ON THE VARIATION OF GRAIN DENSITY WITH SPECIFIC ENERGY LOSS IN NUCLEAR EMULSIONS

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ABSTRACT. An attempt has been made to explain the experimental curves showing the relation between grain density (ionisation parameter) and specific energy loss in Nuclear Emulsions. A model based on the recombination of positive holes and electrons during the process of latent image formation has been suggested. The theoretical results agree satisfactorily with the experimental ones.

INTRODUCTION

The analysis of nuclear tracks in photographic emulsions caused by the passage of charged particles mainly depends on the ionization (density or length) parameters. Experimental observations show that for low values of energy losses grain density is directly proportional to the rate of loss of energy of the charged particle producing the track but at higher values of energy loss it deviates from linearity (Fowler, 1950; Fowler and Perkins, 1951; Powell *et al*, 1959; and Sharma and Gill, 1962).

Beriman (1951) tried to explain this variation by considering a simplified model of track formation assuming grains of constant size and sensitivity and a uniform rate of loss of energy of the parent particle but his theoretical curves do not agree with the experimental ones.

A linearity between grain density and energy loss can be expected only if the entire energy loss in various grains is used in creating positive holes which are used up in latent image formation. The deviation at high energy losses from linearity clearly indicates that only a part of the positive holes created due to the passage of the charged particle is used up in latent image formation and the rest may be lost somewhere without making any contribution to the development of the grain.

In this paper, we have tried to explain theoretically the experimental variation of grain density with energy loss assuming the presence of the recombination of positive holes with electrons during latent image formation as suggested quantitatively by many workers (Demers, 1947; Webb, 1948; Dell-Corte *et al*, 1953; Mitchell, 1957; Mitchell and Mott, 1957; Hamilton *et al*, 1965). Considering the positive hole theory of Mitchell and Mott (1957) for latent image formation a simple model is presented for evaluating the effective number of positive holes

(effective ionization) available for latent image formation (Sharma and Gaur, 1969). This model helps in formulating the mechanism of track formation in nuclear emulsions and in calculating primarily the probability that a silver halide grain in the path of the particle may be impressioned so as to render it developable and finally the grain density at any specific energy loss.

THEORY

a) *Effective Ionization in a Grain.*

It is well known fact that a charged particle traversing the nuclear emulsion loses its energy in releasing electrons from the normal state of halide ion. As a result of this equal number of positively charged vicinities are created in the lattice of the crystal which are referred as "positive holes". The number of positive holes (n_0) depends on the rate of energy loss (dE/dR) and the path length (d) through an emulsion grain. It is probable that electron and positive hole may recombine immediately after their production to form a halide ion from which they were originated.

The recombination being a phenomenon involving two ions whose concentrations are equal to 'n', will be governed by the following relation

$$-\frac{dn}{dt} = \lambda n^2 \qquad .. \quad (1)$$

where dn/dt represents the rate of change of ion concentration and n is the number of electrons or positive holes present at any instant t and λ, the coefficient of recombination which depends on the nature of the medium through which the charged particle traverses. The nature of the medium depends on the number of sensitivity centres (sensitivity specks, clumps of silver or impurity atoms, kink sites or edge dislocations) which act as trapping centres for the ions (+ ve holes and the interstitial Ag^+ ions). The greater the number of sensitivty centres the smaller will be the probability that positive holes are left behind to recombine i.e., less will be the recombination and vice-versa. Hence the constant λ can be defined as $\lambda = C/S$, S being the number of sensitivity centres and C a different constant of proportionality. On substituting the value of λ in equation (1) we have

$$-dn/n^2 = (C/S)dt \qquad .. \quad (2)$$

On integrating it we get

$$1/n = (C/S)t + C' \qquad .. \quad (3)$$

where C' is a constant of integration. By applying the boundary condition when $t = 0$ (the instant when holes are created), $n = n_0$ (the maxm. number of holes present at that moment), from the above relation we get $C' = 1/n_0$ Therefore,

$$n = \frac{n_0 S}{S + n_0 C t} \qquad .. \quad (4)$$

This shows a relation between n_0, the total number of holes produced at some specific energy loss in a grain and n, the number of effective holes at any instant t left after recombination. The number of positive holes (n_0) depends on the rate of energy loss and the diameter (d) according to the following relation

$$n_0 = \frac{(dE/dR \text{ ev/micron}) \times (d \text{ microns})}{5{\cdot}8 \text{ ev.}}$$

