5 g of high purity vanadium nitride powder that had been compressed into disks 20 mm in diameter and 0.5 mm in thickness. Corrections due to the contribution of the V in VN are estimated to be small, as discussed in Appendix II. The thickness of the targets was such that the maximum energy loss was only ~6 MeV at 150 MeV, and less than 2 MeV at 600 MeV.

Some additional details of our isotope separator technique, which has been outlined previously,[10] are given below. After several months of cooling time, each of the targets was dissolved up with ~2 mg of ^{9}Be carrier and subjected to a relatively simple chemical treatment. The main purpose of this treatment was to remove the large quantity of target material and provide a chemically pure sample of BeO for insertion into the ion source of the separator. Because of the high dispersion factor of the isotope separator, separation of the ^{7}Be and ^{10}Be (as $BeCl^+$) was no problem (with one minor exception, discussed below). Our main objectives were therefore to avoid, at the mass-42 and -45 collection positions, (a) any stable species (such as ^{45}Sc) which could cause sputtering of the collected products; (b) any radioactive atoms (such as ^{45}Ca) or even highly unusual molecules (for example ^{22}Na as Na_2^+ = mass 45) which are always possible in a discharge ion source. In addition, of course, it was desirable to reduce, as much as possible, all of the radioactive species since there is always some scattering due to residual gas pressure in the separator.

In order to minimize problem (a), an ion source was constructed of previously unused components, and care was taken to avoid materials which could cause interferences (example: boron nitride insulators which can give $^{10}B^{35}Cl$ = mass 45). Blank runs were made until the "sputtering" currents at the collection positions were established to be negligible. With regard to problem (b), after separation from the bulk of the target material the Be was put through a purification procedure consisting of repeated Be $(OH)_2$ precipitations (in the presence of EDTA) and Fe $(OH)_3$ scavenges in a strong NaOH medium.

The final Be $(OH)_2$ precipitate was converted to the oxide and loaded into a graphite capsule suitable for insertion into a "furnace" at the entrance to the ion source of the isotope separator SIDONIE.[11] A stream of CCl_4 gas was passed over the sample, which was heated to ~500 °C, and the Be collected as $BeCl^+$ ions on thin Al foils. The mass-44 ($^{9}Be^{35}Cl^+$) current, measured through a 25 × 1-mm slit in the focus plane, was used to monitor the run and maintain the focus conditions during the separations, which lasted 15–45 min. When the decrease in the mass-44 current indicated that the separation was completed, the collector section was isolated from the ion source and the Al foils removed. The foil at mass 42 ($^{7}Be^{35}Cl^+$) was folded into a geometry suitable for direct γ counting of the ^{7}Be in a NaI well crystal. The foil containing the ^{10}Be was sectioned with a razor blade into three strips 36 × 3 mm (the focus conditions of the separator being such that essentially all of the activity is localized in a strip 25 × 1 mm). Each of these strips was then cut into three pieces, so that for ^{10}Be counting purposes we had an effective rectangle of 9 × 12 mm. These samples were counted in a previously calibrated[10] low-level β counter having a background of 0.38 cpm, and an efficiency for ^{10}Be of 0.30 ± 0.02. The two 3-mm strips on either side of the mass-45 ($^{10}Be^{35}Cl^+$) position were used as background determinations, and always showed negligible activities (< 5% of the mass 45).

As was noted earlier, there was one minor aspect in which separation of ^{7}Be and ^{10}Be was not as complete as was expected, on the basis of

TABLE I. Cross sections for the production of ^{10}Be in proton-irradiated targets of Mg, Si, and N.

Target	Proton energy (MeV)	^{10}Be activity (cpm)	^{10}Be activity (dpm)	^{7}Be activity (μc) at $t=0$	$\sigma(^{10}\text{Be})/\sigma(^{7}\text{Be})$	$\sigma(^{7}\text{Be})$ [a] (mb)	$\sigma(^{10}\text{Be})$ (mb)
Mg	600	0.19±0.03	0.65±0.10	15.5 ±0.8	0.19±0.04	6.5±1.5	1.3±0.4
Si	600	0.05±0.01	0.17±0.04	5.36±0.27	0.13±0.05	5.3±1.2	0.7±0.3
VN	600	0.07±0.01	0.23±0.04	4.98±0.25	0.20±0.05		
(N) [b]	600				0.18±0.06	7.0±2.0	1.3±0.6
VN	150	0.12±0.02	0.40±0.07	18.0 ±0.9	0.11±0.03		
(N) [b]	150				0.10±0.03	6.0±1.5	0.6±0.2

[a] See Appendix I.

