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ABSTRACT

A semiempirical four-parameter formula, following the formalism of Gupta, is proposed in order to systematise spallation yields. A preliminary test made by comparing calculated and experimentally determined cross sections for 2-GeV bremsstrahlung-induced spallation in natural copper gave very encouraging results (a coefficient of reproducibility R = 1.7 or better). The formula will be used for an exhaustive study of intermediate- and high--energy photospallation of medium-weight nuclei.

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Knowledge of cross sections for intermediate- and high-energy nuclear reactions is essential in several branches of science such as geochemistry, cosmochemistry, lunar and meteoritic studies, and astrophysics.

Basically, two different methods of computation provide estimates of the cross sections not available experimentally. Monte Carlo calculation on one hand, which are based on the Serber([1]) theoretical model of interaction (i.e. a fast intra-nuclear cascade followed by a much slower particle evaporation), and semiempirical multiparameter formulae, on the other hand. The latter method is in general more widely used, due to its greater computational speed.

A number of semiempirical formulae([2-19]) are available that reproduce, with a more or less high degree of accuracy, the cross sections of formation of products arising from spallation or other reactions. As far as spallation is concerned, the formula which up now gives the best results and has therefore been used over a wider range of both target nuclear masses and incident energies is the well known five-parameter charge-distribution-mass--distribution (CDMD) Rudstam formula([3]). This formula bases on the observed trend of spallation yields to decrease exponentially with increasing both the (nominal) number of nucleons ejected from the struck nucleus and the distance between the atomic numbers of the produced nuclide (spallation product) and target nucleus. Some correspondence is found between the Serber model and the CDMD formula (mainly for the evaporation step), although the latter furnishes a phenomenological description of spallation only, without giving deeper information about the physics involved in the process.

Quite a different approach has been proposed by Gupta *et al.*([7]). They treat, in fact, the interaction in a purely statistical way by considering the target nucleus as a collection of A_t non-interacting particles consisting of a uniform mixture of two types of populations, Z_t protons and N_t neutrons. The phenomenon of spallation is thus regarded as the successive emission of nucleons from the nucleus, no distinction being made between cascade and evaporation steps. The physical reality enters in their treatment by taking into consideration that the number of protons expected to be emitted when a given number of neutrons are emitted from the target is dependent on the proton-neutron ratio in the target and, what is still more important, on the number of low-energy protons to be suppressed due to the Coulomb barrier. The great advantage of this formalism (henceforth abridged as G-formalism, G-formula, etc.) lies in the fact that only three "free" parameters are needed for to obtain theoretical distributions of spallation yields when a given number of neutrons leave the nucleus.

In the next section we shall try to extend the G-treatment, originally proposed for high-energy incident protons, to photon- and bremsstrahlung- induced spallation, and, at the same time, we shall introduce some modifications in the formula in order to attain distributions of yields for any Z and N values of the nuclides photoproduced on the same target nucleus.

PHOTOSPALLATION AND THE FOUR-PARAMETER FORMULA

A photospallation reaction that leads to a product nucleus P from a target nucleus T through a nominal nucleon loss of x protons and y neutrons is written as

$$ {}^{A_t}_{Z_t}T_{N_t} \ (\gamma,\ xp\ yn)\ {}^{A_t-(x+y)}_{Z_t-x}P_{N_t-y} \quad . $$

We consider as <u>true</u> spallation reactions only those for which

$$ x \geq 1 \quad , $$
$$ y \geq 1 \quad . $$

Furthermore, to avoid complication due to the occurrence of other reactions such as fission and/or fragmentation, at least at the higher energies[20-23], we shall confine ourselves to those events only for which the total number of emitted nucleons $(x + y)$ is not greater than $A_t/2$ (in other words: $x \lesssim Z_t/2$, $y \lesssim N_t/2$).

