

Title:

Measurement of Proton Production Cross Sections
of ^{10}Be and ^{26}Al from Elements Found in Lunar Rocks.

RECEIVED
JUL 19 1996
OSTI

Author(s):

J.M. Sisterson, K. Kim, P.A.J. Englert, M. Caffee,
A.J.T. Jull, D.J. Donahue, L. McHargue,
C. Castaneda, J. Vincent, and R.C. Reedy

Submitted to:

Proceedings of the 7th International Conference
on Accelerator Mass Spectrometry (held in Tucson
in May 1996), to be published in Nucl. Instrum
& Methods.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

MASTER

Los Alamos
NATIONAL LABORATORY



Los Alamos National Laboratory, an affirmative action/equal opportunity employer, is operated by the University of California for the U.S. Department of Energy under contract W-7405-ENG-36. By acceptance of this article, the publisher recognizes that the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or to allow others to do so, for U.S. Government purposes. The Los Alamos National Laboratory requests that the publisher identify this article as work performed under the auspices of the U.S. Department of Energy.

Paper 8B-1(9A-1)

Measurement of proton production cross sections of ^{10}Be and ^{26}Al from elements found in lunar rocks.

J.M. Sisterson¹, K. Kim², P.A.J. Englert², M. Caffee³, A.J.T. Jull⁴, D.J. Donahue⁴, L. McHargue⁴,
C. Castaneda⁵, J. Vincent⁶, and R. C. Reedy⁷.

¹Harvard Cyclotron Laboratory, Harvard University, Cambridge MA 02138, USA.

²Department of Chemistry, San Jose State University, San Jose CA 92192, USA.

³Nuclear Chemistry, Lawrence Livermore National Laboratory, Livermore, CA 94550, USA.

⁴NSF Arizona AMS Facility, University of Arizona, Tucson, AZ 85721, USA.

⁵Crocker Nuclear Laboratory, University of California, Davis, CA 95616, USA.

⁶TRIUMF, 4004 Wesbrook Mall, University of British Columbia, Vancouver B.C. V6T 2A3, Canada.

⁷Los Alamos National Laboratory, Group NIS-2, MS D436, Los Alamos, NM 87545, USA

Proton production cross sections for ^{10}Be , ^{26}Al , ^7Be and ^{22}Na produced by protons in silicon dioxide (for oxygen) and silicon targets over a wide proton energy range, 25 - 500 MeV, have been measured using accelerator mass spectrometry or non-destructive γ -ray spectroscopy. These cross sections are important for interpreting measurements of the concentration of these nuclides made in extraterrestrial materials by cosmic rays.

Address all correspondence to: J.M. Sisterson, Harvard Cyclotron Laboratory, 44 Oxford Street, Cambridge MA 02138, USA.

Tel: (617)495-2885; Fax: (617)495-8054; e-mail: sisterson@huhepl.harvard.edu

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

1. Introduction

Cosmic rays penetrate the lunar surface and interact with the lunar rocks to produce both radionuclides and stable nuclides. Production depth profiles for long-lived radionuclides produced in lunar rocks are measured using Accelerator Mass Spectrometry (AMS). For a particular radionuclide these production depth profiles can be interpreted to give an estimate for the solar proton flux over a time period characterized by the half life of the radionuclide under study [1, 2]. This analysis is possible if and only if the all the cross sections for the interactions of all cosmic ray particles with all elements found in lunar rocks are well known. In practice, the most important cross sections needed are the proton production cross sections, because 98% of solar cosmic rays and ~87% of galactic cosmic rays are protons.

The cross sections for the production of long-lived radionuclides were very difficult to measure before the development of AMS and only in recent years has significant progress been made in determining these essential cross sections [3, 4].

Oxygen and silicon are major constituents of lunar rocks. We have reported already ^{14}C production cross sections from O and Si [4, 5] for proton energies 25 -500 MeV, and $\text{O}(\text{p},\text{x})^{10}\text{Be}$ from 58 - 160 MeV [6]. Here we present new measurements for the cross sections $\text{O}(\text{p},\text{x})^{10}\text{Be}$, $\text{O}(\text{p},\text{x})^7\text{Be}$, $\text{Si}(\text{p},\text{x})^{10}\text{Be}$, $\text{Si}(\text{p},\text{x})^7\text{Be}$, $\text{Si}(\text{p},\text{x})^{26}\text{Al}$, and $\text{Si}(\text{p},\text{x})^{22}\text{Na}$ from ~30 - 500 MeV.

