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Comments on the capture cross-section of electrons in silver bromide grains

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Abstract. The values in the range of 10^{-15} cm^2 to 10^{-14} cm^2 have been reported by various authors for the capture cross-section of electrons at the traps in silver bromide grains. In this paper we have given a suitable value of this cross-section which is used for computing the track characteristics in nuclear emulsions (e.g. effective ionization, probability of development, grain density and mean gap length) and the ratio of the rate constant of recombination and trapping. Theoretical results agree well with the experimental observations and the available data of other workers and give a convincing support to our choice of this cross-section parameter.

Keywords. Capture cross-section of electrons; nuclear emulsions; silver bromide grains.

1. Introduction

The process by which a latent image is formed in silver bromide grains in nuclear emulsions is related to the absorption of energy by the grains due to the passage of charged particles through them releasing electron-hole pairs. The electrons migrate through the crystal lattice until they are trapped at the electron traps and get neutralized there by the arrival of interstitial Ag^+ ions. The process of production of electron-hole pairs is a reversible one, and hence it is probable that the electrons and holes may recombine immediately after their production to form halide ions from which they were originated. The rate constant of recombination can be determined from the capture cross-section and the thermal velocities of electrons (Rose 1951 and Seitz 1951). The normal observed value for electron capture cross-section reported is of the order of 10^{-15} cm^2 for semiconductors (Bube 1960 and Rose 1963). Seitz (1951) has reported the value of cross-section for the capture of an electron at the trap $\sim 3 \times 10^{-15}$ cm^2. According to Bayer and Hamilton (1965) and Hamilton and Bayer (1965) the value of capture cross-section of electron by a recombination centre is about 25 times less than that offered by a silver ion kink site. In fact the exact value of the capture cross-section of an electron by recombination centres is not known.

In this paper we have tried to make a suitable choice of this parameter, ' capture cross-section of electron ', and have given a justification regarding its magnitude by deriving the theoretical relationships of track characteristics (effective ionization, probability of development, grain density and mean gap length) and the ratio of rate constant of recombination and trapping on the basis of this parameter and comparing the results with the available experimental data.

2. Theory

2.1. *Track characteristics*

Any ionizing particle while passing through an emulsion grain (AgBr) loses energy and ejects electron-hole pairs from its normal state in a Br^- ion. The holes so formed can be trapped very rapidly at the hole traps forming a recombination centre. The recombination of free holes with free electrons in emulsions is not possible but there is a possibility of recombination of free electrons and trapped holes at the recombination centre. The possibility of recombination of free hole and electron at the electron trap is also negligible. (see figure 1).

The rate of loss of electrons due to the recombination process will be governed by the following relation:

$$\frac{dn}{dt} = (\sigma_r V_e V_g^{-1})\, n_e n_h \tag{1}$$

where $\sigma_r V_e V_g^{-1}$ defines the rate constant of recombination (σ_r being the capture cross-section of electron by recombination centre; V_e, the thermal velocity of electrons and V_g, the volume of an emulsion grain). Also n_e and n_h represent the concentration of electrons and holes respectively at any instant t.

As the number of electrons is initially equal to the number of holes i.e., $n_e = n_h = n$ (say), the equation of continuity is given by:

$$\frac{dn}{dt} = (\sigma_r V_e V_g^{-1})\, n^2 \tag{2}$$

Integrating this equation and applying the boundary condition, $t=0$, $n=n_0$, we get:

$$n = \frac{n_0}{1+(\sigma_r V_e V_g^{-1}) n_0 t} \tag{3}$$

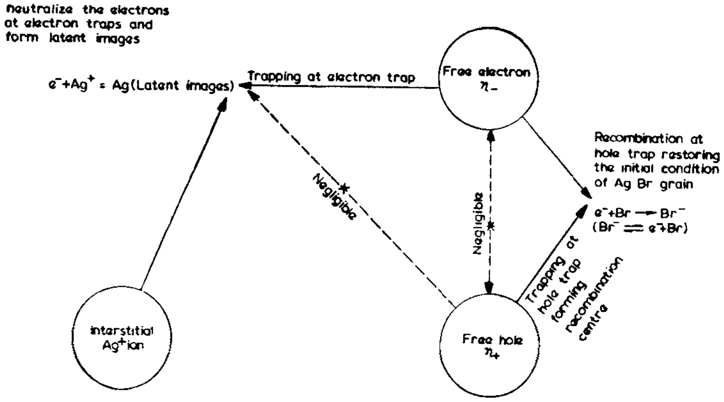

Figure 1. Diagram of the model of trapping of ions and possibility of their recombinations.

