High-Energy Proton Spallation of Argon†

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The cross sections for production of ^{39}Cl, ^{38}Cl, ^{37}S, ^{34m}Cl, ^{29}Al, ^{28}Al, ^{28}Mg, ^{27}Mg, ^{24}Na, ^{22}Na, ^{18}F, and ^{7}Be from the proton spallation of argon at 310, 425, and 578 MeV are reported. Experimental production rates and experimental production ratios of the various radioisotopes to ^{38}Cl are compared to theoretical spallation yield calculations. Applications of the cross sections to health physics problems associated with high-energy accelerators and to studies of atmospheric processes are discussed.

I. INTRODUCTION

The high-energy proton spallation of argon is of considerable interest from a health physics standpoint.[1] In addition, a study of this spallation process is required to determine appropriate tracers for various atmospheric processes.[2-5] Argon comprises 0.9% of air, and is the dominant source of high-Z spallation products produced by cosmic rays in the atmosphere or by high-energy particles within the vault of a particle accelerator. The type and quantities of activation products of the air in an accelerator vault are governing factors in determining the exchange rate of the air and the amount of time before a vault can be opened after an irradiation. The steady-state concentrations of the cosmic-ray-induced spallation products in the atmosphere are of prime importance for use as tracers of precipitation scavenging processes. Studies of precipitation scavenging and associated rapid atmospheric mixing rates[5-8] have been hampered by both the low concentrations and unknown production rates of the spallation products of argon.

The concentrations and production rates could be calculated if the cross sections for production were known as a function of energy. Previously, one determination of the 1000-MeV proton spallation cross sections of ^{38}Cl and ^{39}Cl from argon had been made.[9] Production cross sections can be calculated according to the semiempirical method of Rudstam,[10] but even the best of his equations [charge distribution, mass distribution (CDMD)] is uncertain by approximately 75% and frequently differs from experimental data by factors ranging from 2 to 10.

In order to more accurately determine the production cross sections of as many high-energy proton spallation products of argon as possible, liquid argon was irradiated directly with protons of 310, 425, and 578 MeV; and activation products were determined with the aid of a Ge(Li) diode γ-ray spectrometer. The experimental results were then compared with theoretical calculations, and the calculated data were extrapolated to other energies.

II. EXPERIMENTAL PROCEDURES

Liquid argon was bombarded with 310-, 425-, and 578-MeV protons at the Space Radiation Effects Laboratory cyclotron. The argon was contained in a 6-cm-diam by 11-cm-deep glass Dewar which was sealed with a rubber stopper having only a small pressure vent. Proton beams from 4.6- to 6.8-cm in diam were incident normal to the axis of the Dewar. Proton fluxes were monitored by a calibrated proportional counter and by the spallation products induced in aluminum foils and thin Lucite sheets placed in the beam path ahead of the argon. Integral doses of $(1-10)\times10^{13}$ protons were obtained during irradiation times, which varied from 17 to 60 min. Beam currents appeared to be constant within ±15% during all irradiations. A maximum of approximately 15% loss of argon was observed during any irradiation.

Immediately after each irradiation, the liquid argon was transferred from the "irradiation" Dewar to a precooled "counting" Dewar and counted at varying distances up to 12 in. from a 3.6-cm^3 Ge(Li) diode γ-ray spectrometer. Pulses were stored in a Nuclear Data multichannel analyzer, utilizing 1024 channels calibrated at either 3 or 6 keV per channel. Successive counting periods of 20 to 8000 sec were taken for several hours after each irradiation so that decay curves were obtained for use in the data analysis. The counting efficiency as a function of γ-ray energy deposited in the diode was obtained by calibration of known radionuclide sources which were standardized against IAEA standards. The various radionuclides were contained in the same counting geometry and position in the Dewar as was the irradiated argon.