2. Experimental

These cross section measurements consist of four basic steps; target irradiations, short-lived radionuclide determination using non-destructive γ -ray spectroscopy, sample preparation and AMS determination.

Target irradiations: Three accelerator facilities are used for the irradiations. Irradiations are made for proton energies < 68 MeV at the University of California, Davis (Davis); for ~55 - 160 MeV at the Harvard Cyclotron Laboratory, Harvard University (HCL); for 200 - 500 MeV, the Tri-

Universities Meson Facility at the University of British Columbia (TRIUMF). At all three facilities the overall irradiation philosophy is the same, but details of the irradiation schemes vary.

At all facilities, thin target conditions are always used. This is achieved by using thin target foils and short target stacks, such that at most ~2 MeV would be lost in an individual target and < 10 MeV in the entire target stack. These conditions minimize both the loss due to protons scattering out of the stack, and the production of neutrons within the stack. Si targets are 0.0754 - 0.076 cm thick, 1.0 - 1.5 cm in diameter and weigh ~0.138 - ~0.309g. SiO₂ (for O) targets are 0.0508 - 0.056 cm thick, 1.0 - 1.5 cm in diameter and weigh ~ 0.095 - 0.214g. Catcher foils were used to minimize the effects due to recoils.

At all three facilities, irradiations are designed so that the entire proton beam is intercepted by both the target stack and the monitor chamber used to get a direct measurement of the number of protons through the target. The monitor chamber is calibrated for each proton energy using a Faraday cup. Aluminum monitor foils are included at the front of the target stack and the reactions $^{27}\text{Al}(p,x)^{24}\text{Na}$ and $^{27}\text{Al}(p,x)^{22}\text{Na}$ used as an additional dosimetric check [5].

For most irradiations, irradiation times were designed to produce $\sim 10^7$ ^{14}C atoms in the central target foil and one of the catcher foils was used for ^{10}Be and ^{26}Al determination by AMS.

γ -ray spectroscopy: ^7Be and ^{22}Na are determined in all targets at HCL using non-destructive γ -ray spectroscopy.

Sample preparation: Following the addition of Be and Al carriers, with the exception of the Al targets, which required no Al carrier, the targets are dissolved in acidic solutions. After dissolution several steps of HClO₄ fuming are performed. Standard ion-chromatographic techniques are used to separate Be and Al from other species. Since the elution of Be and Al from cation columns differ this technique also separates the Be from the Al. Both Al and Be are precipitated in a basic solution as hydroxides. The Al⁻ and Be⁻ hydroxides are then dried and converted to oxides at high temperature (~ 800 C). The samples are mixed with Ag prior to AMS analysis. Successful AMS

analysis of Be requires a sample preparation protocol that yields a sample relatively free of boron, an isobaric interference. With only a few exceptions this procedure produced both Al and Be samples free of interfering species. In those instances where interferences created problematic analyses repeat measurements were performed.

AMS determination: ^{10}Be analyses for some of the silicon dioxide targets were made at the University of Arizona and the results reported [6]. All other targets were analyzed at the Lawrence Livermore National Laboratory AMS Facility (LLNL) [7] with the chemistry done at San Jose State University. Both ^{10}Be and ^{26}Al measurements are made utilizing a dE/dX detector. For the ^{10}Be measurements the boron background is reduced by using a thin Havar foil as the entrance window for the dE/dX detector. For most of the analyses <5% of the counts accumulated in the ^{10}Be spectrum are attributed to boron reactions in the Havar. A correction based on a “dirty boron blank”, which is prepared at LLNL, is applied in all instances.

3. Results

New cross section measurements for $\text{Si(p,x)}^7\text{Be}$, $\text{Si(p,x)}^{10}\text{Be}$, $\text{Si(p,x)}^{22}\text{Na}$, and $\text{Si(p,x)}^{26}\text{Al}$, are given in Table 1. and new cross section measurements for $\text{O(p,x)}^7\text{Be}$ and $\text{O(p,x)}^{10}\text{Be}$ from oxygen in SiO_2 targets in Table 2; Table 2 includes corrected values for the cross sections previously reported [6].

The principal sources of error are as follows; 2% for the number of atoms/cm² of the target element; 5% for the number of protons through the target at all three facilities, - except for some irradiations of Si and SiO_2 targets at HCL where this error was 7%; and a range of values for the AMS determination depending on the number of ^{10}Be or ^{26}Al atoms made in the target. The irradiation conditions were optimized for ^{14}C production, so for some target/energy combinations, too few ^{10}Be atoms were produced for optimum AMS determinations leading to an increased error

in the AMS measurement. However, even under these conditions, most cross sections were measured to better than 10%.