The value of n_0 (number of electrons produced initially due to the passage of charged particles in a grain) can be calculated from the relation,

$$n_0 = 46{\cdot}55\ (\mathrm{d}E/\mathrm{d}R)$$

where $\mathrm{d}E/\mathrm{d}R$ is the rate of energy loss in eV/μm (Sharma and Gaur 1968).

Substituting this value of n_0 in eq. (3), we get:

$$n = \frac{46{\cdot}55\ (\mathrm{d}E/\mathrm{d}R)}{1+46{\cdot}55\ \sigma_r V_e V_g^{-1}\, t\ (\mathrm{d}E/\mathrm{d}R)} \tag{4}$$

The probability of development (π) of a grain can be represented by the following relation (Sharma and Gill 1962):

$$\pi = 1-\exp[-(\theta/g_0\phi)\exp(-\theta)(1-\phi^{-1})\{1+(\theta-\phi)(1-\phi^{-1})+\ldots\}] \tag{5}$$

where $\theta = n/S$, $\phi = B/S$.

Here S is the average number of sensitivity centres in a grain, B is the limiting value of effective ionization at an energy loss of several thousand keV/μm, n is the effective number of electrons (given by equation 4).

Substituting the values of θ, ϕ and n in eq. (5) and solving it for the first approximation, we get:

$$g_0 B(B/S-1)\log(1-\pi) = \frac{46{\cdot}55\ (\mathrm{d}E/\mathrm{dR})}{1+46{\cdot}55\,\sigma_r V_e V_g^{-1}\, t\ (\mathrm{d}E/\mathrm{d}R)} \tag{6}$$

Fowler and Perkins (1951) have given the following dependence of the specific energy loss ($\mathrm{d}E/\mathrm{d}R$) on the charge ($\mathcal{Z}$) and velocity (β) of the particle traversing a G-5 emulsion:

$$\frac{\mathrm{d}E}{\mathrm{d}R} = \frac{0{\cdot}587\ \mathcal{Z}^2}{\beta^{1{\cdot}46}}\ \mathrm{keV}/\mu\mathrm{m} \tag{7}$$

Thus substituting this value of $\mathrm{d}E/\mathrm{d}R$ in equation (6) we have:

$$\pi = 1-\exp\left[\frac{27{\cdot}325\ \mathcal{Z}^2\beta^{-1{\cdot}46}}{g_0 B(B/S-1)\ (1+27{\cdot}325\ \sigma_r V_e V_g^{-1}\, t\ \mathcal{Z}^2\beta^{-1{\cdot}46})}\right] \tag{8}$$

The grain density is defined as the product of probability of development (π) and the total number of grains per hundred microns of an undeveloped emulsion, $\mathcal{N}$, and can be estimated by the following expression:

Grain density, $g = \pi\mathcal{N}$

$$\text{or } g = \mathcal{N}\left[1-\exp\left\{\frac{27{\cdot}325\ \mathcal{Z}^2\,\beta^{-1{\cdot}46}}{g_0 B(B/S-1)\ (1+27{\cdot}325\ \sigma_r V_e V_g^{-1}\, t\ \mathcal{Z}^2\beta^{-1{\cdot}46})}\right\}\right] \tag{9}$$

Thus a charged particle traversing through the nuclear emulsion renders some grains developable along its path during the process of development. The linear array of such developed grains forms a track which consists of discrete grains, blobs (aggregate of continuous grains due to overlapping) and gaps (discontinuity in the track). The magnitude of the average gap length per 100 μm of path length is defined as the mean gap length. The relation between the mean gap length and the probability of development is as follows (Sharma and Gill 1962 and Gaur and Sharma 1970).

$$\pi = \frac{d}{\bar{w}+d}\ \text{(neglecting the cut off in order to increase the accuracy of } \pi\text{).}$$

where d is the diameter of the grain and $\bar{w}$ is the mean gap length. Substituting the value of π from the above equation in equation (6), we get:

$$-g_0 B(B/S-1)\log(1+d/\bar{w}) = \frac{46{\cdot}55\,(\mathrm{d}E/\mathrm{d}R)}{1+46{\cdot}55\;\sigma_r V_e V_g^{-1}\,t\,(\mathrm{d}E/\mathrm{d}R)} \tag{10}$$