[b] Results for N have been calculated after correction for contribution of V, as described in Appendix II.

the known characteristics of SIDONIE. We have measured the tailing and scattering of ^{7}Be as a function of position in the collector and shown that, at mass-45 (^{10}Be ^{35}Cl) position, the expected contribution is $\sim 10^{-4}$ of the collected ^{7}Be activity. In making γ measurements on the ^{10}Be samples, however, we found that the actual contribution was $\sim 5\times 10^{-4}$ and that this was concentrated exactly at the mass-45 position and not spread out as would be expected from the two effects mentioned above. We believe (without any direct proof) that this activity may represent ^{7}Be $^{19}F_2^+$ (=mass 45) arising from fluorine impurity in the CCl_4. Although we used research grade CCl_4, it is easy to show that a fluorine impurity of ~0.01% would account for the effect we observe. The net result of this effect was to give a slight (<20%) contribution to the ^{10}Be counting rate which decayed with a ~53 day half-life (although ^{7}Be decays by pure electron capture, the 10% branching ratio giving γ rays can produce Compton electrons and we have measured the over-all efficiency of the β counter to ^{7}Be to be ~0.01%.) In order to make sure that this contribution to the ^{10}Be was negligible we have counted the samples at intervals of several months after the separation. As in earlier work[1,10] the count rates reported here are those obtained after all traces of ^{7}Be had decayed. In the future we believe such a problem might be avoided by repurifying the CCl_4 before using.

From the relative activities of the ^{7}Be and ^{10}Be samples the cross section ratios $\sigma(^{10}\text{Be})/\sigma(^{7}\text{Be})$ are directly determined. In order to have the absolute ^{10}Be cross sections it is necessary to have the values for σ (^{7}Be). Although there are experimental values in the literature for all the targets involved in this work, we have independently measured the ^{7}Be cross sections for Mg and Si. The details of these measurements are given in Appendix I.

RESULTS

In Table I we give a summary of our results, including the measured ^{10}Be and ^{7}Be activities after isotope separation, the ^{10}Be/^{7}Be production ratio, the adopted ^{7}Be cross section, and the ^{10}Be cross sections. The errors on the ^{10}Be and ^{7}Be activities are statistical counting errors (one standard deviation), plus the uncertainty in the counter efficiencies. The ^{10}Be/^{7}Be ratio includes these uncertainties plus the uncertainties in the relative separator collection efficiencies (equal to within 5%),[10] and the half-lives of the two species. For ^{7}Be we have used a half-life of (53.17±0.07) days,[12] and a branching ratio of 10.3%.[13] For ^{10}Be we have used our measured value of $(1.5\pm 0.3)\times 10^{6}$ yr for the half-life.[1] Support for the essential accuracy of this latter determination includes recent corrections to both previously published values[14,15] and an independent measurement.[15] The error in the ^{10}Be cross sections in Table I include also the uncertainty in the adopted ^{7}Be cross section. Of course any improvement in either the ^{10}Be half-life and/or the ^{7}Be cross sections will bring an automatic improvement in the ^{10}Be cross sections.

In the case of the results quoted in Table I for "nitrogen" there is a correction necessary to take account of the contribution of the vanadium in the VN target. In Appendix II, we show that the uncertainty introduced by this correction is quite small.

DISCUSSION

The results presented here for ^{10}Be production in N, Mg, and Si, together with previous measurements in targets of C and O and our new determination for the ^{10}Be half-life, constitute a set of data permitting for the first time a more precise discussion of the source processes responsible for ^{10}Be in its role as an astrophysical

and geophysical "chronometer." In particular, one is in a better position to examine the relative importance of various target elements in each individual situation. One interesting aspect of such a procedure is that it also gives some insight into the controversy and confusion regarding some earlier measurements, particularly the apparent discrepancy between mass-spectrometrically measured cross sections and a variety of direct and indirect evidence which has been cited to cast doubt on those measurements. For example, the very small production cross sections measured for ^{10}Be formed in proton irradiated O targets have been challenged on basically three types of evidence: (a) their "disagreement" with similar radiochemical measurements[9]; (b) their "inconsistency" with production rates measured in meteorites[16] and lunar soil[4]; (c) their "discrepancy" with production rates in the earth's atmosphere, as implied from measurements of marine sediments.[17] We hope that it will become apparent in the discussion that follows that such discrepancies have arisen in large measure not from experimental contradictions but from incomplete or inaccurate analysis of the relevant data.

Finally, it will also be clear from the discussion that our measurements are a long way from answering all the questions, and that a considerable amount of work still remains to be carried out before one is in a position to utilize fully the information provided by this interesting and important nuclide. Our most important contribution is, perhaps, to emphasize the necessity for applying a generous amount of caution in this area before drawing any firm and far-reaching conclusions.

A. Comparison with other results

There are, effectively, no previous results which can be directly compared to the present work. Nevertheless, a brief chronological summary of earlier ^{10}Be measurements may be useful in understanding the context in which the present experiments were undertaken, and to give a background for the discussion which follows in later sections.

