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Thin-Target Cross Sections for Some Cr, Mn, Fe, Co, Ni, and Zn Nuclides Produced in Copper by 82- to 416-MeV Protons*

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Thin-target cross sections were determined by radiochemical means for the production of the radionuclides ^{65}Zn, ^{63}Ni, ^{56}Ni, ^{60}Co, ^{58}Co, ^{57}Co, ^{56}Co, ^{59}Fe, ^{55}Fe, ^{54}Mn, ^{52}Mn, and ^{51}Cr by 82-, 111-, 187-, 283-, and 416-MeV protons incident on natural copper. The experimental results are compared with values in the literature, expected trends in the excitation functions, and values calculated via empirical formulas and Monte Carlo treatments.

INTRODUCTION

Recent work on radionuclide production has concentrated on cross sections for reactions induced by GeV protons or heavy ions. A review of the literature, however, indicates many gaps and apparent discrepancies in existing data for reactions produced by lower energy protons. The present work was undertaken because of an interest in a number of problems that require cross-section data in the energy range 0.1 to 1 GeV. Such data are of use in the elucidation of the mechanisms of nuclear reactions and as input data in studies of the effects of internuclear cascades in thick targets (e.g., meteorites and the lunar surface). Therefore, cross sections for production of 12 nuclides (^{65}Zn, ^{63}Ni, ^{56}Ni, ^{60}Co, ^{58}Co, ^{57}Co, ^{56}Co, ^{59}Fe, ^{55}Fe, ^{54}Mn, ^{52}Mn, and ^{51}Cr) were determined for 82-, 111-, 187-, 283-, and 416-MeV protons incident on copper.

EXPERIMENTAL

Three series of bombardments were performed on thin (29.8-mg/cm^2), pure (>99.99%) copper metal foils using the internal beam of the University of Chicago synchrocyclotron. Separate irradiations by protons of three different energies were included in each series, with one run at 111 MeV being common to all as a check on reproducibility among the series. Beam monitoring was achieved by determining the production of ^{24}Na in two thin (7.28-mg/cm^2), pure (>99.9%) aluminum foils. Cross-section values of Cumming[1] for the production of ^{24}Na from proton bombardment of aluminum (10.1 mb at 82 MeV, 10.0 mb at 111 MeV, 9.3 mb at 187 MeV, 10.1 mb at 283 MeV, and 10.5 mb at 416 MeV) were used to calculate the total number of protons traversing the target. The proton energy values listed were derived from the nominal values (determined by target location) by applying corrections for the radial oscillations of the beam.[2] This phenomenon also causes a spread in the energy of the protons striking the target. The full widths of such proton energy dispersions are estimated to vary from about 10% at 416 MeV to about 20% at 82 MeV.[2]

The irradiated foils were subjected to radiochemical separation and purification procedures to yield various elements of interest. The chemical treatment and radioactivity measurement techniques were fairly standard.[3-5] Instruments were calibrated and sample decay was followed as per usual radiochemical practice.

The nature of the radiation measured for each nuclide, as well as branching ratios and half-lives assumed (generally taken from Lederer, Hollander, and Perlman[6]), are presented in Table I.

RESULTS AND DISCUSSION

Individual cross sections determined in this work are given in Table I, along with averages for replicate determinations at 111 and 187 MeV. These cross sections refer to copper of natural isotopic composition. No corrections for contributions from decay of short-lived precursors have been applied to these data.[7] The three separate irradiation series are indicated by letters G, D, and S. Sequential radioactivity measurements on a given sample have been appropriately combined in calculating the value given in the table. Only relative values have been reported for ^{63}Ni because of insufficient knowledge of the absolute detection efficiency for the low-energy radiation (0.067-MeV β^-) sought.

A standard deviation (1σ) of ±20% is assigned to each individual cross-section value. Such uncertainties are larger than the statistical uncertainties of the radioactivity measurements (typically ~1%) or the variation observed in sequential measurements on an individual (decaying) sample

TABLE I. Thin-target cross sections, in mb, for 82- to 416-MeV protons on natural copper.