Thus substituting the value of $\mathrm{d}E/\mathrm{d}R$ from equation (7) in the above equation, we have:

$$d/\bar{w} = \exp\left[\frac{-27{\cdot}325\;\mathcal{Z}^2\beta^{-1\cdot46}\,S}{g_0 B(B-S)(1+27{\cdot}325\;\sigma_r V_e V_g^{-1}\,t\,\mathcal{Z}^2\beta^{-1\cdot46})}\right]-1 \tag{11}$$

The value of S involving the temperature dependence is given as (Singh, Sharma and Chauhan, 1972):

$$S = \frac{2\pi x_0 d^3}{3a^3\exp(w_0/2kT)} \tag{12}$$

where x_0 is a constant, the magnitude of which is a reasonable value as given by Mott and Gurney (1957), d is the grain diameter, a is the cell size of AgBr crystal, w_0 is the maximum energy required to take an interstitial Ag^+ ion away from the lattice point, k is Boltzmann's constant and T is the absolute temperature in degrees Kelvin.

Substituting the value of S from equation (12) in equation (11), we get:

$$\bar{w} = d/\exp\left[\frac{-27{\cdot}325\;\mathcal{Z}^2\beta^{-1\cdot46}\{2\pi x_0 d^3/3a^3\exp(W_0/2kT)\}}{dg_0 B\{B-(2\pi x_0)d^3/3a^3\exp(W_0/2kT)\}(1+27{\cdot}325\;\sigma_r V_e V_g^{-1} t\,\mathcal{Z}^2\beta^{-1\cdot46})}\right]-1 \tag{13}$$

or $\bar{w}=\psi(\mathcal{Z}, \beta, T)$, where ψ denotes a function. This expression gives a relation between $\bar{w}$, $\mathcal{Z}$, β and T as other values are mere constants whose magnitudes can be substituted as follows:

d $= 0{\cdot}27\ \mu\mathrm{m}$ (for G-5 emulsion grains)
x_0 $= 30{\cdot}0$ (Mott and Gurney 1957)
a $= 5{\cdot}755\times10^{-4}\ \mu\mathrm{m}$ (for AgBr crystals)
k $= 0{\cdot}8617\times10^{-7}$ keV/10°K (Boltzmann's constant)
g_0 $= 275$ grains per 100 μm (Sharma and Gill 1962, Voyvodic 1954)
B $= 457$ ($n_{\max}$ from our calculations using equation 4; *see also* Sharma and Gill 1962)
V_g $= (4/3)\,\pi\,(d/2)^3 = 1{\cdot}03\times10^{-14}\ \mathrm{cm}^3$ (volume of G-5 emulsion grain)
V_e $= 10^7$ cm/sec (Bayer and Hamilton 1965 and Hamilton and Bayer 1965)
σ_r $= \sigma_t/25$, where σ_t is the capture cross-section of electron at the trap (Bayer and Hamilton 1965 and Hamilton and Bayer 1965)

2.2. *Ratio of recombination and trapping constants*

Within the grain under consideration, the number of electrons will decrease with time as a result of (i) recombination process (recombination of electrons with positive holes), and (ii) combination of electrons with interstitial Ag^+ ions for the formation of latent images.

If $n_e(t)$ is the number of electrons within the grain at any instant t, the contribution to the rate of decrease of n_e, due to process (i), is a term proportional to n_e and also proportional to the number of vacant places, n_h. The contribution to the rate of decrease due to process (ii) can be expressed by a term proportional to n_e alone.

Therefore, for total loss of electrons we have:

$$dn_e/dt = -an_e n_h - C_e n_e \tag{14}$$

where a is the rate constant of recombination and C_e is a trapping or neutralization constant.

The ratio of recombination and trapping constant can be defined as a/C_e. The rate constant of recombination a is defined as $a = \sigma_r V_e V_g^{-1}$ (eq. 1) where $\sigma_r = \sigma_{t/25}$, σ_t being the capture cross-section of the electron at the trap. Substituting the values of constants $\sigma_t = 3 \times 10^{-15}$ cm^2 (Seitz 1951 and also a value preferred by us) and V_e and V_g as shown above, we get $a = 1{\cdot}164 \times 10^5$ sec^{-1}.