The earliest ^{10}Be cross-section measurements were the radiochemical results of Honda and Lal[8] for 220-MeV protons on carbon and semicarbazide (CN_3O) targets, and 730-MeV protons on Fe. The cross-section ratios for ^{10}Be/^{7}Be from these targets were given as 0.21 ± 0.06, 0.4 ± 0.2, and 0.40 ± 0.10 respectively (where possible we prefer to quote the ^{10}Be/^{7}Be ratio rather than the absolute ^{10}Be cross sections since the reference value for the adopted ^{7}Be cross section has not always been the same and this introduces an additional variable into quoted absolute cross sections). From that work it was argued that the ^{10}Be/^{7}Be ratio in N and O targets was probably somewhat greater than that measured in C. These results were the basis for essentially all discussion of ^{10}Be spallation production until 1968 when mass-spectrometric measurements for the production of ^{10}Be from targets of O irradiated by protons of 150, 600 and 19 000 MeV were reported.[18] For a similar energy these latter results give a ^{10}Be/^{7}Be value almost an order of magnitude lower than had been estimated from the radiochemical results quoted above. In 1971 mass spectrometric cross sections for C targets irradiated by protons of 150 and 600 MeV were reported[19] and, while considerably larger than the oxygen results, were also somewhat lower than the radiochemical values of Honda and Lal.[8] Most recently, Amin *et al.*[9] using the same oxygen targets used for the mass-spectrometric measurements, reported radiochemical cross sections which were smaller than estimated from the earlier radiochemical work, but still a factor of 3 to 4 higher than the mass-spectrometric measurements. Meanwhile, the half-life for ^{10}Be had been remeasured[1] and was found to be $(1.5 \pm 0.3) \times 10^{6}$ yr instead of $(2.7 \pm 0.4) \times 10^{6}$ yr. Such a half-life modification, of course, reduces all radiochemical ^{10}Be cross-section measurements by the same factor. Accepting, for the moment, the validity of this new half-life, it is interesting to reexamine the situation as it now exists. A summary to this effect is shown in Table II.

The semicarbazide (CN_3O) target of Honda and Lal[8] will now give ^{10}Be/^{7}Be $= 0.24 \pm 0.13$, a value still somewhat in excess of that measured here (interpolated to 220 MeV) for a nitrogen target. Making corrections to the semicarbazide target for the contributions from the carbon and oxygen atoms makes this discrepancy even greater. However, in view of the large statistical error limits given for that work, we do not feel that this disagreement can be considered significant.

The radiochemical result for the carbon target at 220 MeV when adjusted for the new half-life is, contrary to the argument of Amin *et al.*,[9] in good agreement with the mass-spectrometric value interpolated for this energy (those authors have apparently compared that result to the 600-MeV mass-spectrometric value, which does not seem reasonable to us).

Finally, the most recent radiochemical results for oxygen[9] should now also be adjusted for the smaller ^{10}Be half-life. Although that work was reported after the new half-life had been communicated to those authors, they preferred to

TABLE II. Comparison of ^{10}Be cross-section measurements in light targets by different techniques.

Target	Proton energy	Technique	$\sigma(^{10}Be)/\sigma(^{7}Be)$ Published value	Present estimate [a]	References
C	220	Radiochemistry	0.21 ± 0.06	0.12 ± 0.04	Honda and Lal (8)
C	150	Mass spectrometry	0.09 ± 0.01		Fontes *et al.* (19)
C	600	Mass spectrometry	0.25 ± 0.02		Fontes *et al.* (19)
C	(220)	Mass spectrometry		(0.14 ± 0.03)	Fontes *et al.* (19)
CN_3O	220	Radiochemistry	0.4 ± 0.2	0.24 ± 0.13	Honda and Lal (8)
N [b]	220	Radiochemistry		0.38 ± 0.20	
N [c]	150	Isotope separator	0.10 ± 0.03		This work
N [c]	600	Isotope separator	0.18 ± 0.06		This work
N [c]	(220)	Isotope separator		(0.12 ± 0.04)	
O	135	Radiochemistry	0.19 ± 0.01	0.11 ± 0.02	Amin *et al.* (9)
O	600	Radiochemistry	0.32 ± 0.02	0.18 ± 0.04	Amin *et al.* (9)
O	135	Mass spectrometry	0.07 ± 0.02		Yiou (18)
O	600	Mass spectrometry	0.09 ± 0.03		Yiou (18)

[a] Values in this column have been calculated on the basis of a ^{10}Be half-life = $(1.5 \pm 0.3) \times 10^6$ yr (with the uncertainty in this half-life being included in the listed errors). Values in brackets have been interpolated to an energy of 220 MeV on the basis of measurements at 150 and 600 MeV, with a somewhat increased error included to account for the uncertainty in this procedure.

[b] Value of N has been calculated by making corrections for C and O contributions (using mass spectrometric results for these contributions).