Results of the new cross section measurements are shown in Figures 1 - 6 for the cross sections for $\text{Si}(p,x)^{10}\text{Be}$, $\text{Si}(p,x)^{26}\text{Al}$, $\text{Si}(p,x)^7\text{Be}$, $\text{Si}(p,x)^{22}\text{Na}$, $\text{O}(p,x)^{10}\text{Be}$, and $\text{O}(p,x)^7\text{Be}$. These new measurements are compared to those in the literature and in most cases, the agreement is very good [3,8-25].

4. Discussion and Conclusions

These irradiations are made using thin targets and small target stacks to minimize both the production of neutrons within the stack and the scattering of protons out of the stack. All irradiations are designed to include a direct measurement of the number of protons through a target stack, with monitor foils used as an additional check. These experimental conditions allow a simple calculation for the cross section. The majority of the new cross section values reported here, agree well with the cross sections measured over the past decade by others [3, 8, 9].

Production depth profiles of cosmogenic radionuclides measured in lunar rocks can be analyzed to give estimates of the solar proton flux over a time period characterized by the half life of the radionuclide under study [1]. Accurate and precise cross section measurements of all the relevant proton production cross sections are essential for reliable estimates of the solar proton flux to be made. The progress in measuring these essential cross sections over the past decade is evident from Figures 1 - 6. Many cross sections have now been measured by more than one independent technique, with consistent results. These values can now be used with confidence to get better estimates of the solar proton flux from the ancient past to present solar cycles.

Cross sections for the production of ^{10}Be below ~ 150 MeV have only recently been measured and are similar to the estimated cross sections used by Nishiizumi [26] to interpret ^{10}Be measured in lunar rock 68815. These recent ^{10}Be cross section measurements eliminate the possibility noted

by Nishiizumi [26] that the cross sections assumed for ^{10}Be production at low energies were wrong, and support the case for a soft solar proton spectrum with relatively few high-energy protons over the last few million years.

Acknowledgments

Grant NAGW 4609 from the National Aeronautics and Space Administration provided partial support for this work at Harvard University, San Jose State University and Lawrence Livermore National Laboratory. Part of this work was performed under the auspices of the Department of Energy by Lawrence Livermore National Laboratory under contract W-7405-Eng-48. The work performed at the Los Alamos National Laboratory was supported by NASA and done under the auspices of the Department of Energy.

Table 1. Cross sections for the production of radionuclides from Si targets.

Proton Energy*	Si(p,x) ⁷ Be	Si(p,x) ¹⁰ Be	Si(p,x) ²² Na	Si(p,x) ²⁶ Al
MeV	mb	mb	mb	mb
500	5.7±0.43	0.554±0.033	17.7±1.31	18.7±1.09
400	4.25±0.32	0.397±0.029	17.5±1.29	20.4±1.18
300	3.9±0.3	0.262±0.0159	17.5±1.31	22.1±1.3
200	2.9±0.26	0.114±0.0073	15.1±1.16	24.8±1.44
157.3±1.0	1.41±0.15	0.066±0.0064	16.1±1.45	32.6±2.54
148.2±2.0	1.3±0.15	0.062±0.005	16.1±1.46	29.1±2.22
128.7±2.0				32.3±1.88
128.0±2.0	1.19±0.15	0.045±0.0044	16.5±1.49	34.6±2.64
127.5±2.0			17.9±0.99	
127.0±2.0	1.11±0.34	0.048±0.0038		
97±3.0	0.87±0.16	0.023±0.0026	16.8±1.53	36.4±2.76
87.4±3.0	0.84±0.14	0.019±0.0019	18.1±1.63	38.8±3.01
77.2±3.5	0.75±0.13	0.011±0.0013		43.9±3.36
67.4	0.66±0.24			
66.9	0.81±0.16	0.0097±0.0012	19.9±1.82	47.0±3.57
65.6		0.005±0.0005	25.5±2.4	50.5±2.87
65.2	0.88±0.203			
61.7±4.0	0.41±0.16	0.0065±0.0009	20.0±1.84	53.0±4.04
59.0			22.4±2.08	
58.7			22.0±2.09	
56.5±4.0	0.31±0.15	0.0024±0.0009	16.8±1.54	51.3±5.04
52.4	0.5±0.105			
50.2		0.0009±0.0002	11.6±1.09	70.2±4.05
44.5				67.0±3.95
41.0			1.16±0.12	
29.8				5.99±0.38

*200-500 MeV TRIUMF;

lower energies; with energy spread indicated, HCL; no energy spread indicated, Davis;

Table 2. Cross sections for the production of radionuclides from O using SiO₂ targets.