The "irradiation" Dewar was swabbed with alcohol and dry tissues which were counted to determine what, if any, quantity of spallation products

had adhered to the sides of the Dewar. The empty, wiped Dewar was also counted to ensure that no spallation products remained. That activity not transferred to the "counting" Dewar was less than 5% of the total activity, which also demonstrates that an insignificant fraction of any measured radionuclide is produced by spallation of the elemental constituents of the Dewar itself. After all the argon had evaporated from the "counting" Dewar, it also was swabbed clean in a similar procedure, and the swabs were counted in a standard 0.5-in.-thick by 2-in.-diam counting geometry on a large crystal multidimensional γ-ray spectrometer[11,12] at our Richland laboratories to determine the long-lived radioisotopes ^{24}Na, ^{22}Na, and ^{7}Be. The collection efficiency of the swabs for these radioisotopes was found to be greater than 98% based on a comparison of the activity of ^{24}Na measured in the Dewar containing the argon and in the swabs.

III. RESULTS

A. Experimental

The radioisotopes ^{39}Cl, ^{38}Cl, ^{37}S, ^{34m}Cl, ^{29}Al, ^{28}Al, ^{28}Mg, ^{27}Mg, ^{24}Na, and ^{18}F were observed in the diode counts of the liquid argon. In addition, ^{24}Na, ^{22}Na, and ^{7}Be were observed in the swabs of the "counting" Dewar. A short-lived positron emitter(s), probably ^{14}O, ^{15}O, and/or ^{17}F, was observed at 578 MeV, but no quantitative yields could be derived from the data. Not all radioisotopes were observed at all energies, but this is principally due to the type and length of counts which were made at each energy, rather than the lack of production of a radioisotope. For example, ^{18}F was observed only at 300 MeV since that target was the only one counted long enough to measure ^{18}F after the interfering ^{34m}Cl radioactivity had decayed. Subsequent counts were not made at the higher energies until after all of the ^{18}F had decayed.

Given in Tables I through III are the measured proton-spallation cross sections of argon at 310, 425, and 578 MeV, respectively. The error values listed in the tables for the experimental data are consistent with the errors associated with counting statistics, counter calibration, proton flux, effective target thickness, background, and Compton production. Since a rather large uncertainty is associated with the proton flux, the ratio of the cross section of the various radioisotopes to that of ^{38}Cl is also given. Only statistical uncertainties remain in the ratios; hence, the ratios are much

TABLE I. 310-MeV proton spallation of argon cross sections in mb.

Radioisotope	σ (Expt.)	σ (Calc.)	σ/σ (^{38}Cl) (Expt.)	σ/σ (^{38}Cl) (Calc.)
^{39}Cl	17.8 ± 5.9	2.97	1.20 ± 0.12	0.25
^{38}Cl	14.7 ± 4.6	11.9	1	1
^{37}S	0.77 ± 0.25	1.17	0.052 ± 0.002	0.099
^{34m}Cl	1.09 ± 0.75	4.20[a]	0.075 ± 0.045	0.353[a]
^{28}Mg	0.51 ± 0.19	0.16	0.035 ± 0.007	0.013
^{27}Mg	1.93 ± 0.66	0.80	0.130 ± 0.017	0.067
^{24}Na	1.71 ± 0.54	1.35	0.116 ± 0.012	0.113
^{22}Na	0.94 ± 0.30	0.70	0.064 ± 0.006	0.059
^{18}F	3.3 ± 1.1	0.35	0.222 ± 0.029	0.029
^{7}Be	0.80 ± 0.25	0.0047	0.054 ± 0.005	0.00039

[a]Calculated for ^{34}Cl, not for ^{34m}Cl.

TABLE II. 425-MeV proton spallation of argon cross sections in mb.

Radioisotope	σ (Expt.)	σ (Calc.)	σ/σ (^{38}Cl) (Expt.)	σ/σ (^{38}Cl) (Calc.)
^{39}Cl	11.4 ± 3.0	2.48	1.13 ± 0.15	0.237
^{38}Cl	10.1 ± 2.4	10.5	1	1
^{37}S	1.16 ± 0.28	1.09	0.116 ± 0.007	0.104
^{34m}Cl	0.56 ± 0.19	4.54[a]	0.056 ± 0.014	0.435[a]
^{27}Mg	1.13 ± 0.28	1.24	0.112 ± 0.008	0.119
^{24}Na	1.71 ± 0.51	2.46	0.170 ± 0.032	0.235
^{22}Na	0.70 ± 0.17	1.41	0.070 ± 0.007	0.135
^{7}Be	0.49 ± 0.12	0.021	0.049 ± 0.005	0.0020

[a]Calculated for ^{34}Cl, not for ^{34m}Cl.