C_e is a constant which depends upon the concentration of interstitial Ag$^+$ ions and therefore it can be written as (Hamilton and Brady 1962, Tan 1972 private communication):

$$C_e = CN_I = 1/\tau_n \tag{15}$$

where τ_n is the mean neutralization time of Ag$^+$ ions with electrons and C is another constant of proportionality. N_I is defined as the total number of interstitial Ag$^+$ ions per unit volume and is expressed by the following relation (Singh and Sharma 1972):

$$N_I = \sqrt{2}N\ [\sqrt{2} \exp(-G_I/kT) + \exp(-G_F/2kT)] \tag{16}$$

where N is the number of AgBr ion-pairs per unit volume, G_F is the free energy of formation ($\sim 0{\cdot}92$ eV) for a Frankel defect pair in AgBr (Muller 1965, Trautweiler 1968), G_I is the free energy of formation ($\sim 0{\cdot}32$ eV) of an interstitial Ag$^+$ ion at the surface (Muller 1965).

The time of neutralization of Ag$^+$ ion (τ_n) is given by (Hamilton and Brady 1962):

$$\tau_n = 1/N_I V_i \sigma_i \tag{17}$$

where σ_i is the capture cross-section of one of these Ag$^+$ ions at the trapping centre and V_i is its thermal velocity. The magnitude of V_i is defined as:

$$V_i = \mu kT/a_e \tag{18}$$

where μ is the ionic mobility, k is the Boltzmann's constant, T is the absolute temperature, a is the lattice constant and e is the electronic charge.

From equations (15), (17) and (18) we have:

$$\tau_n = a_e/\mu kT\sigma_i N_I \tag{19}$$

Thus we get, $C_e = 1/\tau_n \quad = N_I V_i \sigma_i$

$= 6 \times 10^7$ sec^{-1} (at room temperature)

Therefore, the ratio of a/C_e will be:

$$\frac{a}{C_e} = \frac{1{\cdot}164 \times 10^5 \text{ sec}^{-1}}{6 \times 10^7 \text{ sec}^{-1}} \simeq 2 \times 10^{-3} \tag{20}$$

This value of a/C_e calculated on the basis of $\sigma_t = 3 \times 10^{-15}$ cm^2 is exactly the same as calculated by Della Corte *et al* (1953) on the basis of a different model involving some experimental considerations.

3. Results and discussion

Theoretical values of effective ionization (n), probability of development (π) and grain density (g) for various values of specific ionization ($\mathrm{d}E/\mathrm{d}R$) are calculated from equations 4, 8 and 9 respectively by assuming the values of the cross-section parameter as $\sigma_t=1\times10^{-15}$ cm^2, 3×10^{-15} cm^2, 5×10^{-15} cm^2 and 10×10^{-15} cm^2 and taking $t=10^{-8}$ sec, $1{\cdot}88\times10^{-8}$ sec, for each value of σ_t. Calculations are made only for the room temperature ($\sim$293 K). The reason for choosing the value of the capture cross-section, σ_t between 10^{-15} cm^2 and 10^{-14} cm^2 is that these are the limiting minimum and maximum values as suggested by various authors (Bube 1960, Hamilton and Bayer 1965, Rose 1951, Rose 1963 and Seitz 1951), and we may expect some suitable magnitude of σ_t somewhere between these limiting values. The value of the trapping time, t considered by us in this work is as suggested by others (Hamilton and Bayer 1965, Mitchell and Mott 1938, and Sharma and Gill 1962).

Figures 2–4 show the relationship between n vs $\mathrm{d}E/\mathrm{d}R$, π vs $\mathrm{d}E/\mathrm{d}R$ and g vs $\mathrm{d}E/\mathrm{d}R$ respectively for various sets of σ_t and t values. In all these figures the comparison of theoretical curves due to various sets of σ_t and t is made with the experimental curves (Della Corte *et al* 1953, Fowler and Perkins 1951, and Gaur and Sharma, 1970). The curves indicate that the most suitable value of σ_t is 3×10^{-15} cm^2 and that of t is $1{\cdot}88\times10^{-8}$ sec, as in all the three figures the theoretical curves based on these values are very close to the experimental curves.

The theoretical values of mean gap length ($\bar{w}$) computed from equation (13) for room temperature and relativistic particles ($\beta=1$) in a charge interval $1\leqslant Z\leqslant12$ are plotted