Nuclide	Half-life	Radiation sought and branching ratio assumed	Bombardment series	Cross section[a]				
				82 ± 8 MeV	111 ± 10 MeV	187 ± 13 MeV	283 ± 17 MeV	416 ± 21 MeV
^{65}Zn	245 day	1.11 MeV γ[b]	*S*	2.6	1.7	0.9	...	...
			G	...	...	1.1	...	0.35
			D	...	1.6	...	0.52	...
					1.7 Ave.	1.0 Ave.		
^{63}Ni	92 yr	0.067 MeV β^- 100%	*S*	≡1.0[c]	1.0	1.2	...	...
^{56}Ni	6.1 day	1.56 MeV γ 15%	*S*	0.07	0.06	0.08	...	...
^{60}Co	5.26 yr	1.17 + 1.33 MeV γ 100% + 100%	*S*	15	11	...	...	...
			G	...	11	11	...	8
			D	...	12	...	10	...
					11 Ave.			
^{58}Co	71.3 day	0.811 MeV γ 100%	*S*	51	39	...	...	...
			G	...	37	38	...	20
			D	...	40	...	29	...
					39 Ave.			
^{57}Co	272 day[d]	0.123 MeV γ 89%	*S*	46	45	...	...	...
			G	...	41	38	...	24
			D	...	45	...	34	...
					44 Ave.			
^{56}Co	77.3 day	3.25 MeV γ 12.5%[e]	*S*	17	11	...	...	...
			G	...	12	9	...	11
			D	...	14	...	14	...
					12 Ave.			
^{59}Fe	45.0 day	1.10 + 1.29 MeV γ 54% + 44%	*S*	0.8	1.0	...	...	...
			G	...	1.0	1.2	...	1.4
			D	...	1.0	...	1.3	...
					1.0 Ave.			
^{55}Fe	2.7 yr	5.9 keV x ray[b]	*S*	11	11	...	...	...
			G	...	10	11	...	11
			D	...	11	...	11	...
					11 Ave.			
^{54}Mn	312 day	0.835 MeV γ[b]	*S*	3.7	10	15	...	...
			G	...	10	16	...	21
			D	...	11	...	19	...
					11 Ave.	16 Ave.		
^{52}Mn	5.7 day	1.43 MeV γ 100%	*S*	0.9	2.1	5.3	...	...
			G	...	2.0	5.1	...	8.9
			D	...	2.2	...	7.1	...
					2.1 Ave.	5.2 Ave.		
^{51}Cr	27.8 day	0.32 MeV γ 9.0%	*G*	...	...	...	...	18
			D	...	3.7	...	8.3	...

[a] Cross-section values have estimated standard deviations of ±20% (see text). No corrections to the observed values for contributions from isobaric precursors have been made [see text and note (Ref. 7)]. The proton energy values represent means, with all incident protons assumed to be within the limits indicated (see text).

[b] Standard samples prepared from National Bureau of Standards calibrated solutions were used in detection efficiency determinations for the nuclides so indicated.

[c] ^{63}Ni cross sections are relative to that at 82 MeV (see text).

[d] D. J. Goldman and J. R. Roesser, *Chart of the Nuclides* (General Electric Company, Schenectady, N.Y., 1966).

[e] H. L. Scott and D. M. Van Patter, Phys. Rev. 184, 1111 (1969).

(typically ~5%). However, these quoted uncertainties of ±20% reflect typical agreement among values obtained from replicate experiments, and an assessment of the uncertainties in the values for other parameters used in calculating cross sections from experimental data. In principle, a better measure of the true uncertainties could be obtained by a critical evaluation of the cross-section values (at nearly the same energies) reported by different workers. Such estimates well could exceed the ±20% quoted here (a value already much larger than those typically given in the literature).

A comparison with other work on proton-induced nuclear reactions with copper is provided in Fig. 1. Values from Table I and corresponding data from the literature[5,8-21] for 40-MeV to 30-GeV protons on copper are plotted. Wherever possible, literature values have been adjusted to conform with the values of Cumming[1] for the production cross section of ^{24}Na from aluminum. A yield of 83 mb for the production of ^{64}Cu from natural copper[20] was used to convert the relative 49-MeV values of Carleson.[9] For consistency, production cross-section values for ^{65}Zn have been adjusted to refer to natural copper (not ^{65}Cu).