TABLE III. 578-MeV proton spallation of argon cross sections in mb.

Radioisotope	σ (Expt.)	σ (Calc.)	σ/σ (^{38}Cl) (Expt.)	σ/σ (^{38}Cl) (Calc.)
^{39}Cl	11.4 ± 4.0	2.06	1.44 ± 0.18	0.228
^{38}Cl	7.9 ± 2.6	9.04	1	1
^{37}S	0.92 ± 0.32	0.98	0.117 ± 0.011	0.108
^{34m}Cl	0.47 ± 0.18	4.61[a]	0.060 ± 0.010	0.510[a]
^{29}Al	19.7 ± 6.9	3.09	2.50 ± 0.27	0.34
^{28}Al	4.2 ± 1.6	9.7	0.53 ± 0.10	1.08
^{28}Mg	10.2 ± 5.4	0.30	1.29 ± 0.53	0.033
^{27}Mg	1.18 ± 0.42	1.67	0.150 ± 0.021	0.185
^{24}Na	2.05 ± 0.68	3.7	0.259 ± 0.027	0.411
^{22}Na	0.84 ± 0.28	2.31	0.107 ± 0.011	0.255
^{7}Be	0.76 ± 0.25	0.062	0.096 ± 0.010	0.0068

[a] Calculated for ^{34}Cl, not for ^{34m}Cl.

more precisely known. This uncertainty was calculated according to

$$\sigma_{\text{ratio}} = (\text{ratio}) \times [(\sigma_y/D_y)^2 + (\sigma_{38}/D_{38})^2]^{1/2},$$

where σ_y and σ_{38} are the uncertainties, and D_y and D_{38} the disintegration rates of the various radioisotopes and ^{38}Cl, respectively.

B. Theoretical

The values of the various cross sections were calculated according to the CDMD cross-section equation of Rudstam.[10] This is a five-parameter equation corresponding to an exponential yield-mass distribution and a Gaussian charge distribution. Rudstam evaluated another form of this type of distribution (CDMD-G) and two distributions in terms of isotopic distributions and the elemental distribution (IDED-G and IDED) with aid of an IBM 7090 computer. The parameter values in the various equations were determined by fitting the functions to the available experimental data. The CDMD equation was demonstrated to be the best choice for the cross-section distribution.

The calculated cross sections and ratios of cross sections are listed in the tables beside the experimental data for comparison. Complete excitation functions were generated for the radioisotopes ^{38}Cl, ^{37}S, ^{27}Mg, ^{24}Na, and ^{22}Na. Four of the calculated excitation functions were multiplied by constant values in order to better fit them to the experimental data points. These multipliers were 0.562 for ^{22}Na, 0.787 for ^{24}Na, 0.752 for ^{37}S, and 0.935 for ^{38}Cl. These values are certainly all within the basic error limit of the Rudstam equation,

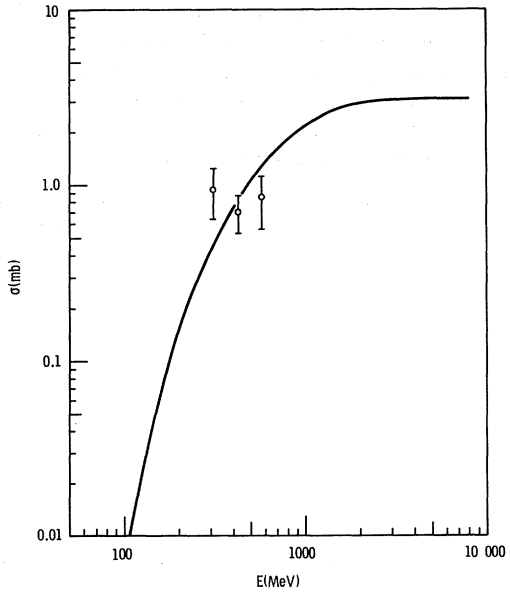

FIG. 1. Proton spallation cross sections of ^{22}Na from argon.