In case of G-5 emulsions the diameter 'd' of a grain is $\simeq 0{\cdot}27$ micron. Hence expressing n_0 as function of energy loss, we have

$$n_0 = 46{\cdot}55\ dE/dR \tag{5}$$

where dE/dR is in Kev/μm.

Substituting the value of n_0 in eqn. (4) we get

$$n = \frac{46{\cdot}55\ (dE/dR)}{1 + 46.55\ (dE/dR)(Ct/S)} \qquad \ldots (6)$$

The constant of proportionality C can be calculated with the help of equation (2) and comes out to be $1{\cdot}3356 \times 10^6$ (Appendix 1).

At any instant t, the number of positive holes left after possible recombination and available for latent image formation is given by relation (6). Initially at $t = 0$, the number of holes available for latent image formation is n_0, but as the time elapses some of the holes recombine and only the rest at that instant are used for latent image formation. To what extent does this process continue ? The recombination will continue till the holes survive.

The minimum value of the survival time or life time of positive holes as shown by Malinowski (1967) is $2{\cdot}5 \times 10^{-6}$ sec. This can easily be taken as the limiting value of time t used in relation (6).

We take $S = 2000$, the number of sensitive centres or trapping centres (Sharma and Gill, 1962; Mess, 1948). Substituting the values of C, t and S (estimated as above) in equation. (6), we get,

$$= \frac{46{\cdot}55(dE/dR)}{1 + 0{\cdot}0777(dE/dR)} \tag{7}$$

The effective number of positive holes or effective ionization for different dE/dR values can be calculated from equation (7) and the values of total number of positive holes, n_0, from equation (5).

b) *Theoretical Grain Density.*

The passage of a charged particle through a grain is followed by an excess of positive charge at the surface of the crystal in the form of positive silver ions

and a corresponding negative charge in the form of electrons in the conduction band (Powell *et al*, 1959). The positive silver ions are distributed in the sensitivity centres where they are neutralized with electrons resulting in the formation of latent image.