[c] Value for N has been corrected for contribution of V, as indicated in Appendix II.

adopt the older value in presenting their results. As we have noted earlier, recent reports[14,15] tend to remove most of the reasons for doubting this smaller half-life measurement. The adjusted radiochemical results for oxygen targets will thus now be in quite acceptable agreement with the mass spectrometer results at 150 MeV, and barely a factor of 2 larger at 600 MeV. Taking into account the remaining uncertainties in both measurements, this is probably not too unreasonable. In this regard it should be pointed out that the value given in Table II for the uncertainty in the mass-spectrometric measurement at 600 MeV is taken from the original work[18] and differs significantly from that which has been incorrectly quoted in Ref. 9.

The only other published ^{10}Be cross sections which are relevant to the present discussion are those measured in C and N using an emulsion technique.[20] However, in view of the very limited statistics and the inability of that technique to reproduce even the well-established ^{7}Be cross section, we feel justified in discounting that work.

We thus come to the conclusion that there are probably no significant discrepancies among any of the direct measurements of ^{10}Be cross sections. Objections which have been raised on the basis of indirect evidence are considered in later sections.

B. ^{10}Be production in meteorites and lunar material

In this section we consider the ^{10}Be production in extraterrestrial bodies such as stone meteorites and lunar crust. The cosmogenic production of ^{10}Be has previously been assumed to arise primarily from the oxygen component of such materials[3,4,16] (the reason being that this is the element having the smallest mass difference between the target and product nuclei). We have previously expressed the necessity for considerable caution about such an assumption,[21] especially in view of the small cross sections measured for the production of ^{10}Be by protons in oxygen targets. In fact the lack of agreement between the ^{10}Be found in meteorites and lunar material compared to that calculated using such small oxygen cross sections has been cited by others as one of the arguments against the reliability of those cross section measurements.[4,16] We have suggested[21] two possible explanations for this apparent anomaly: (a) Oxygen is indeed the most important target element but it is secondary particles, notably high energy neutrons, which are responsible for the majority of the ^{10}Be production. (b) Other heavier but abundant target nuclei, such as Mg, Si, and Fe, may contribute appreciably to the ^{10}Be yield. It was the possibility of exploring this

second hypothesis which was one of our motivations for undertaking the present experiments with Si and Mg targets. Indeed, with 600-MeV protons the ^{10}Be cross sections in Si and Mg are already as large or larger than σ (^{10}Be) measured for oxygen at the same energy.[18] For higher energies it is expected that the ^{7}Be cross section in Mg and Si will rise to ~10 mb.[22] Assuming that the ^{10}Be/^{7}Be ratio remains constant (by analogy with results for C and O we feel that in fact it is likely to increase slightly, although this is certainly an area that must be studied), the cross section for ^{10}Be in Si and Mg at higher energies will thus be 1.5 to 2 times larger than in oxygen. Since the combined quantity of Si and Mg in chondritic material is comparable to oxygen, it is evident that, considering only the primary cosmic-ray protons, Si and Mg would in fact be more important than oxygen in producing ^{10}Be. Of course the effective threshold of the oxygen reaction is lower, so secondary particle contributions will be important and the question becomes more complicated. As we have pointed out elsewhere,[1] there are valid nuclear physics reasons for believing that the neutron-induced ^{10}Be cross sections for a light target such as oxygen will be significantly larger than the corresponding proton-induced cross sections. An interesting experiment would be to try to identify the most important progenitor nuclides by measuring ^{10}Be activities in different meteorites (or different mineral components of the same meteorite) having different chemical compositions, and then applying a least-squares fitting technique as has been done for ^{26}Al.[23,24]

In any event it seems obvious that the production of ^{10}Be by the irradiation of a thick multicomponent target (i.e., meteorite or lunar rock) by a spectrum of various projectiles (i.e., the primary cosmic rays plus all their secondaries) is a complex process and cannot, in general, be reliably identified with a simple nuclear reaction such as monoenergetic protons on oxygen.

C. ^{10}Be production in cosmic dust

The source, extent, and composition of the dust particles which pervade interplanetary space are still largely open questions. It has been noted that, if the composition of these particles is similar to chondritic material, the relative cosmogenic production of ^{10}Be/^{26}Al will be significantly different than in the earth's atmosphere, and that measurement of this ratio in geophysical reservoirs may permit a determination of the infall of this extraterrestrial dust on the earth.[5] We note here several interesting aspects about nuclear production processes in such dust particles.