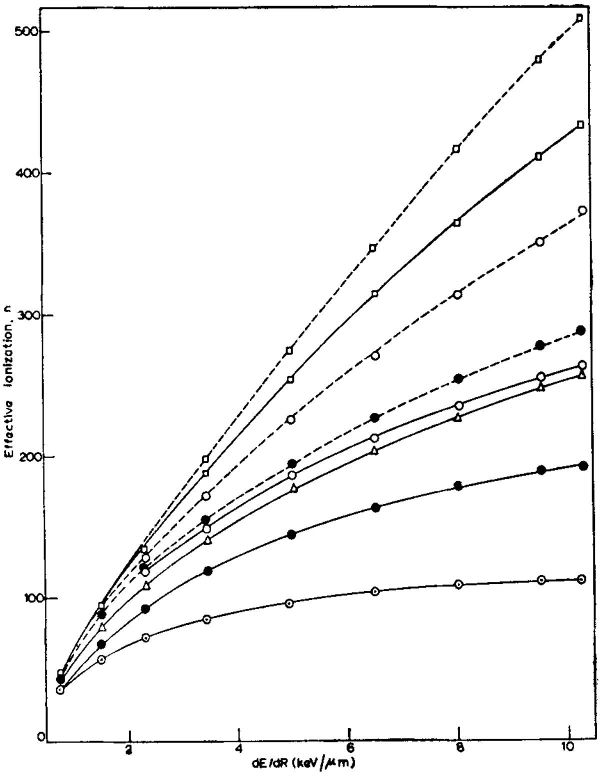

Figure 2. Variation of n with $\mathrm{d}E/\mathrm{d}R$.

(a) The dotted curves indicate $t=1\times10^{-8}$ sec with $\sigma t=1\times10^{-15}$ cm^2(□), $\sigma_t=3\times10^{-15}$ cm^2(○), $\sigma_t=5\times10^{-15}$ cm^2 (●).

(b) The continuous curves indicate, $t=1{\cdot}88\times10^{-8}$ sec with $\sigma_t=1\times10^{-15}$cm^2 (□), $\sigma_t=3\times10^{-15}$ cm^2 (○), $\sigma_t=5\times10^{-15}$ cm^2 (●), $\sigma_t=10\times10^{-15}$ cm^2 (◉).

(c) △ points indicate the experimentally observed values.

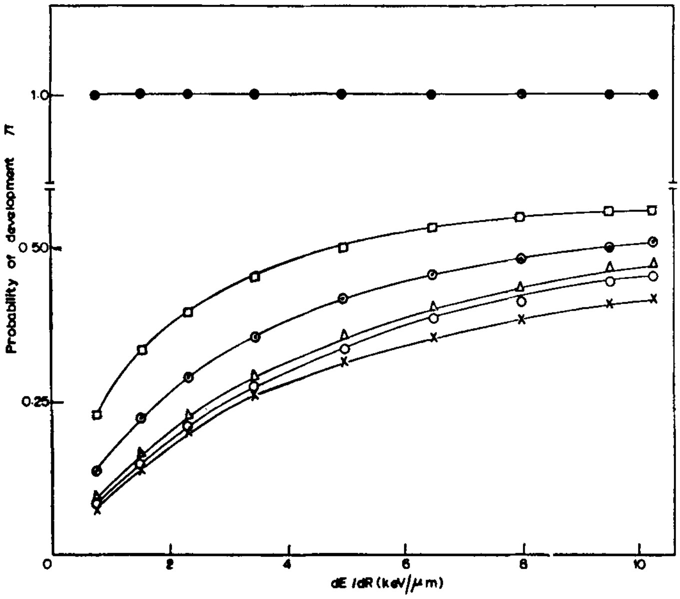

Figure 3. Variation of π with d*E*/d*R*.

(a) x points indicate $t=1\times10^{-8}$ sec and $\sigma_t=3\times10^{-15}$ cm².

(b) All other points indicate $t=1{\cdot}88\times10^{-8}$ sec with $\sigma_t=1\times10^{-15}$ cm² (●), $\sigma_t=3\times10^{-15}$ cm² (○), $\sigma_t=5\times10^{-15}$ cm² (◎), $\sigma_t=10\times10^{-15}$ cm² (□).

(c) △ indicate the experimentally observed values.

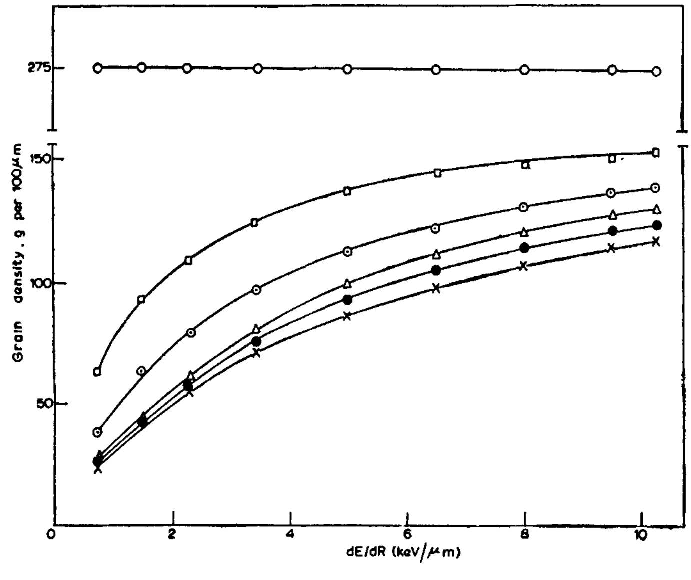

Figure 4. Variation of with d*E*/d*R*.