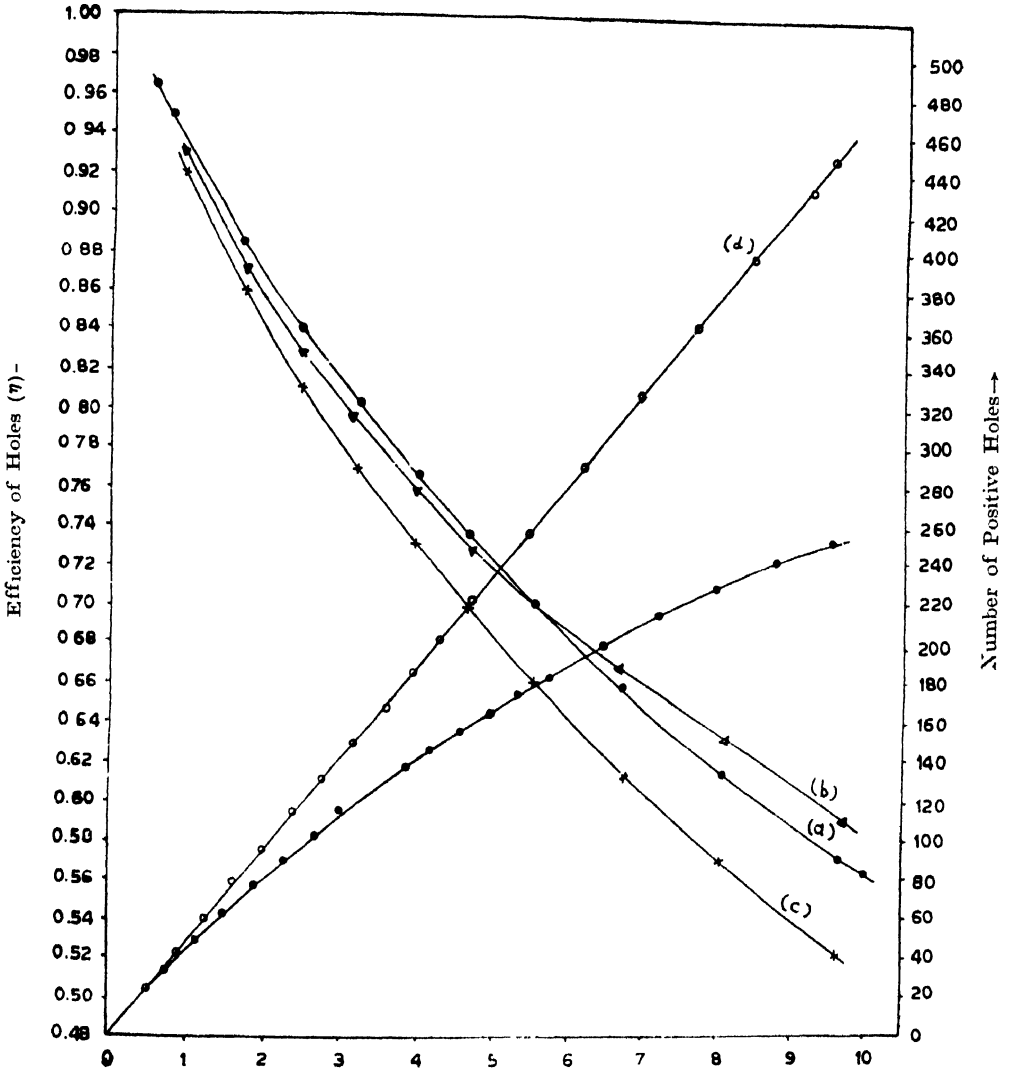

Figure 1. (A) Variation of η with dE/dR.

Δ Points indicate the work of Della Corte *et al.*, (1953).

× Points indicate the work of Sharma and Gill (1962)

O Points indicate the present work.

(B) Variation of n_0 and n with dE/dR

Curve (a) indicates the variation of $n_0 - dE/dR$

Curve (b) indicates the variation of $n - dE/dR$.

For calculating the probability of development of a grain we have considered a model similar to that of Sharma and Gill (1962) with a slight modification. We consider the distribution of Ag^+ ions in the sensitivity centres as a primary process. Moreover, we have made emphasis on the probability of development of active centres within individual grains rather than concentrating on the active centres of all the grains simultaneously lying along a certain path length. On these considerations (Appendix 2) the number of active centres S' in a grain which may become developable due to the passage of charged particle through it can be expressed as

$$S' = 1{\cdot}327n\, e^{-0.0005n}[1+6{\cdot}635\times10^{-4}(n-B)+2{\cdot}2013\times10^{-7}(n-B)(n-2B)+4{\cdot}83\times10^{-11}(n-B)(n-2B)(n-3B)+8{\cdot}075\times10^{-15}(n-B)(n-2B)(n-3B)(n-4B)+\text{negligible terms}..] \quad .. \quad (8)$$

For the first approximation

$$S' = 1{\cdot}327\, ne^{-0.0005n}[1+6{\cdot}635\times10^{-4}(n-B)] \quad .. \quad (9)$$

Where n is the effective number of positive holes produced in a grain at some specific energy loss and B is the limiting number of positive holes ($B = 493$) as suggested by one of the authors (Sharma and Gill ,1962) earlier.