For the majority of nuclides, yields determined

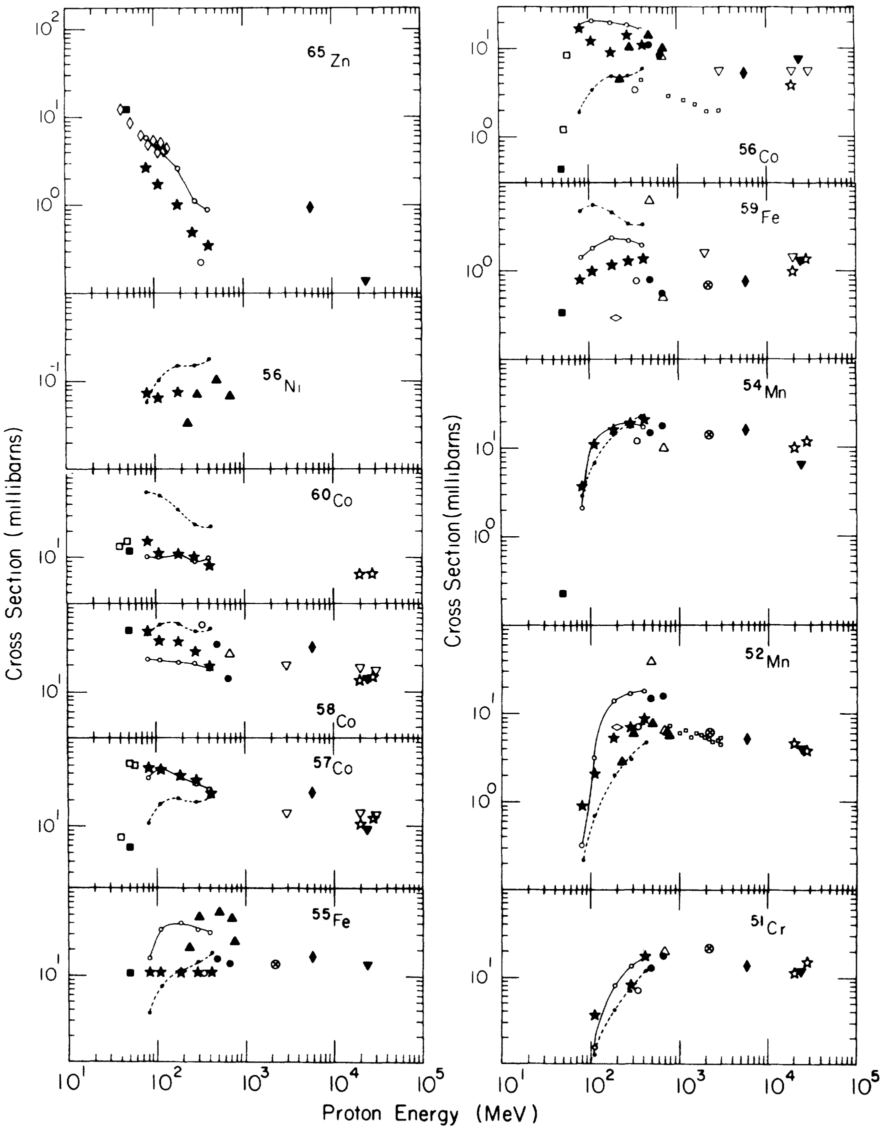

FIG. 1. Production cross sections for 40-MeV to 30-GeV protons on copper. Values are taken from: ★, this work; ◇, Ref. 5; □, Ref. 8; ■, Ref. 9; ◇, Ref. 10; ▲, Ref. 11; ○, Ref. 12; ●, Ref. 13; △, Ref. 14; ⊗, Ref. 15; ◆, Ref. 16; ▼, Ref. 17; ▽, Ref. 18; □, Ref. 19; -●-●-●-, calculated using Rudstam CDMD formula, Ref. 22; -○-○-○- , calculated using VEGAS-DFF treatment, Refs. 23 and 24; ☆, Ref. 21.

in this work are in fair agreement with yield patterns suggested by the bulk of existing data in the energy range 40 MeV to 30 GeV. For seven of the nuclides (^{60}Co, ^{58}Co, ^{57}Co, ^{56}Co, ^{55}Fe, ^{52}Mn, and ^{51}Cr), our values appear to lie (within the uncertainties) on the excitation curves which may be inferred from the major portion of the previous work. ^{56}Ni appears to be produced more abundantly at lower energies than might be inferred from the one previous study. The present results for ^{54}Mn and ^{59}Fe are systematically higher by about a factor of 2 than might be expected on the basis of the values in the literature, while our values for ^{65}Zn are systematically lower by about a factor of 2 on a similar basis.