Since these particles are small (0.1–1000 μm), there will be little effect of buildup of secondary particles such as is the case in large solid bodies (the implicit assumption here being that the cosmic-ray irradiation has taken place when the dust was in its divided form and not during an earlier period when it may have been part of a larger body). Thus, in this case cosmogenic products such as ^{10}Be will indeed reflect the production processes from the primary galactic (or solar) proton flux. Therefore, assuming a composition, the yield of these products can in principle be calculated directly from experimental proton cross sections. There is, however, one complicating feature which must be taken into account in such a procedure. Since the dust particles are small it is necessary to consider possible losses due to the recoil effect in the nuclear reaction. Lal and Venkatavaradan[25] have estimated the loss of ^{10}Be from a 1-μm particle as ~30%. They do not give a detailed quantitative analysis for this estimation, but it appears to be based on experimental data for spallation type products (i.e., the residual nucleus after emission of one or more individual nucleons). Perhaps more relevant to ^{10}Be, however, is to consider the case of a typical fragmentation product like ^{8}Li. Experimental evidence indicates that, even in a target as light as C, and certainly in a target such as Al, a large fraction of the ^{8}Li product nuclei have an energy $\geq$5 MeV.[26] But, the range (in chondritic material) of a 5-MeV ^{10}Be is ~8 μm.[27] We arrive at the conclusion, therefore, that recoil loss of ^{10}Be will probably be very severe in the smallest cosmic dust particles (approaching 100% for a particle of 1 μm). Fortunately, any such effect would be in a favorable direction as far as trying to distinguish between cosmic dust and atmospherically produced ^{26}Al/^{10}Be ratios (i.e., the retained fraction of ^{26}Al/^{10}Be will be, if anything, greater in dust than that calculated simply on the basis of composition and cross-section considerations).

D. ^{10}Be production in the atmosphere

The high-energy cosmic-ray proton flux arriving at the top of the earth's atmosphere interacts with the constituent nuclei to produce a broad energy spectrum of secondary particles. Since the mean free path of these particles is relatively small compared to the total atmospheric depth there is a type of equilibrium set up in which the particle spectra become approximately independent of altitude. Because the neutrons are affected only by nuclear interactions, while the charged particles are also slowed down by ionization losses,

this steady-state flux is dominated by neutrons at energies below ~500 MeV.[2] Because of this, and because the atmosphere is composed of ~80% nitrogen, the most important process for cosmic-ray ^{10}Be production in the atmosphere involves the reaction of medium energy (50–500-MeV) neutrons on nitrogen nuclei. Although this fact is well known, it seems often to have been forgotten (or at least ignored) in the discussion regarding cosmogenic ^{10}Be formation in the atmosphere. The cross section for this neutron-induced reaction has not yet been measured. Calculations on the production rate of ^{10}Be have therefore necessarily relied on a variety of estimates or indirect measurements. Unfortunately the degree of applicability of these estimates and/or measurements has not always been very carefully analyzed or stated.

As noted earlier, the earliest quantitative information in this direction came with the radiochemical measurements of Honda and Lal[8] for proton-induced spallation of carbon and semicarbazide (CN_3O) targets. These results were the basis for the estimate by Lal and Peters[2] that the over-all ^{10}Be/^{7}Be production ratio in the atmosphere would be ~0.5. Knowing the production rate in the atmosphere, and making several reasonable assumptions, one can calculate the deposition rate of undisturbed sea sediments by measuring their ^{10}Be activities. Alternatively, using other means of determining the sedimentation rate, one can calculate the atmospheric production rate of ^{10}Be. Using such a procedure, Amin[17] has measured ^{10}Be activities in several sea-sediment cores and has shown that they are reasonably consistent with the production estimate quoted above, although somewhat better agreement with other dating techniques was obtained in assuming ^{10}Be/^{7}Be ~0.3. In this case, also, lack of agreement between such a ratio and the mass spectrometrically measured ratio for ^{10}Be/^{7}Be in proton-irradiated oxygen targets has been cited as evidence against those cross-section data.[9,17] Here again it hardly seems necessary to point out that the connection between the indirect evidence and the experimentally measured value is rather tenuous. As we have mentioned elsewhere[1] there is ample reason for believing that a reaction such as ^{14}N$(n, 3p\,2n)^{10}$Be could have a significantly different cross section than ^{16}O$(p, 5p\,2n)^{10}$Be.

It is also interesting to reexamine the geophysical data in light of the new ^{10}Be half-life. Neglecting all other changes, a reduction in the half-life will cause a corresponding increase in the sedimentation rate as calculated by the ^{10}Be technique (assuming the dated layers are much younger than the ^{10}Be half-life). In order to continue to have the best agreement between this rate and that calculated by other techniques it would therefore be necessary to decrease the assumed atmosphere ^{10}Be production rate even further. Thus, while such a calculation is quite indirect, and involves a number of assumptions, a ratio of ~0.2 is probably the best estimate that can be made for the atmospheric ^{10}Be/^{7}Be production ratio from this point of view. While this is not inconsistent with the experimental results presented here for protons on nitrogen, it is obvious from what we have emphasized above that a satisfactory solution to this problem still awaits the measurement of neutron-induced cross sections. We are presently working on this problem.