Proton Energy*	O(p,x) ⁷ Be	O(p,x) ¹⁰ Be
MeV	mb	mb
500	10.2±0.77	
400	10.3±0.77	1.18±0.068
300	11.1±0.85	0.952±0.058
200	10.3±0.77	0.712±0.046
158.9±1.0	6.82±0.63	0.56±0.048
149.9±2.0	7.11±0.66	0.51±0.057
129.9±2.0	7.3±0.69	0.42±0.044
129.9±2.0	6.27±0.56	
99.9±3.0	7.57±0.74	0.44±0.047
89.9±3.0	7.25±0.68	0.33±0.038
79.9±3.5	7.66±0.73	0.27±0.032
69.9±3.5	7.69±0.75	
67.4		0.33±0.02
64.8±4	7.68±0.75	0.21±0.037
59.8±4	8.44±0.83	0.18±0.022
49.9		0.038±0.0029
41.8		0.0079±0.0008
30.9		0.0052±0.0004

*200-500 MeV TRIUMF;

lower energies; with energy spread indicated, HCL; no energy spread indicated, Davis;

References

1. Rao R. M. et al., *Geochim. et Cosmochim. Acta*, 58, (1994), 4231.
2. Reedy R.C. and Marti K. *The Sun in Time*, (U. of Arizona Press), (1991), 260.
3. Michel R. et al., *Nucl. Instr. and Meth. Phys. Res.*, B103, (1995), 183.
4. Sisterson J. M. et al., *Lunar Planet. Sci. XXVII*, (1996), 1209.
5. Sisterson J. M. et al., *Nucl. Instr. and Meth. Phys. Res.*, B92, (1994), 510.
6. Sisterson J. M. et al., *Lunar Planet. Sci. XXIII*, (1992), 1305.
7. Southon J. R. et al., *Nucl. Instr. and Meth. Phys. Res.*, B52, (1990), 301.
8. Bodemann R. et al., *Nucl. Instr. and Meth. Phys. Res.*, B82, (1993), 9.
9. Michel R. et al., *Analyst*, 114, (1989), 287.
10. Dedieu J., Thesis, Université de Bordeaux (1979).
11. Raisbeck G. M. and Yiou F., *Phys. Rev. C*9, (1974), 1385.
12. Raisbeck G. M. and Yiou F., 15th Int. Cosmic Ray Conf. Plovdiv, Bulgaria 2, (1977), 203.
13. Furukawa M. et al., *Nucl. Phys.* 174, (1971), 539.
14. Bimbot R. and Gauvin H., *Compt. Rend B*273, (1971) 1054.
15. Sheffey D. W. et al., *Phys. Rev.* 172, (1968), 1094.
16. Raisbeck G. M. and Yiou F., *Phys. Rev. C*12, (1975), 915.
17. Regnier S. et al., *C. R. Acad. Sci. Paris* 280, (1975), 513.
18. Furukawa M. et al., *Nucl. Phys.* 69, (1965), 362.
19. Korteling R. G. and Heymann D., *Phys. Rev C*1, (1970), 1960.
20. Heydegger H. R. et al., *Phys. Rev. C*14, (1976), 1506.
21. Amin B. S. et al., *Nucl. Phys. A*195, (1972), 311.
22. Yiou F. et al., *J. Geophys. Res.* 74, (1969), 2447.
23. Yiou F. et al., *Phys. Rev* 166, (1968), 968.

24. Lafleur M. S. et al., Can. J. of Chem. 44, (1966), 2749.
25. Inoue T. and Tanaka S., J. Inorg. Nucl. Chem. 38, (1976), 1425.
26. Nishiizumi K. et al., Proc. 18th Lunar Planet. Sci. Conf. (Cambridge University Press), (1988), 79.

Figure Captions

Figure 1. $\text{Si}(p,x)^{10}\text{Be}$; errors are as shown or are smaller than the plotted symbol.