The variations of cross sections with bombardment energy observed in this work (Table I and Fig. 1) are in qualitative agreement with well-established general features of nuclear reactions in this energy region. As the bombardment energy increases, cross sections for the production of nuclides considerably removed from the target nucleus ($\Delta A > 10$) increase sharply, due to greater typical nuclear excitation energies. On the other hand, yields for very "simple" reactions, in which the product is close to the target in mass number, decrease rapidly. There is also a broadening of the yield pattern about the most probable product curve as the projectile energy is increased. These qualitative features are exhibited in the data. Production cross sections for ^{54}Mn, ^{52}Mn, and ^{51}Cr increase by factors of from 6 to 11 as the proton bombardment energy increases from 82 to 416 MeV. Also, the isotopic cross-section ratio $\sigma(^{52}\text{Mn})/\sigma(^{54}\text{Mn})$ increases from about 0.2 to 0.4 over this energy region. However, the cross section for ^{65}Zn decreases by about an order of magnitude over the projectile energy range involved. As expected, the variation with energy of the yields of the remaining eight intermediate-mass nuclides in the table is moderate. These cross sections change by less than a factor of 2 as the irradiation energy increases from 82 to 416 MeV.

Two general approaches to estimating radionuclide production probabilities have been taken in the past, and it is of interest to compare predicted values obtained thereby with those reported in this work.

Rudstam[22] has derived several empirical formulas from which the cross sections for spallation products can be estimated. We have chosen to use Rudstam's Eq. (25) with ancillary Eqs. (15), (19), (28), and (29), in performing these calculations. The resultant cross sections for the nuclides of interest have been plotted in Fig. 1; a dotted line has been used to join these calculated values solely in order to distinguish them more clearly from the points representing experimental values. ^{65}Zn and ^{63}Ni were omitted due to their close proximity to the target nuclei, cases in which the treatment of Rudstam is not applicable.[22]

An alternative approach involves Monte Carlo simulations of the intranuclear cascade and evaporation processes with the aid of a computer, utilizing rather basic nuclear data as input. Several models have been applied, one of the most recent being VEGAS[23] (for the cascade step)-DFF[24] (for the evaporation step). The results of calculations using this model as performed by Friedlander and Harp at Brookhaven National Laboratory for incident proton energies of 82, 111, 187, 283, and 400 MeV with ^{63}Cu and ^{65}Cu as targets were made available to us. Results (using the STEPNO model[23]) were combined in the appropriate ratio to yield thin target cross sections for the production in a natural Cu target of all of the nuclides of interest in this work (as well as those for many other nuclides). These values are shown in Fig. 1, again with the individual data points joined by a solid curve merely for the sake of clarity. Results for ^{56}Ni are not presented due to the poor precision obtained from only 2000 cascades. For all VEGAS-DFF results plotted, the quoted standard deviation is at most ±50%; for most it is less than ±30%.

In general, agreement between our experimental values and the calculated values is only moderate, with the former usually falling between the two sets of calculated values. Both approaches give reasonable shapes for excitation functions of products furthest removed from the target, but a better fit over the whole range of products and projectile energies seems to be provided by VEGAS-DFF.

Although the sums of the experimental cross sections for all nuclides considered (except ^{65}Zn,

TABLE II. Comparison of observed and calculated cross sections.

Calculation method	Cases in category[a] $0.5 > Q_i$	$0.5 \le Q_i < 1.0$	$1.0 \le Q_i \le 2.0$	$2.0 < Q_i$
Rudstam	11	7	14	11
VEGAS-DFF	7	18	15	3

[a] $Q_i \equiv \sigma_{\text{exp}}/\sigma_{\text{calc}}$.

^{63}Ni, and ^{56}Ni) at any given energy are in good agreement with the results of both calculations (generally within 20%), certain distinctions between the typical results obtained from the two methods may be drawn. There is a tendency for the Rudstam approach to give individual cross section estimates which are lower than both our experimental values[25] and those from VEGAS-DFF. VEGAS-DFF, on the other hand, just as frequently provides values which are higher than our values[26]; the relative discrepancies are, generally, not as large, however, as with the Rudstam formulas.