As mentioned in the previous section, knowledge of the production rate of ^{10}Be in the atmosphere is also important to the problem of trying to measure the infall of cosmic dust on the earth. For example, using the early estimates for ^{10}Be/^{7}Be production, Lal and Venkatavaradan[25,28] have argued that the ^{26}Al/^{10}Be ratio measured in deep sea sediments indicated an infall of ~10^3 to 10^4 tons/day of such extraterrestrial material. Yokoyama,[29] using essentially the same data but basing his ^{10}Be production estimates on the much smaller mass spectrometrically measured cross sections for protons on oxygen, has concluded that there was no positive evidence for infall of such material. It seems to us that neither of these positions is tenable until the appropriate (i.e., neutron-induced) ^{10}Be production processes have been measured.

E. Cosmic ray ^{10}Be

To date we have been considering the ^{10}Be produced by the interaction of high-energy cosmic-ray protons and their secondaries on various target nuclei. Indeed, by far the largest fraction of the nuclear component of cosmic rays consists of protons (~90%) and α particles (~10%). However, the heavier nuclides (~1%) have been the object of considerable experimental study during the past decade because they are extremely rich in the information they can provide about the origin and history of cosmic rays. We deal in this section with the production of ^{10}Be by the interaction of these heavier nuclei with the interstellar hydrogen (protons). From the point of view of nuclear physics, the cross sections are identical to those for protons of the same velocity (MeV/nucleon) striking stationary target nuclei.

For some years now this laboratory has been engaged in a systematic study of the nuclear reactions which modify the composition of cosmic-ray nuclei during their passage through interstellar matter. As mentioned earlier, ^{10}Be is an

TABLE III. Abundance and $^{10}Be/^{7}Be$ production ratio for most prominant cosmic ray nuclides.

Element	Cosmic-ray abundance[a] observed	source	$\sigma(^{10}Be)/\sigma(^{7}Be)$ at 600 MeV/nucleon	References
B	27	0	3.3 ± 0.8[b]	Raisbeck and Yiou (10)
C	100	144	0.25 ± 0.02	Fontes *et al.* (19)
N	25	16	0.18 ± 0.06	This work
O	86	147	0.09 ± 0.03	Yiou (18)
Ne	20	28		
Mg	20	39	0.19 ± 0.04	This work
Si	14	32	0.13 ± 0.05	This work
Fe	11	32	0.24 ± 0.06[c]	Honda and Lal (8)

[a] M. M. Shapiro, R. Silberberg, and C. H. Tsao, in Proceedings of the Twelfth International Conference on Cosmic Rays, edited by A. G. Fenton and K. B. Fenton (Univ. of Tasmania Press, Hobart, Tasmania, 1971), Vol. 1, p. 221. Values are normalized to 100 for carbon arriving at earth.

[b] Cross-section data is for a ^{11}B target, corrected for ^{10}Be half-life = $(1.5 \pm 0.3) \times 10^{6}$ yr.

[c] Cross-section data at 730 MeV, corrected for ^{10}Be half-life = $(1.5 \pm 0.3) \times 10^{6}$ yr.

isotope of particular interest because its radioactive half-life is believed to be of the same order as the storage time of cosmic rays in the galactic disk. Thus, the determination of the spallation yield of ^{10}Be produced in cosmic rays, together with measurement of the fraction which survives to reach the earth, should be able to provide important information on the source, acceleration, and propagation conditions of cosmic rays.

In Table III we show the most abundant cosmic-ray nuclides which can produce ^{10}Be, together with available cross-section results. It is seen that the targets studied here represent three important species in that list for which there existed, up to now, no experimental measurements. Si and Mg, together with Ne, are the most important members of what is generally referred to in cosmic-ray terminology as the "H" group, and are believed to be important primary constituents of the cosmic rays. There is some question at present as to what fraction (if any) of the observed nitrogen is primary, but its abundance and its low mass number make it a potentially important contributor to ^{10}Be.

We should perhaps emphasize that our experimental results are for natural targets, whereas the isotopic composition of cosmic rays is not yet established. For example, it is expected (and recent results appear to confirm[30]) that a substantial fraction of the observed cosmic-ray N is secondary ^{15}N. By analogy with our recent experimental results for ^{13}C,[31] we would expect ^{15}N to have a significantly greater cross section than ^{14}N for production of the neutron-rich ^{10}Be.

In the case of Mg, Shapiro and Silberberg[32] have already pointed out the possible importance of the neutron-rich isotope ^{26}Mg in the cosmic-ray production of ^{10}Be. Their suggestion is based on the semiempirical formula of Silberberg and Tsao[33] which predicts a ^{10}Be cross section ~3 times larger in ^{26}Mg than in ^{24}Mg. It would be interesting to measure experimentally the magnitude of this enhancement for such a target since, in contrast to ^{13}C and ^{15}N, the dominant nuclear mechanism involved is probably different (fragmentation instead of spallation).