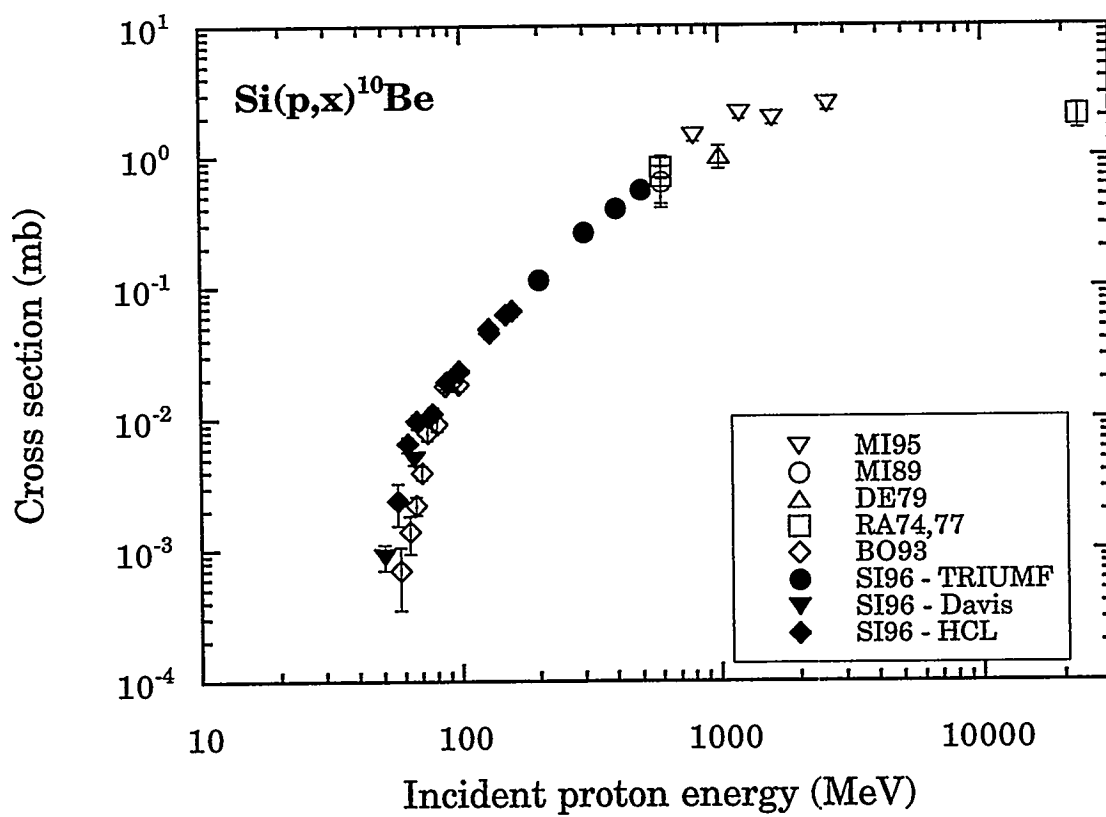
Figure 2. $\text{Si}(p,x)^{26}\text{Al}$; errors are as shown or are smaller than the plotted symbol.

Figure 3. $\text{Si}(p,x)^7\text{Be}$; errors are as shown or are smaller than the plotted symbol.

Figure 4. $\text{Si}(p,x)^{22}\text{Na}$; errors are as shown or are smaller than the plotted symbol.