A more quantitative indication of the agreement between our results and those obtained using the two calculation techniques may be provided by examination of the ratio: $(\sigma_{\text{exp}}/\sigma_{\text{calc}}) \equiv Q_i$. Table II provides a crude indication of the distribution of Q_i values from both approaches.

Although the Rudstam-type approach gives a particularly large relative discrepancy for ^{56}Co at 82 MeV, it might be noted that both calculational approaches have difficulty in fitting the experimental values at the lower projectile energies, especially for the lower-A products (where the cross section may be a steeply changing function of projectile energy). The larger relative uncertainties in our experimental projectile energies at the lower values could, through some undetected systematic inaccuracies in these values, reduce the discrepancies somewhat, but do not seem likely to eliminate completely (for either calculational approach) the dispersion encountered throughout the entire product-mass-energy spectrum studied here. Nevertheless, it is clear (both from Fig. 1 and from Table II) that the VEGAS-DFF calculations fit the present data better than the Rudstam approach.[27] This is interesting, since Rudstam's formula is based on a fit to experimental data while VEGAS-DFF utilizes average values of more fundamental nuclear properties.

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[1] J. B. Cumming, Ann. Rev. Nucl. Sci. 13, 261 (1963).

[2] R. C. Koch, Ph.D. thesis, University of Chicago, 1955 (unpublished).

[3] A. Van Ginneken, Ph.D. thesis, University of Chicago, 1966 (unpublished).

[4] H. R. Heydegger, Ph.D. thesis, University of Chicago, 1968 (unpublished).

[5] C. K. Garrett, Ph.D. thesis, University of Chicago, 1970 (unpublished).

[6] C. M. Lederer, J. M. Hollander, and I. Perlman, *Table of Isotopes* (Wiley, New York, 1967), 6th ed.

[7] Such corrections could be made, for example, by calculating the relevant isotopic cross-section ratios by use of the equation proposed by Rudstam (1966) (see text) and, on this basis, would be: ^{57}Co, 5.6%; ^{56}Co, 1.5%; ^{59}Fe, 2.1%; ^{55}Fe, 6.3%; ^{52}Mn, 3.6%; ^{51}Cr, 8.5%. VEGAS-DFF (1971) (see text) predicts contributions at similar, though energy-dependent, levels except for ^{51}Cr, where the effect ranges from 13 to 23% between 82 and 400 MeV.

[8] I. R. Williams and C. B. Fulmer, Phys. Rev. 162, 1055 (1967).

[9] G. Carleson, Acta Chem. Scand. 8, 1697 (1954).

[10] G. Albouy, M. Gusakow, N. Poffee, H. Sergolle, and L. Valentin, J. Phys. Radium 23, 1000 (1962).

[11] J. Warren and W. Forster, University of California at Davis, Report No. UCD-34P75-1, 1966 (unpublished).

[12] R. E. Batzel, D. R. Miller, and G. T. Seaborg, Phys. Rev. 84, 671 (1951).

[13] A. N. Murin, B. K. Preobrazhensky, I. A. Yutlandov, and M. A. Yakimov, in *Proceedings of the Conference of the Academy of Sciences of the U.S.S.R. on the Peaceful Use of Atomic Energy, Moscow, July, 1955* (Akad. Nauk. S.S.S.R., Moscow, 1955) [translation by Consultants Bureau, New York: U. S. Atomic Energy Commission Report No. TR-2435, 1956 (unpublished), p. 160].

[14] A. P. Vinogradov, I. P. Alimarin, V. I. Baranow, A. K. Lavrukhina, T. V. Baranova, and F. I. Pavlotskaya, in *Proceedings of the Conference of the Academy of Sciences of the U.S.S.R. on the Peaceful Use of Atomic Energy, Moscow, July, 1955* (see Ref. 13), p. 132.

[15] G. Friedlander, J. M. Miller, R. Wolfgang, J. Hudis, and E. Baker, Phys. Rev. 94, 727 (1954).

[16] D. W. Barr, University of California Radiation Laboratory Report No. UCRL-3793, 1957 (unpublished).

[17] G. Rudstam, E. Brunnix, and A. C. Pappas, Phys. Rev. 126, 1852 (1962).

[18] J. Hudis, I. Dostrovsky, G. Friedlander, J. R. Grover, N. T. Porile, L. P. Remsberg, R. W. Stoenner, and S. Tanaka, Phys. Rev. 129, 434 (1963).