Further the probability of development (π) of a grain can be estimated by considering the number of undeveloped grains along a certain path length of the charged particle and imposing a condition that a grain will become developable if it acquires at least one active development centre in it so as to render the whole of the grain developable during the process of development. Thus by theoretical considerations the value of probability of development can be given by the following relation.

$$\pi = 1-e^{-S'} \quad .. \quad (10)$$

where S' can be estimated from equation (9). The value of S' (hence π) involves the theoretical parameters which depend on the characteristics of unprocessed emulsion. Relation (10) can be used to estimate the expected number of grains/100 μm i.e., grain density by multiplying π with N (the number of grains/100 μm. in an unprocessed emulsion).

$$\text{Grain density,} \qquad g = \pi\times N(1-e^{-S'}) \quad .. \quad (11)$$

As number of grains/100 μm. for unprocessed G-5 emuslion is around 275–300 (Sharma and Gill, 1962), we can estimate the theoretical grain density from equation (11). The calculated values of grain density for various energy losses are shown in figure 2.

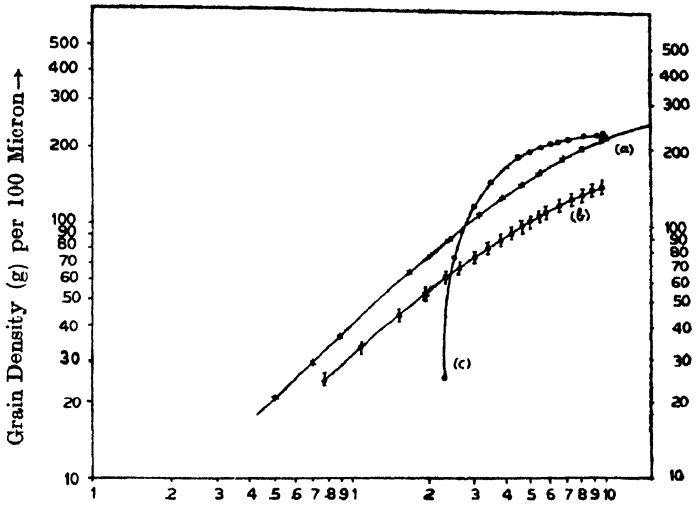

Figure 2. Variation of grain density with specific energy loss.
Curve (a) indicates the experimental work of Fowler and Perkins (1951).
Curve (b) indicates the present theoretical work. The upper limit of ϕ denotes the grain density for $S = 2000$ and the lower limit indicates grain density for $S = 1500$ and the centre points (O) indicate grain density for the average number of sensitivity centers between 1500 and 2000.
Curve (c) indicates the throretical work of Beriman (1951).

RESULT AND DISCUSSION

The values of number of positive holes produced/grain (n_0) and number of effective holes (n) left after the recombination process for various values of energy loss (dE/dR), calculated from relation 5 and 7 are shown in the table (column 2 and 3). The variation of n_0 and n with dE/dR is also shown in figure 1. This shows that the number of effective positive holes at .low dE/dR is almost equal to the number of positive holes produced (i.e., the recombination being negligible in this region) while it is considerably reduced at high energy losses.

The grain density depending on the probability of development of a grain must depend on the number of positive holes produced in the grain. If it would have depended on the number of positive holes produced in a grain, there would have been no deviation from the straight line curve (a) of figure 1. The experimental relation between the specific ionization and the grain density for G-5 emulsion (figure 2*a*) also remains linear up to an specific ionization of 3 Kev/μm. after which the deviations start rapidly and at very high energy losses the curve tends to become saturated.

The theoretical curve (b) of figure 1 showing relation of effective number of positive holes with dE/dR has exactly the same nature as that of experimental

curve (a) of figure 2 due to Fowler and Perkins (1951) and Fowler (1950) between the grain density (g) and (dE/dR). The similar nature of these curves indicates a close relation between the grain density and the number of effective holes available for latent image formation.

Experimental curve (a) of figure 2 between grain density and specific energy loss has been explained by Beiser (1952) and Blau (1949) only qualitatively with the idea of space charge due to large number of electrons around the sensitivity centres. The quantitative picture of recombination considered by us on the basis of the +ve hole theory of Mitchell and Mott (1957) clearly explains the experimental curve between the grain density and dE/dR in the same way as that of Sharma and Gill (1962) based on the electron theory of Gurney and Mott (1938). Thus it does not make much difference whether one considers the distribution of electrons or of positive holes in the sensitivity centres of the grains to render them developable. It is true that the grain density depends on the number of effective positive holes left after recombination.