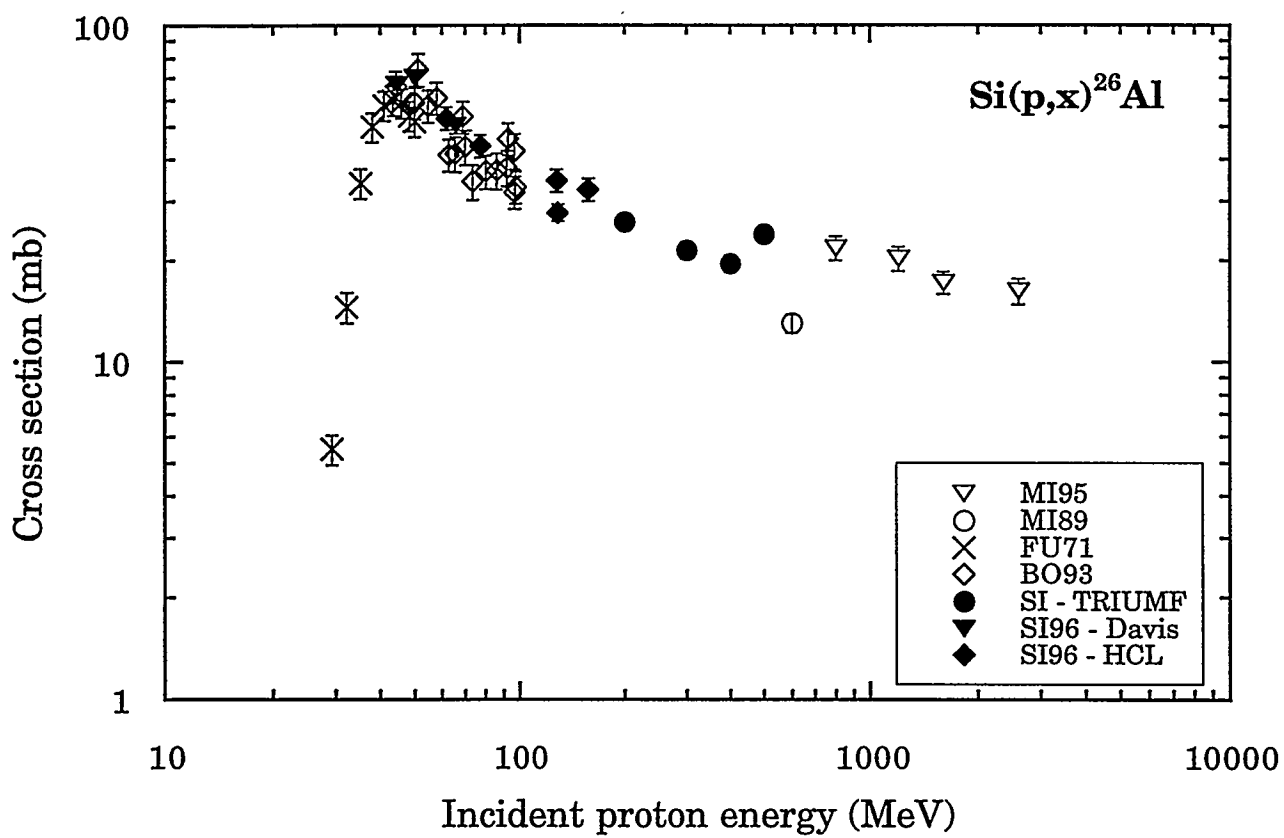
Figure 5. $\text{O}(p,x)^{10}\text{Be}$; errors are as shown or are smaller than the plotted symbol.

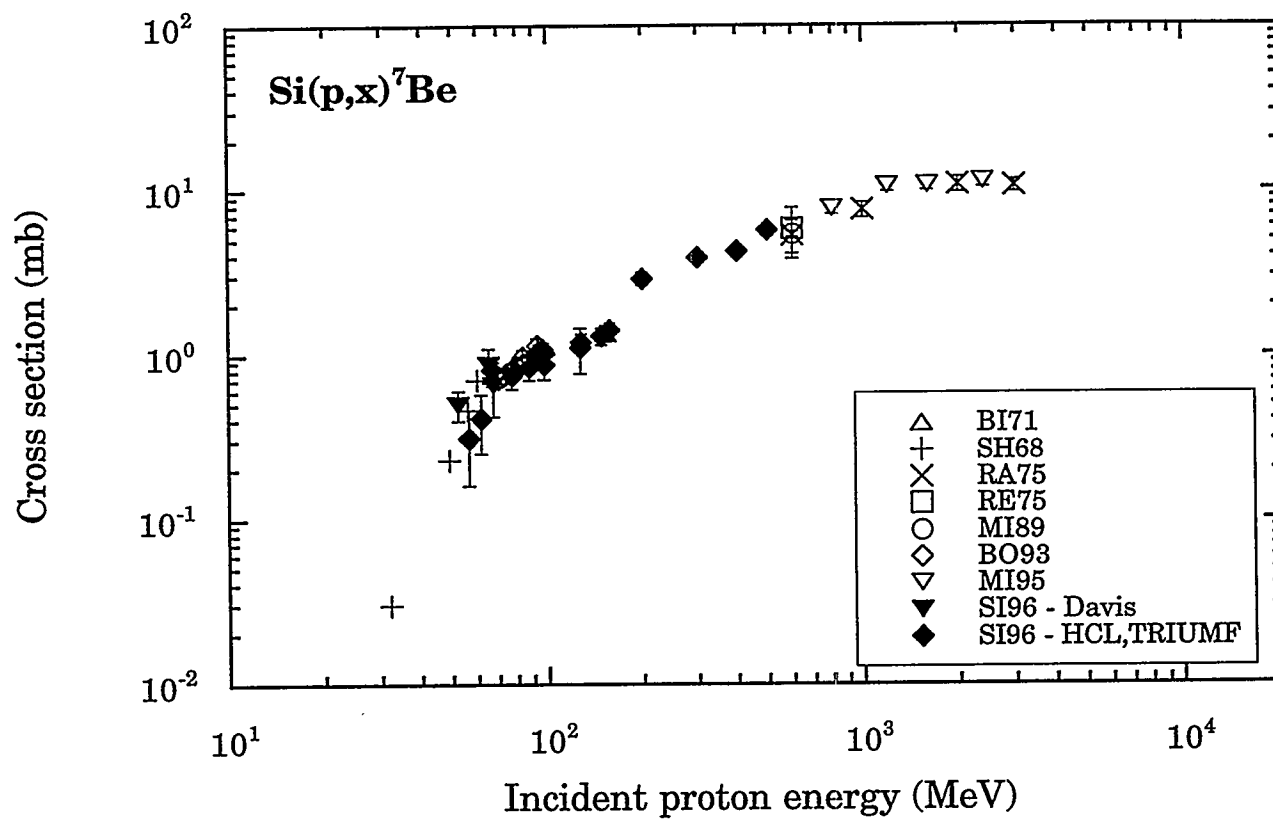
Figure 6. $\text{O}(p,x)^7\text{Be}$; errors are as shown or are smaller than the plotted symbol.