[19] J. A. Miskel, M. L. Perlman, G. Friedlander, and J. M. Miller, Phys. Rev. 98, A1197 (1955); G. Friedland-

er, private communication.

[20] J. W. Meadows, Phys. Rev. 91, 885 (1953).

[21] J. Hudis, CERN NP DIV. Internal Report No. 68-22, 1968 (unpublished); private communication.

[22] G. Rudstam, Z. Naturforsch, 21a, 1027 (1966).

[23] K. Chen, Z. Fraenkel, G. Friedlander, J. R. Grover, J. M. Miller, and Y. Shimamoto, Phys. Rev. 166, 949 (1968).

[24] I. Dostrovsky, Z. Fraenkel, and G. Friedlander, Phys. Rev. 116, 683 (1960).

[25] Such an observation is not inconsistent with the results reported by Rudstam (1966) for Cu as target at similar projectile energies. However, Brodzinski *et al.* [Phys. Rev. C 4, 1250, 1257 (1971)] recently reported that the Rudstam approach provided systematically higher cross-section estimates than their own experimental values for various products in Ti and Fe targets irradiated with protons of comparable energy to those employed in this work.

[26] Although experimental values should be systematically higher than those predicted by the calculations due to contributions to the former from short-lived precursors, such effects are of insufficient magnitude to be important here.

[27] Since the range of products studied in this work is confined to those rather close to stability, the relative performances of the two calculational techniques for other products (or projectile energies or targets) can not, necessarily, be inferred from those encountered here.

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Variation of the ^{32}S Compound-Nucleus Width with Energy and Spin*

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Coherence widths of the compound nucleus ^{32}S were determined from excitation functions of the differential cross section for the reactions ^{31}P(p, α)^{28}Si and ^{16}O(^{16}O, α)^{28}Si. The proton-induced reaction provided a lower average spin of the compound nucleus. Excitation functions were measured for ^{31}P(p, α)^{28}Si from 13.90- to 14.24- and from 26.62- to 30.56-MeV compound-nucleus energy. The average coherence widths were 11 ± 1.2 and 95 ± 15 keV, respectively. For the oxygen-induced reaction, excitation functions were measured from 28.99 to 34.42 MeV with a resulting 73 ± 7-keV coherence width, which is nearly the same as for the lower-spin proton-induced case.

These coherence widths were used to test the Gilbert and Cameron level-density formulation. Good agreement with the above data was obtained if the compound nucleus ^{32}S is considered to be spherical in this formulation. The increase in the calculated width for the ^{16}O induced reaction with increasing excitation energy is greater than indicated by combining our data with another measurement at higher excitation.

I. INTRODUCTION

Information on the level densities $\rho(E, J)$ of nuclei as a function of excitation energy E and spin J has several uses. Gadioli and Zetta[1] note that measurements of level widths are the main method of determining densities for $E \gtrsim 6$ MeV, while for lower energies more direct and quantitative methods are available. The present work is an extensive investigation of the compound nucleus ^{32}S at excitation energies from 14 up to 34 MeV. The span of spin conditions in the reactions studied and the span of energies covered combine to make this a particularly rich collection of data on which to test energy and spin dependence of level-density expressions.

Experimentally, the present studies were measurements of excitation functions made in sufficiently small steps of incident energy and with sufficiently good resolution to permit fluctuation analyses.[2] Use of the compound nucleus ^{32}S provided several advantages:

(1) The minimum spin $\frac{1}{2}$ of both the proton and the target nucleus ^{31}P allow use of the convenient reaction ^{31}P(p, α)^{28}Si as the low-spin case for the study of the intermediate nucleus ^{32}S.

(2) The nucleus ^{32}S has a sufficiently large mass number that levels have the necessary overlap[3] at 14-MeV excitation energy, but has a sufficiently small mass number that the cross section for ^{16}O(^{16}O, α)^{28}Si at 34-MeV excitation energy is adequate to allow fluctuation measurements. This allows a large span of energies.

(3) Other fluctuation measurements have been reported for the compound nucleus ^{32}S. Measurements have been made of the ^{31}P(p, α)^{28}Si reaction at intermediate energies,[4, 5] the ^{16}O(^{16}O, α)^{28}Si reaction and ^{16}O-^{16}O scattering at a slightly higher