Following the G-formalism, for a given number of emitted neutrons, say y_i, a true gaussian-shaped distribution of nuclide yields (yields of isotones), centered at x_i (mean number of emitted protons) is obtained as

$$ \sigma_i(x,y_i) = H_i \ \exp\left[-k_i(x-x_i)^2\right] \ , \qquad 1 $$

where k_i gives the width of the distribution and H_i is the normalising factor ($\sigma_i = H_i$ when $x = x_i$). The law of regression of x_i on y_i accounts for the ratio $\alpha = Z_t/N_t$ of the number of protons to the number of neutrons of the target nucleus and for proton suppression due to the Coulomb barrier. The regression line is written as

$$ x_i = \alpha y_i - \alpha y_i \ \exp\left[-D/\alpha y_i\right] \ , \quad D > 0 \ . \qquad 2 $$

In eq. 2, the first term of the right hand side represents the number of protons expected to be emitted if there were no suppression, and the second term the number of suppressed protons. Eq. 2 follows the asymptotic behaviour

$$x_i \rightarrow 0 \quad \text{as} \quad y_i \rightarrow 0$$

and

$$x_i \rightarrow D \quad \text{as} \quad y_i \rightarrow \infty .$$

Also, it can be proved that $x_i \simeq \alpha$ when $y_i = 1$ and $x_i = (Z_t/2)[1-\exp(-D/Z_t/2)]$ when $y_i = N_t/2$.

Suppose now y_i varies from 1 up to about $N_t/2$. A number of distributions of type 1 will be obtained with the same width (as arising from the same physical process) but different normalising factors H_i ($i = 1,2,\ldots$). We assumed the following dependence of H_i upon the variable x_i to be valid

$$H_i = \sigma_M \exp\left[-B(x_i-1)\right] \quad , \; B > 0 \qquad 3$$

and, consequently, a more general mass-yield distribution is obtained

$$\sigma(x,y) = \sigma_M \exp\left[-B(x-1)\right] \exp\left[-k\{x-\alpha y[1-\exp(-D/\alpha y)]\}^2\right], \qquad 4$$

which may be written with a more compact formalism by setting

$$C = 1 - \exp\,(-D/\alpha y) \qquad 5$$

and

$$w = x - C\alpha y \quad , \qquad 6$$

thus obtaining

$$\sigma(x,y) = \sigma_M \exp\left[-B(x-1)\right] \exp\left[-kw^2\right] \quad . \qquad 7$$

Equation 7 allows one to calculate the cross section of any spallation product for a given target nucleus at any incident photon (or bremsstrahlung) energy. Apart from those quantities directly correlated with the target characteristics (α) and type of reaction (x and y), it contains only the four free parameters σ_M, k, D (or C), and B; σ_M being the cross section of formation of the nuclide with $Z = Z_t-1$ and $N = N_t-1/\alpha C$ (i.e. $x = 1$, $y = \frac{1}{\alpha C} \simeq 1$).

We should have tested the validity of this formula by comparing its numerical results with a number of available experimentally determined yields. As a matter of fact, however, this paper is only a preliminary one, and we shall make a comparison with the measured cross sections reported by Bachschi *et al.*([24]) for 2-GeV bremsstrahlung irradiation of natural copper. We wish to point out that this particular choice is not a casual one. The experimental data by Bachschi *et al.* have been obtained at a bremsstrahlung energy E_0 = 2 GeV which is sufficiently high to reduce the background tail from low--energy processes (mainly for those produced nuclides very close to the target mass) and cover a wide range of <u>true</u> spallation products (from $Z = Z_t-1$ up to $Z = Z_t-11$, see Table 1). Moreover, they have already been compared, quite satisfactorily indeed, with the pattern for spallation reactions as predicted by the CDMD five-parameter Rudstam formula, modified by Jonsson and Lindgren([13,17]) for bremsstrahlung-induced-spallation. Finally, copper is a very representative medium-weight element (both the CDMD formula and the modified G-formula are expected to fail for either very light or very heavy target nuclei([4,10])).

COMPARISON WITH EXPERIMENTAL RESULTS AND DISCUSSION

Natural copper consists of the two isotopes $^{63}_{29}Cu_{34}$ (69.1% isotopic abundance) and $^{65}_{29}Cu_{36}$ (30.9%). For the sake of simplicity we considered, in the course of the calculation, a target of ^{63}Cu (α = 0.853). This assumption simplifies a great deal the numerical computations and influences the final results only to a very low extent.

A least-squares analysis (with no data rejection) of the measured cross sections per equivalent quantum σ_Q reported in ref. 24 has been carried out to obtain the best values of the parameters σ_M, k, D, and B. The following set was obtained: $\sigma_M = 4.0 \times 10^3$ μb/*equivalent quantum*; k = 0.46; D = 25; B $\doteq$ 0.41. The statistical uncertainty that affects each parameter does not exceed 10% (for σ_M and B, it was less than 5%).