JMS 5/6/96

Figure 1





JMS 5/6/96

Figure 3

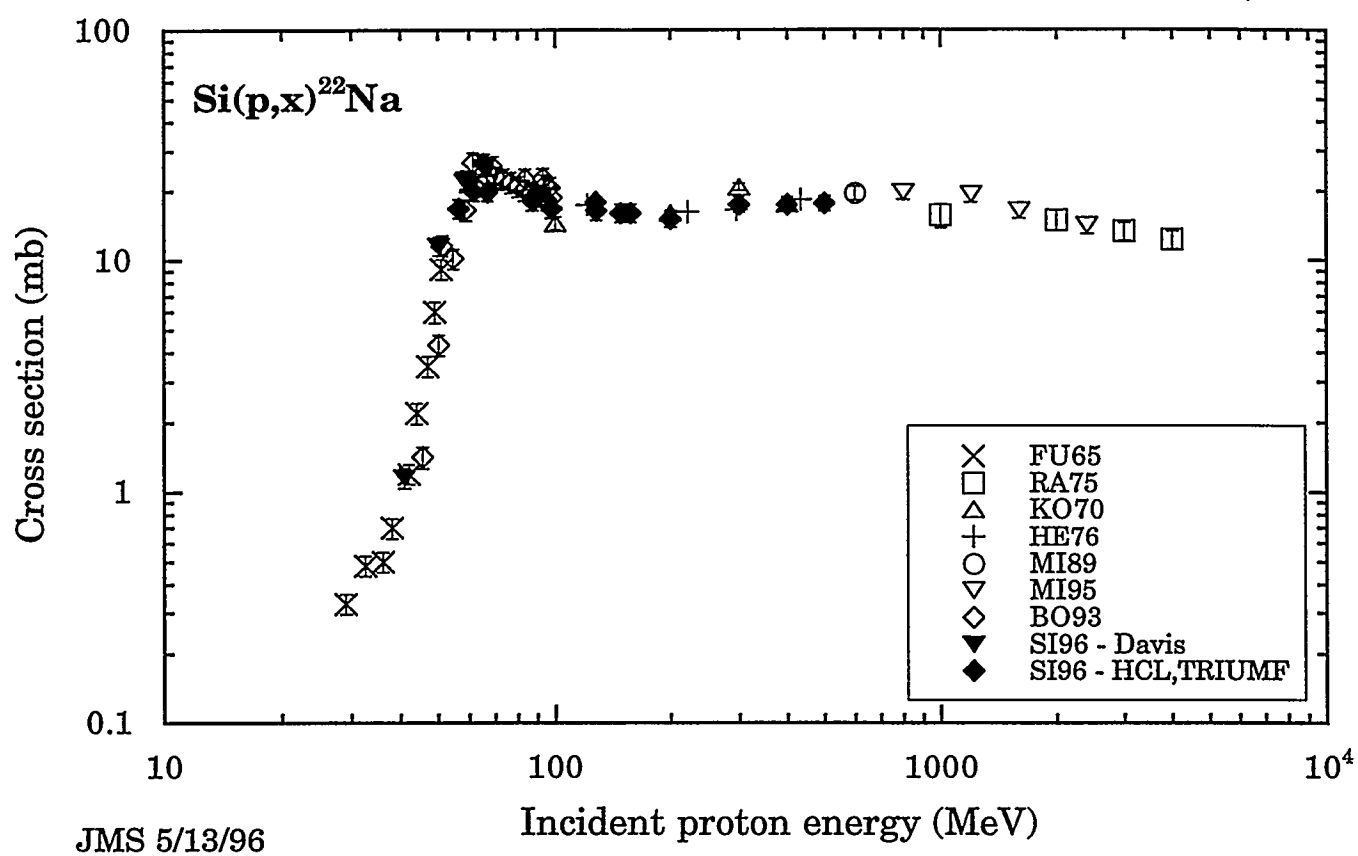