The variation of efficiency $\eta(= n/n_0)$ of utilizing the positive holes for latent image formation with dE/dR is shown in figure 1. It is clear from the curves that our theoretical values of the efficiency η for different dE/dR values are in better agreement with the semiempirical values of Della Corte *et al.*, The maximum efficiency η is observed when dE/dR is very small.

The values of the probability of development of a grain calculated from relation (10) for different energy losses are shown in the table (column 5). Our theoretical values complete well with the experimental values of Della Corte *et al.* (1953) and Sharma and Gill (1962) (Column 6 of the table).

The average theoretical value of π between successive intervals of dE/dR perfectly agree with the experimental values of column 6 for small specific energy losses. The small discrepancy in the throretical and experimental values of π at large energy loss intervals can be attributed to the production of a large number of positive holes.

Figure 2 clearly indicates that our theoretical curve (b) of figure 2 showing a variation of grain density with dE/dR is exactly of the same nature as the experimental curve (a) of figure 2 due to Fowler and Perkins. The theoretical curve (c) of figure 2 based on Beriman model does not agree at all with these two curves. Thus the present model of grain density vs. dE/dR after taking into account the process of recombination of positive holes and electrons seems quite satisfactory.

The discrepancy in the values of calculated grain density at various dE/dR values indicates that actually more grains/100 μm. are developed in comparison to what we expect theoretically. This discrepancy in grain density values may be attributed to the fact that some grains lying around the passage of a charged

particle are also developed by induction process, through they are not traversed and affected by the incident particle.

Table 1. Values of n_0, n, η and π at various energy losses

1		2	3	4	5	6	
dE/dR		No. of positive holes produced/ grain 'n_0'	No. of effective positive holes (calculated)/ grain 'n'	Efficiency η	Theoretical values of π (calculated)	Exporimental values of π between successive intervals of dE/dR	
mev/g cm^{-2}	in Kev/ μm.					Values due to Sharma and Gill (1962)	values due to Dell Corte *et al.*, (1953)
2.0	0.76	35.38	33.36	0.9429	0.0861	0.085	0.087
3.0	1.14	53.07	48.69	0.9175	0.1219	0.133	0.133
4.0	1.52	70.76	63.22	0.8934	0.1563	0.147	0.148
5.0	1.90	88.45	77.07	0.8712	0.1894	0.190	0.189
6.0	2.28	106.14	90.16	0.8495	0.2134	0.221	0.220
7.0	2.66	123.83	102.60	0.8286	0.2442	0.250	0.253
8.0	3.04	141.52	114.47	0.8089	0.2666	0.268	0.264
9.0	3.42	159.21	125.77	0.7899	0.2882	0.301	0.302
10.0	3.80	176.89	136.57	0.7720	0.3161	0.305	0.308
11.0	4.18	194.59	146.88	0.7548	0.3297	0.318	0.320
12.0	4.56	212.28	156.73	0.7384	0.3495	0.340	0.346
13.0	4.94	229.97	166.17	0.7226	0.3624	0.359	0.367
14.0	5.32	247.66	175.22	0.7075	0.3812	0.383	0.401
15.0	5.70	265.34	183.89	0.6930	0.3935	0.399	0.406
17.0	6.46	300.72	200.21	0.6658	0.4230	0.420	0.423
19.0	7.22	336.10	215.30	0.6406	0.4457	0.425	0.430
21.0	7.98	371.48	229.39	0.6175	0.4674	0.439	0.449
23.0	8.74	406.86	242.04	0.5949	0.4883	0.459	0.479
25.0	9.50	442.24	254.14	0.5747	0.5083	0.468	0.481