(a) x points indicate $t=1\times10^{-8}$ sec and $\sigma_t=3\times10^{-15}$ cm².

(b) All other points indicate $t=1{\cdot}88\times10^{-8}$ sec with $\sigma_t=1\times10^{-15}$ cm² (○), $\sigma_t=3\times10^{-15}$ cm² (●) $\sigma_t=5\times10^{-15}$ cm² (◎), $\sigma_t=10\times10^{-15}$ cm² (□).

(c) △ points indicate the experimentally observed values.

in figure 5. The comparison of the theoretical curves of figures 5 is made with the experimental curve due to Gaur and Sharma (1970). It is concluded that the curve that fits best (which is very near to the experimental curve) is based on the choice of the parameters as $\sigma_t=3\times10^{-15}$ cm² and $t=1{\cdot}88\times10^{-8}$ sec. These curves also indicate that upto a charge $Z\simeq4$ or 5 the mean gap length is affected considerably by a small change in the value of the capture cross-section while for particles of charge $Z\geqslant5$ the mean gap length is affected very slightly.

The ratio of the rate constant of recombination and trapping, (a/C_e) given by equation (20) and computed on the assumption of the value of $\sigma_t\simeq3\times10^{-15}$ cm², gives a magnitude as $a/C_e\simeq2\times10^{-3}$ which is in agreement with the result calculated by Della Corte *et al* (1953) on the basis of their experimental considerations.

Thus the results presented in figures 2–5 and the computed value of a/C_e, all lead to the conclusion that the most suitable capture cross-section of electrons at the trap in AgBr grains of nuclear emulsion is $\sigma_t \sim 3 \times 10^{-15}$ cm^2 which is similar to the value suggested by Seitz (1951).

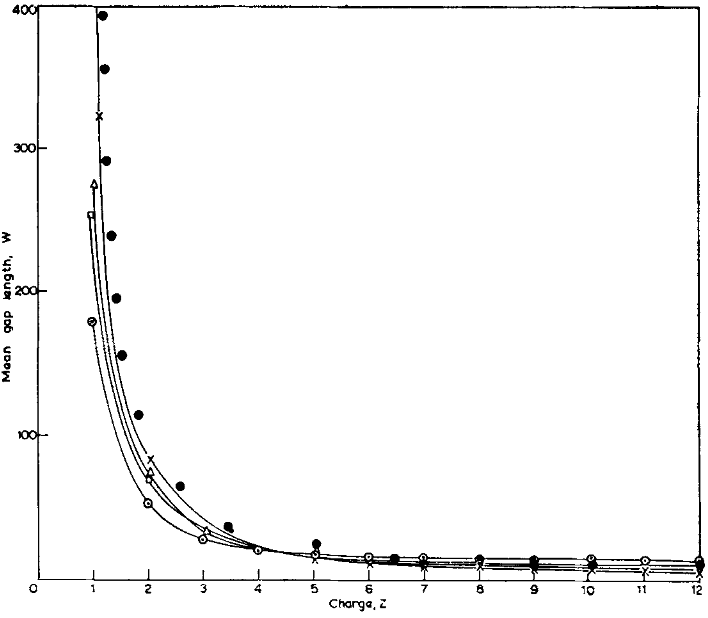

Figure 5. Variation of $\bar{w}$ with Z.

□ points indicate $t = 1{\cdot}88 \times 10^{-8}$ sec and $\sigma_t = 3 \times 10^{-15}$ cm^2; ⊙ points indicate $t = 1{\cdot}88 \times 10^{-8}$ sec and $\sigma_t = 5 \times 10^{-15}$ cm^2; × points indicate $t = 1 \times 10^{-8}$ sec and $\sigma_t = 3 \times 10^{-15}$ cm^2; △ points indicate $t = 1 \times 10^{-8}$ sec and $\sigma_t = 5 \times 10^{-15}$ cm^2; and ● points indicate the experimentally observed values.

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