Excluding the possible contribution of neutron-rich isotopes such as those referred to above, the present experimental results confirm that ^{10}Be is a minor component of total Be spallation yield over the whole range of cosmic-ray primary nuclei. Using cross sections and source compositions such as those given in Table III (and estimates for other less important reactions), we are able to calculate the expected distribution of Be isotopes formed by nuclear reactions in the cosmic rays. We find that the production ratio $^{10}Be/\sum Be = 0.15 \pm 0.03$. As we have stated on previous occasions[1,10] we believe that this fraction is too small to draw any conclusions regarding ^{10}Be survival or decay using only element ratios such as Be/B. Resolution of the age question will therefore require isotopic separation of the cosmic-ray Be.

SUMMARY

^{10}Be cross sections have been measured for targets of Mg, Si, and N irradiated by 600-MeV protons; and N, by 150-MeV protons. These measurements, together with previous data, have been used to show that:

(1) A consideration of all available direct and "indirect" ^{10}Be cross-section measurements indi-

cates that, contrary to previous conclusions, there are no discrepancies that cannot be accounted for by experimental uncertainties and/or lack of appropriate data.

(2) In calculating cosmogenic ^{10}Be production in chondritic-type material, medium-weight nuclides such as Si and Mg cannot be ignored. In fact, for the primary galactic cosmic-ray spectrum alone, such targets appear to be more important than oxygen.

(3) The recoil loss of ^{10}Be produced in micron-sized dust particles will be much more important than previously estimated.

(4) A satisfactory discussion of cosmogenically produced ^{10}Be in the earth's atmosphere and its associated problems such as dating ocean sediments, measuring extraterrestrial dust infall, etc., must await experimental determination of the appropriate neutron-induced cross sections.

(5) The ^{10}Be formed by nuclear spallation in cosmic rays is a minor component of total Be from the whole range of cosmic-ray primary species (excluding possible effects due to neutron-rich source isotopes). The ^{10}Be yield is thus calculated to be too small to give any lifetime information without isotopic measurement of the cosmic-ray ^{10}Be.

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TABLE IV. ^{7}Be and ^{22}Na cross sections in targets of Al, Mg, and Si irradiated by 600-MeV protons.

Target	$\sigma(^{7}\text{Be})$ (mb)	$\sigma(^{22}\text{Na})$ (mb)
Al	4.6 ± 0.9 [a]	15 ± 3 [b]
	4.88 ± 0.35 [c]	
	4.95 ± 0.21 [d]	
Mg	6.5 ± 1.5 [a]	32 ± 7 [a]
	8.0 ± 1.6 [e]	11.7 ± 2.3 [e]
Si	5.3 ± 1.2 [a]	17 ± 4 [a]
	2.5 ± 0.5 [e]	8.5 ± 1.7 [e]
	3.1 [f]	

[a] This work.

[b] Used in this work as normalizing cross section.

[c] A. K. Lavrokhina, P. L. Moskaleva, V. V. Malyshev, and L. M. Satarova, Teor. Fiz. 43, 3 (1962) [transl.: Soviet Phys.—JETP 16, 1 (1963)] for 660 MeV, as corrected by Tobailem *et al.* (Ref. 36).

[d] Wogman and Brodzinski (Ref. 39).

[e] Rayudu (Ref. 37).

[f] R. Régnier, M. Sc. thesis, Université de Bordeaux, Bordeaux, France, 1971.

APPENDIX I

^{7}Be and ^{22}Na cross-section measurements in Si and Mg

As noted in the text it is necessary to have ^{7}Be cross sections in order to calculate absolute ^{10}Be cross sections from our measured ^{10}Be/^{7}Be ratios. There exist published results for all of the targets used in this work, and for N we have adopted values from the curve given by Epherre.[34] For Si and Mg targets, where the published data is much more scarce, we thought it worthwhile to remeasure the ^{7}Be cross sections.

A separate experiment was carried out in which an aligned target stack consisting of 25-mm-diam disks of Al (3×21.2 mg/cm^2), Si (54.1 mg/cm^2), Al (21.2 mg/cm^2), Mg (79.4 mg/cm^2), and Al (21.2 mg/cm^2) was irradiated in the external beam of the CERN 600-MeV synchrocyclotron. The duration of the irradiation (which was in parasite with the ISOLDE target) was ~75 h, and the integrated flux passing through our target ~10^{16} protons.

The irradiated foils were counted directly with a previously calibrated 80-cm^3 Ge(Li) detector (resolution 3.5 keV at 1 MeV) for ^{7}Be (0.477 MeV) and ^{22}Na (0.511 and 1.275 MeV). ^{7}Be and ^{22}Na cross sections were obtained relative to the ^{22}Na cross section in Al. Our choice of σ (^{22}Na) in Al is intermediate between the curve given by Cumming[35] and that proposed by Tobailem *et al.*[36] The uncertainty in this value is the major source of error in our measured cross sections. Because of the similarity of the ^{7}Be cross sections and the thickness of the target disks, errors due

to cross contamination of ^{7}Be recoil atoms were estimated to be small. This was checked by comparing the ^{7}Be/^{22}Na ratio in the Al disk, which was sandwiched between two other Al disks, with that from the two others which were in contact with the Si and Mg respectively. No significant differences could be observed. A summary of the results is given in Table IV. One sees that not only the ^{7}Be in Si but also the ^{22}Na cross sections in Si and Mg differ substantially from the previously reported values.