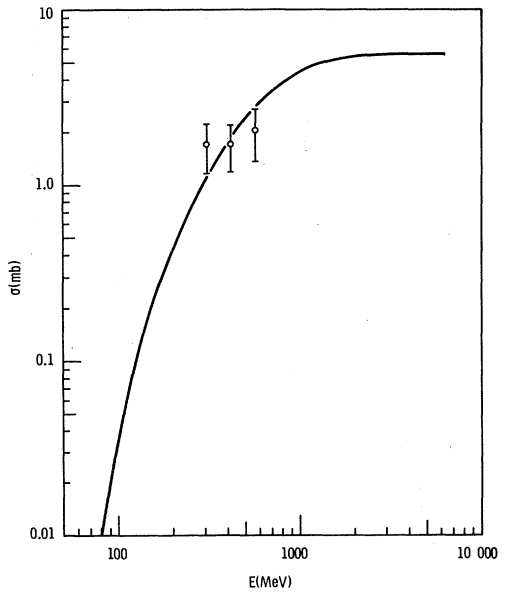

FIG. 2. Proton spallation cross sections of ^{24}Na from argon.

which is ±75%. The excitation functions are plotted in Figs. 1 through 5 as solid lines, and the points are experimental data.

IV. DISCUSSION

Hower and Kaufman[9] have reported the only other experimental cross sections for the high-energy proton spallation of argon. Their bombarding energy (1000 MeV) was higher than those used in this communication, and their measured cross sections for the production of ^{39}Cl (24.0 ± 0.8 mb) and ^{38}Cl (15.7 ± 0.9 mb) are somewhat higher than those reported herein. Their value for the ^{38}Cl cross section is plotted in Fig. 5; and although the Rudstam equation produces a decreasing cross section at energies higher than those used in this work, their experimental datum creates a positive slope. Korteling and Caretto[13] have determined the production cross sections of ^{22}Na and ^{24}Na from a variety of targets at 400 MeV. An interpolation of their ^{22}Na and ^{24}Na yields for S, Cl, K, Ca, and Sc targets agrees well with the ^{24}Na data reported herein. Since the only difference in the determination of ^{22}Na and ^{24}Na in this work is the difference in counter efficiencies, the measured yields of ^{22}Na and ^{24}Na are equally accurate.

The calculated cross sections for ^{39}Cl shown in the tables are all quite low when compared to the experimental data. This is to be expected, however, since the equation breaks down at product masses very near the target mass. The equation is based on and generates a distribution of product yields, and it is therefore oblivious to the fact that some isotopes in the vicinity of the target mass cannot be formed at all. Similarly, the calculated cross sections for ^{7}Be are extremely low, since the equation assumes that the target nucleus is spallated down to the product nucleus and does not take into account the much more probable occurrence of such a light fragment being emitted as an entity from the struck nucleus. Those calculated values shown for ^{34m}Cl are too high, since they represent the total yield of ^{34}Cl for both the ground state and the metastable state, whereas the experimental data are only reported for the yield of the metastable state. The disagreement between experimental and Rudstam calculated values for ^{18}F at 310 MeV and ^{29}Al and ^{28}Mg at 578 MeV indicates the possible error inherent in the semiempirical equation. The experimental cross sections for ^{18}F and ^{29}Al seem to be too high and the calculated values more nearly correct, while the calculated ^{28}Mg datum seems too low and the experimental number more nearly correct in this case. All other data demonstrate excellent agreement between calculated and experimental cross sections.

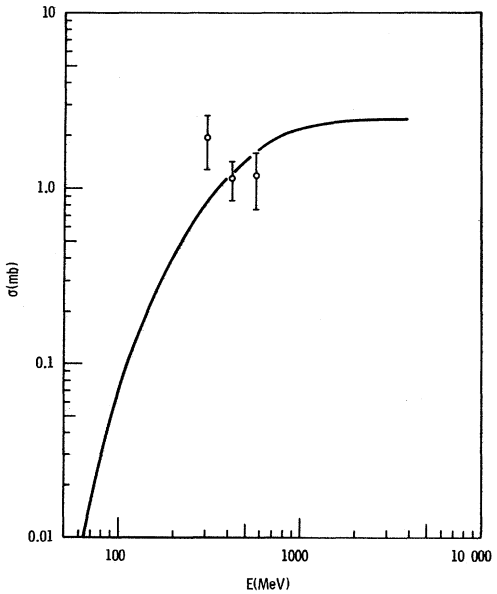

FIG. 3. Proton spallation cross sections of ^{27}Mg from argon.