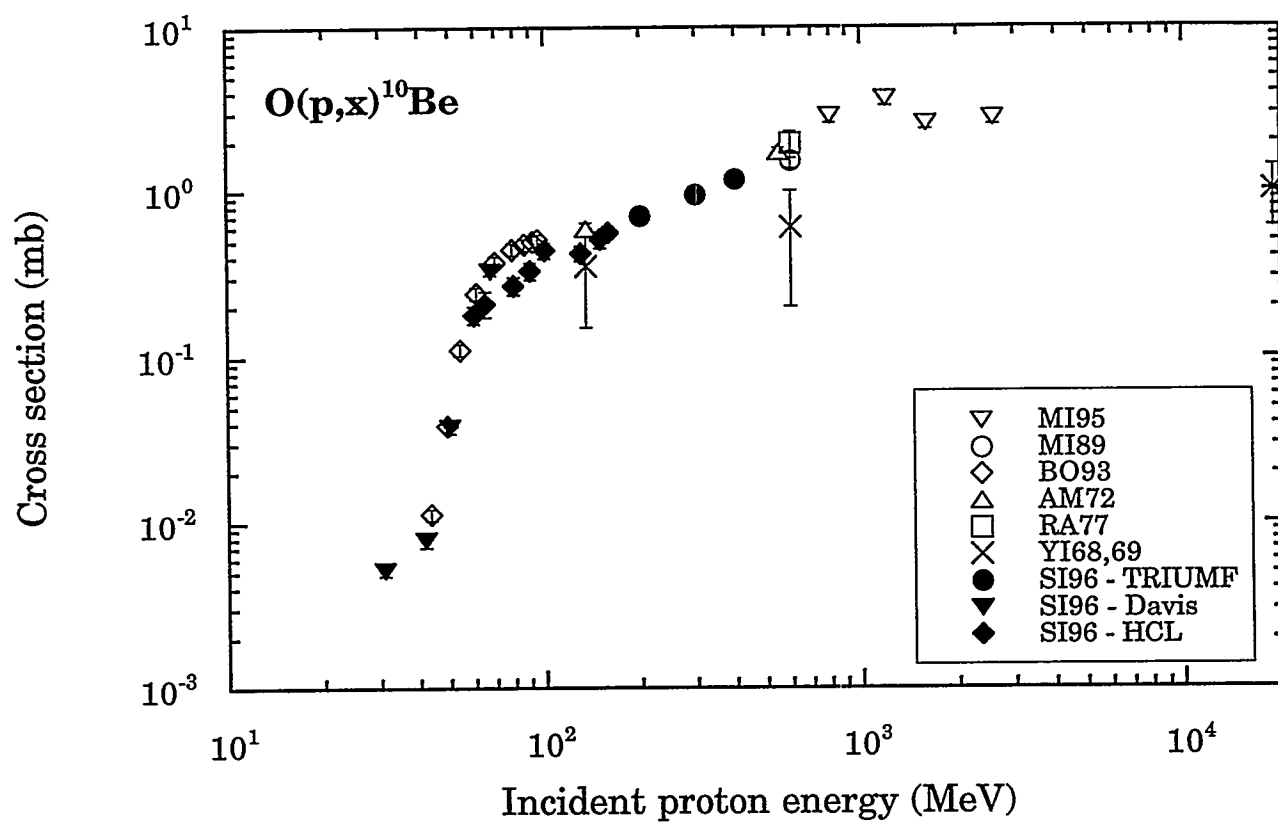


Figure 4



JMS 5/6/96

Figure 5