By using eq. 7 with the numerical set found for the parameters, we calculated the theoretical distribution and plotted the quantity $\varphi_Q(x,y) \times \exp[B(x-1)]$ as a function of w (this variable is defined by eq. 6) in a semilog graph. Figure 1 shows the trend of the calculated distribution. Plotted in the figure are also the experimental points to allow a comparison. As it could have been expected, the calculated trend fits the experimental points very closely

We also used the modified CDMD formula([17]) to calculate values of σ_Q for the radionuclides listed in Table 1. This formula gives the cross section for a product nucleus (A,Z) from a target (A_t,Z_t) and is written as

$$\sigma(A,Z) = \frac{\hat{\sigma} P d'^{2/3} A_t^{-2e'/3} \exp\left[PA - R|Z - SA + TA^2|^{3/2}\right]}{1.79\left[e^{PA_t}\left(1 - \frac{2}{3PA_t}\right) - 1 + \frac{2e'}{3} + \frac{2e'}{3PA_t}\right]} , \quad 8$$

with the following relations for the different parameters

$\hat{\sigma} = (-0.81+0.184\, \ln E_o)A_t^{1.13}\times 10^3$ μb/*eq.quantum*, E_o in MeV,

$P = 7.66\, A_t^{-0.89}$, $R = d'A^{-e'}$,

$S = 0.486$, $T = 0.00038$,

$d' = 11.8$, $e' = 0.45$.

Table 1 reports the ratios σ_e/σ_c between experimentally determined and calculated values of σ_Q for both eq. 7 and eq. 8. These results are also shown in Figure 2 in the form of histograms (in the case of frequency distributions of ratios it turns out to be more convenient the use of a logarithmic scale for the abscissa). It is readily seen that the frequency distributions compare quite favourably with each other, both being centered around unity ($\sigma_e = \sigma_c$). At a first glance, the distribution obtained from eq. 7 seems to be somewhat broader than that from eq. 8, but one must also consider that the first(a) clearly exhibits a higher density around unity and is almost gaussian-shaped, with the exception of a few points at the wings, whereas the second one (b) is rather flat. The following quantitative deductions can be drawn by comparing the two frequency distributions: for eq. 7 (graph a), 41% of the experimental points are reproduced within a factor 1.2, 64% within a factor 1.5, 82% within a factor 2, 91% within a factor 2.5, and 95% within a factor 3; for eq. 8 the corresponding percentages are 41%, 59%, 82%, 91%,and 100%, respectively.

The goodness of least-squares fitting procedures of the type described may be well represented by the coefficient of reproducibility R which is given by $R = e^{\varepsilon}$, with $\varepsilon = \left[\sum_i (\ln \frac{\sigma_{ei}}{\sigma_{ci}})^2/n\right]^{1/2}$. The values of R arising from eq. 7 and eq. 8 are, respectively, 1.67 and 1.61, but, disregarding the point relative to ^{43}Sc solely, eq. 7 gives R = 1.58. This means that 95% of the experimental points are reproduced within a factor $R^2 \approx 2.6$ by both distributions.

It is of some interest to compare the values of the most probable mass number $A_p(Z)$ which gives, for each isotopic distribution of yields, the maximum of the calculated values of σ_Q. A_p values have been calculated, for different Z's, by using eq. 7, eq. 8 and two other semiempirical formulae(8,9,19), and are reported in Table 2. In this case too, the modified G-formula gives very reliable values that are, in general, better than those obtained from eq. 8.

CONCLUSIONS

From inspection of Figures 1 and 2 and of Table 2, we can conclude that the modified G-formula seems to be very suitable in reproducing measured yields of photospallation and in predicting those not yet available from experiment. Of course, we are aware of the fact that a comparison based on one set only of experimental data may be not sufficient to ensure the applicability of the formalism adopted to the entire region of medium-weight and heavy nuclei.

Very preliminary calculations for photospallation of vanadium, manganese, iron, and cobalt at energies between 0.1 GeV and 1 GeV([25]) are giving, however, results which are very encouraging and we believe that a study along these lines will contribute to a better systematics of spallation and to a deeper understanding of this phenomenon.