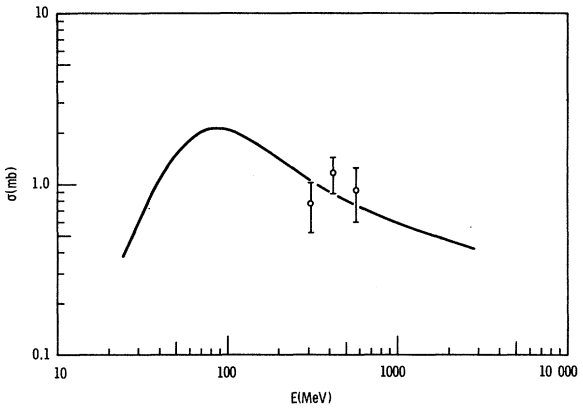

FIG. 4. Proton spallation cross sections of ^{37}S from argon.

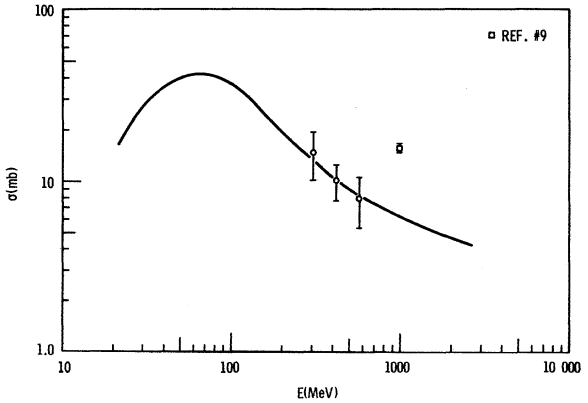

FIG. 5. Proton spallation cross sections of ^{38}Cl from argon.

The number of protons incident on each target was determined by the cyclotron staff from the integrated current generated in a proportional counter as the beam passed through. However, the number of incident protons calculated from the ^{24}Na activity induced in an exposed aluminum foil differed by approximately a factor of 2 from the proportional-counter data for two of the bombardments. This uncertainty, however, does not affect the ratios of cross sections.

From available information on the production rates and air concentrations of the cosmogenic radionuclides[7,8] and the increased μ^- production of ^{39}Cl and ^{38}Cl at low altitudes,[2,5] it has been estimated that in the atmospheric region between 10 000 ft and sea level at 46°N latitude, the atom production rates of ^{24}Na:^{38}Cl:^{39}Cl are in the ratio of 1:20:30.[8] Assuming that the shapes of the Rudstam excitation functions for the production of these radioisotopes from argon are fairly accurate, the production ratio of these three radioisotopes from proton spallation at 250 MeV is 1:22:28 based on the experimentally normalized excitation functions. This means that this production ratio in the atmosphere is possible from direct proton reactions if the average effective proton energy is 250 MeV, and that no muon reaction contributions are required. If the average effective proton energy is less than 250 MeV, any muon contribution would make these ratios much larger. However, if the average effective proton energy is greater than 250 MeV, a contribution from muon reactions would be necessary to achieve a ratio as large as 1:20:30.

Awschalom, Larsen, and Schimmerling[1] have reported several isotopes qualitatively and four isotopes quantitatively observed in argon gas placed in the vicinity of a synchrotron target. In their case, spallation arises from secondary particles generated by the 3-GeV primary proton beam striking a platinum or lead target. Similar results should be obtained by direct irradiation of argon with high-energy protons since high-energy neutrons and high-energy protons interact with a target nucleus in a very similar manner. Indeed, all the same radioisotopes were observed in our direct proton irradiation of liquid argon with the exception of ^{41}Ar. This radioisotope undoubtedly arises from the capture of thermalized secondary neutrons in the experiments of Awschalom, Larsen, and Schimmerling, while an insufficient number of thermal neutrons are available for production of detectable amounts of this radioisotope in our experiments.

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