Unfortunately the CERN 600-MeV accelerator was shut down for modifications before we had an opportunity to repeat this experiment and therefore the results in Table IV are based on a single determination. However, we believe our estimates of the possible errors are realistic, and that the discrepancies with the earlier work are significant. We do not know the cause for this discrepancy, although one can note that the measurements of Rayudu[37] were done with a NaI crystal. It is possible that there was some difficulty in separating the contributions due to ^{7}Be (0.477 MeV) and annihilation radiation from ^{22}Na (0.511 MeV).

In this regard it is interesting to note that Rayudu[37] has reported similar ^{7}Be and ^{22}Na cross sections in Si and Mg targets at higher energies. If the problem discussed here is systematic, then it is very possible that those values also may be in error. We are presently carrying out higher-energy measurements to check this point.

APPENDIX II. CORRECTIONS FOR CONTRIBUTION OF VANADIUM TO ^{10}Be and ^{7}Be PRODUCTION IN VN TARGETS

In choosing a compound for a "nitrogen" target we looked for a material that (a) was readily available in a form convenient for irradiation purposes, and (b) one in which the correction for ^{10}Be and ^{7}Be production would be small and capable of being reliably estimated. Because of (b) we eliminated those compounds containing other light elements (such as C, O) since their cross sections were expected to be comparable to N. The production of ^{7}Be in very heavy elements falls off rapidly at the energies considered here, but the ^{10}Be/^{7}Be ratio is large at higher energies (Poskanzer, Butler, and Hyde[38]) and it is not clear how that ratio should be extrapolated to lower energies. In choosing a vanadium compound we have a compromise in which the ^{10}Be and ^{7}Be cross sections are relatively small compared to N and can be estimated from previous measurements on iron targets at a similar energy.

It can readily be shown that r_2, the ^{10}Be/^{7}Be cross-section ratio in nitrogen, can be expressed as

$$r_2 = \frac{\sigma_V(^{7}\mathrm{Be})}{\sigma_N(^{7}\mathrm{Be})}(R - r_1) + R, \tag{1}$$

where $\sigma_V(^{7}\mathrm{Be})$ and $\sigma_N(^{7}\mathrm{Be})$ are the ^{7}Be cross sections in V and N, and R and r_1 are the measured and assumed ^{10}Be/^{7}Be production ratios in VN and V, respectively. At 600-MeV bombarding energy we have taken $\sigma_V(^{7}\mathrm{Be}) = 2.5 \pm 0.7$ mb[39]; $\sigma_N(^{7}\mathrm{Be}) = 7 \pm 2$ mb; $R = 0.20 \pm 0.05$ (our measured value); and $r_1 = 0.25 \pm 0.07$. Our choice for r_1 is influenced mostly by the measured ratio in Fe at 730 MeV [σ (^{10}Be)/σ (^{7}Be) = 0.24 ± 0.06 when corrected for the new ^{10}Be half-life]. (See Table III).

Using these values we find $r_2 = 0.18 \pm 0.06$ and thus σ_N (^{10}Be) = 1.3 ± 0.5 mb.

Similarly, at 150 MeV we take σ_V (^{7}Be) = 0.2 ± 0.1 mb, σ_N (^{7}Be) = 6.0 ± 1.5, $R = 0.11 \pm 0.03$, $r_1 = 0.25 \pm 0.15$.

Although there is considerable uncertainty in the value of r_1 assumed at this energy (there being no measurements), the error introduced is not significant since σ_V (^{7}Be)/σ_N(^{7}Be) is very small. Thus we find $r_2 = 0.10 \pm 0.03$ and σ_N (^{10}Be) = 0.6 ± 0.2 mb.

It is necessary to emphasize here the distinction between the magnitude of the correction calculated by such a technique and the resulting uncertainty this correction introduces into the final ^{10}Be/^{7}Be ratio. It is clear from Eq. (1) that whenever R and r_2 are similar (i.e., the ^{10}Be/^{7}Be ratio for the compound and the desired component), the calculated correction to r_2 will be small. However, the resulting uncertainty could still be very large. To take an extreme example, if the absolute V cross sections were 10 times those for N, but with the same ^{10}Be/^{7}Be ratio, the calculated correction would be zero but the resulting uncertainty in r_2 very large. This is why we have attempted to minimize such an effect by choosing a compounding element in which the absolute cross sections are smaller than those in N. In targets having a mixture of elements with similar cross sections, such as semicarbizide (CN_3O) used by Honda and Lal,[8] the extraction of the cross section for any particular component element is obviously subject to much larger uncertainties.

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