As it has already been said at the very baginning of this paper, the aim of the present work was to prove that a semiempirical formula with a reduced number of parameters and a very simple analytic expression could have been capable of giving results at least comparable with those of other formulae. Some emphasis should be given to the fact that a reduced number of parameters surely will result in a remarkable reduction of computer time. We wish also put stress on being the four-parameter formula much more than a merely phenomenological one, as it accounts for most of the physical processes involved with spallation, not only from a simple statistical point of view. Finally, some words must be spent on the versatility of eq. 7. From it, in fact, yield distributions either isotopic or isobaric can easily be obtained by choosing suitable values of the two variables x and y.

A much more sophisticated computer program based on a multiple regression analysis, with readjustment of parameter values and data rejection, is presently being carried on in our laboratory. It will consider all existing data on photospallation. The results will be the argument of a further communication.

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Table 1.- Ratios σ_e/σ_c of experimental to calculated cross sections. The calculated values are those obtained from eq. 7 and eq. 8. Experimental values taken from ref. 24.

Product nucleus	x	y	σ_e/σ_c (eq. 7)	σ_e/σ_c (eq. 8)	Product nucleus	x	y	σ_e/σ_c (eq. 7)	σ_e/σ_c (eq. 8)
^{57}Ni	1	5	1.09	0.39	^{49}Cr	5	9	0.81	0.68
^{58}Co	2	3	0.76	0.46	^{48}Cr	5	10	1.04	0.95
^{57}Co	2	4	1.36	0.92	^{48}V	6	9	1.12	0.94
^{56}Co	2	5	1.26	0.76	^{48}Sc	8	7	0.58	1.04
^{55}Co	2	6	2.08	0.84	^{47}Sc	8	8	0.96	1.57
^{59}Fe	3	1	0.47	0.40	^{46}Sc	8	9	0.87	0.97
^{52}Fe	3	8	2.20	0.70	$^{44}Sc^{(g+m)}$	8	11	1.14	1.16
^{56}Mn	4	3	0.37	0.44	^{43}Sc	8	12	0.30	0.64
^{54}Mn	4	5	0.93	0.62	^{43}K	10	10	1.37	1.90
$^{52}Mn^{(g+m)}$	4	7	1.36	0.91	^{42}K	10	11	1.14	1.20
^{51}Cr	5	7	2.02	1.26	^{41}Ar	11	11	1.77	1.83

Table 2. - A_p values obtained from different spallation formulae.

Z	Ref.8 (a)	Ref.19 (b)	Ref.17 (c)	Present work (d)	Mass number(s) of the naturally occurring stable isotope(s)
28	58.3	61.4	60.5	60.8	58(67.76%),60(26.16) 61(1.25),62(3.66),64(1.16)
27	56.2	59.1	58.2	58.6	59(100)
26	54.1	56.8	56.0	56.5	54(5.84), 56(91.68), 57(2.17), 58(0.31)
25	52.0	54.5	53.7	54.3	55(100)
24	50.0	52.3	51.5	52.0	52(83,76), 53(9.55) 54(2.38)
23	47.9	50.0	49.2	49.7	50(0.25),51(99.75)
21	43.7	45.5	44.8	45.0	45(100)
19	39.6	40.9	40.4	40.5	39(93.22),40(0.118),41(6.77)
18	37.5	38.7	38.2	38.1	36(0.337),38(0.063),40(99.600)

(a) $A_p = SZ+TZ^2$, with S = 2.09 and T = 0.00033.

(b) $A_p = 2.27\,Z - 2.20$.

(c) Calculated from $TA_p^2 - SA_p + Z = 0$, with S = 0.486 and T = 0.00038.

(d) Calculated from $A_p = Z_t - x + N_t - \frac{x}{\alpha C}$, with $C = 1-\exp[-D/\alpha y]$ (see text).

FIGURE CAPTIONS

Fig. 1 - Plot of the products $\sigma_Q \times \exp[B(x-1)]$ *vs.* w. The curve is the best parabola obtained from the least-squares analysis of the experimental cross sections. Experimental points: open square, ^{57}Ni; open circles, $^{55,56,57,58}Co$; open triangles, $^{52,59}Fe$; reversed open triangles $^{52(m+g),54,56}Mn$; filled circles, $^{48,49,51}Cr$; filled square, ^{48}V; filled triangles, $^{43,44(m+g),46,47,48}Sc$; reversed filled triangles, $^{42,43}K$; rhomb, ^{41}Ar. The full width at half maximum (FWHM) of the calculated curve is $\Gamma \simeq 2.46$ w units; its full width at the inflection points (the gaussian-shaped curve becomes a parabola in a semilog graph) is $\Gamma_{in} = (2/k)^{1/2} \simeq 2.09$ w units.

Fig. 2 - Frequency distributions of the ratios σ_e/σ_c of experimental to calculated cross sections. Graph a: calculated values from eq. 7. Graph b: calculated values from eq. 8.

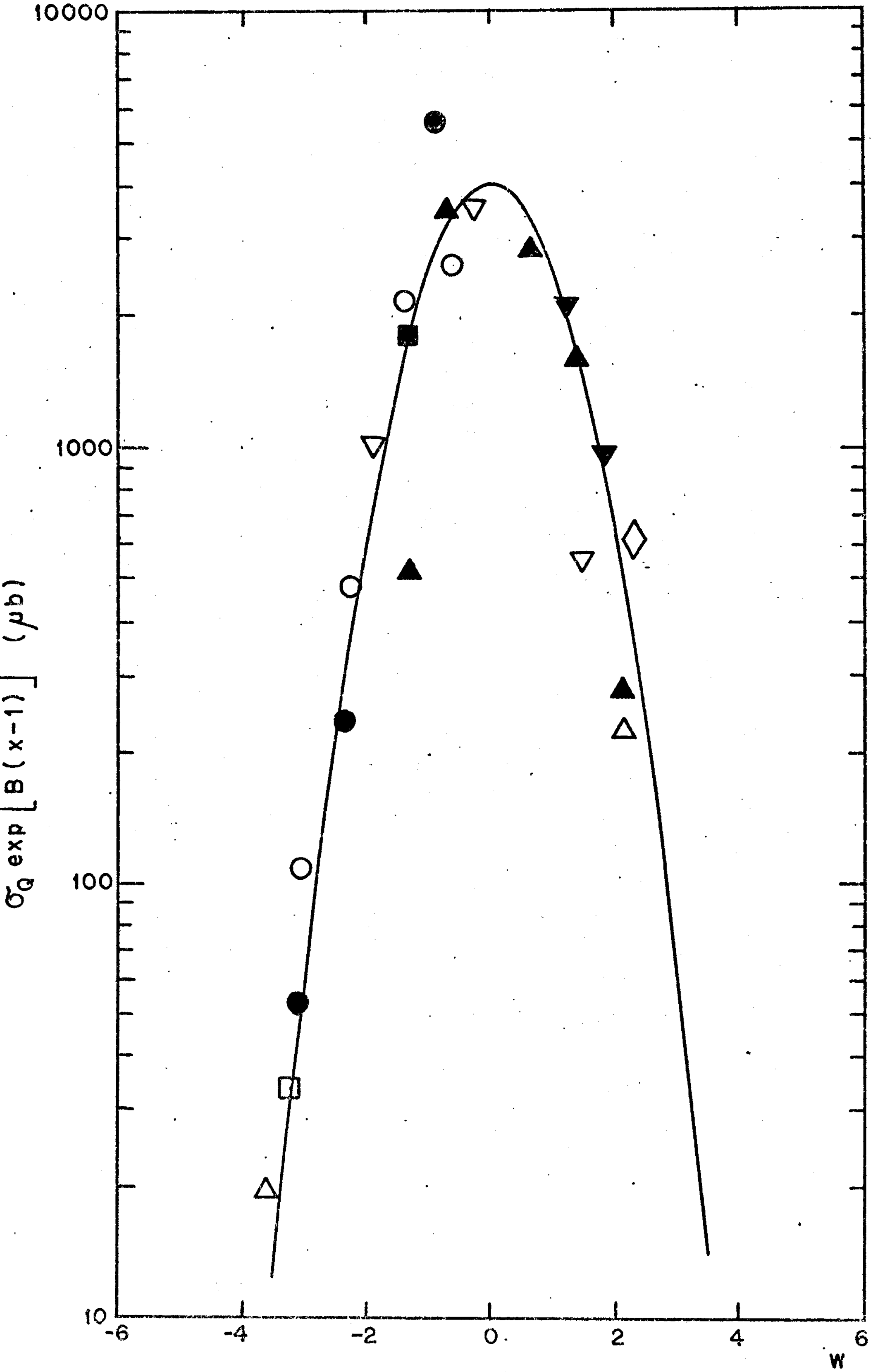

Figure 1

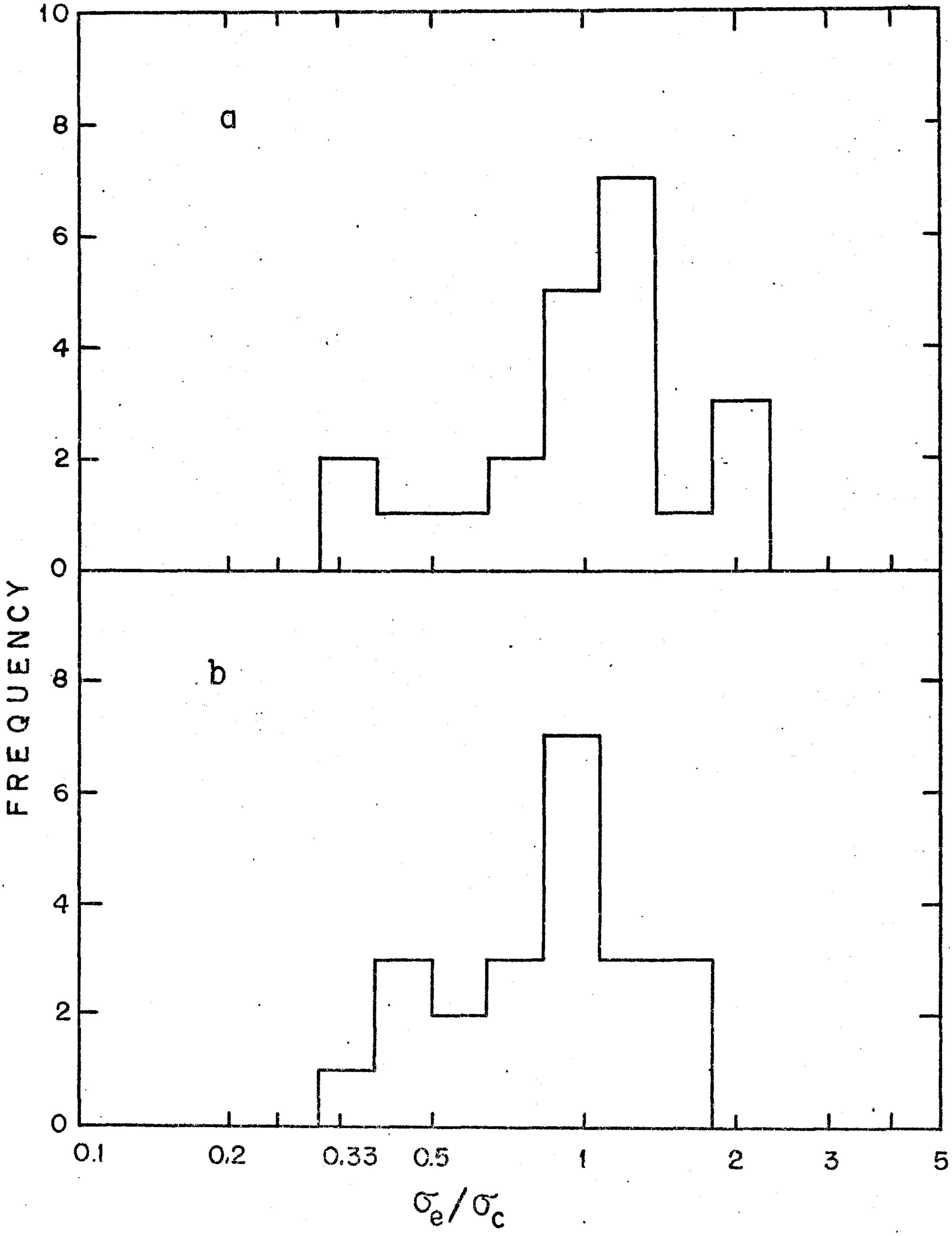

Figure 2