APPENDIX 1

For calculating the constant of the proportionality C, we take the help of the equation (2). Considering the boundary conditions $dt \to 1$ second, $S \to 1$ and $n \to 1$, we get $dn \to C$. As dn is the number of holes attenuated or recombined during time dt., C can be defined numerically equivalent to the number of holes recombined in unit time in the grain having one trapping centre and

with the condition that one hole is left behind after recombination. If the assumptions involved in defining the contant C are justifiable, the above definition of constant C is considered correct. Hence we have to see the validity of these conditions. As $t \to 1$ sec., is a very large value of time as compared to the time of recombination which is of the order of micro seconds, from relation (4), if $t \to \alpha$, $\propto \to 0 \propto$ Also $S = 1$ is a very small number of the trapping centres as compared to their number of the order of 2000 or more for G-5 emulsion grains (Sharma and Gill, 1962). Thus S may be considered tending to zero and according to this condition $n \to 0$ from equation (4). Moreover considering the third condition for defining C, that $n \to 1$ is almost the same as $n \to 0$. All these conditions lead to the same conclusion. When t is maximum (infinitely large) all the holes may recombine during this time and the effective number of holes left behind may tend to minimum or zero. When S is minimum (infinitely small) the positive holes will not be trapped but may recombine. Thus the constant C can be taken equal to the maximum number of positive holes recombined during reasonable interval of time and when the condition $n \to 0$ is satisfied. Since $n \to 0$, the number of recombined holes $(n_0 - n)$ will tend to n_0. As C is defined the maximum possible number of positive holes which may recombine during time t,

$$C \to dn(= n_0 - n)$$

or

$$C \to n_0$$

The maximum distance which a positive hole can move till it survives is defined as the diffusion length or range of diffusion. For calculating the maximum possible number of holes n_0, we imagine a cylinder having a maximum length in the grain and of radius equal to the diffusion length of the holes. This will describe a volume within which the recombination will be possible, otherwise outside the boundary of this imaginary cylinder, the holes will not be able to diffuse due to the fact that they can survive only for few micro seconds. For considering a state of maximum possible recombination in this volume of the grain we assume a maximum number of holes responsible for recombination. This maximum number will be the same as the number of Br ions in this volume. Thus finally the magnitude of constant C will be equal to the maximum number of Br ions in this assumed volume. The effective range of diffusion (diffusion length), l, of positive holes with a life time τ and a diffusion constant D can be calculated from $l = \sqrt{D\tau}$, a relation given by Malinowski (1967) who has recently given the value of diffusion constant (D) equal to 3×10^{-7} cm^2/sec. and the minimum value of $\tau \simeq 2$–3 microseconds while considering the properties of the photo excited holes in silver bromide crystals. The value of diffusion length, l, calculated from the above relation comes out equal to $8{\cdot}66 \times 10^{-7}$ cm. (or ·0086 μm). It clearly shows that the holes while diffusing in the grain perpendicular to the direction of the passage of a charged particle will diffuse to a maximum distance

of ·0086 μm during their life time. We are now interested in calculating the maximum number of holes within the cylinderical volume of radius 'l' and a height 'd' equal to the diameter of the grain. The maximum number of Br ions in this cylinder or the number of Ag Br molecules can be calculated from their crystal structure. Silver bromide grain is a cubic crystal of NaCl type and has a cell size 5·755 A. One cell contains four molecules. Hence the number of Br ions within this volume can be given by the following expression,

$$\frac{\text{Volume of the cylinder} \times \text{number of Br ions in one cell}}{\text{Volume of one cell}}$$

which comes out to be $1{\cdot}3356 \times 10^6$ for G-5 emulsion grains of 0·27 μm diameter. Thus the magnitude of constant C will also be equal to $1{\cdot}3356 \times 10^6$.

APPENDIX 2

For calculating the probability of development of a grain, consider the most general model of the grains having a random distribution in the gelation of the emulsions and also having varying sensitivities and sizes. In a grain n positive ions are to be distributed over its S sensitivity centres having different sensitivities from minimum to maximum. Suppose n_i ions (out of n ions) are distributed over S_i sensivivity centres (out of S sensitivity centres) each requiring i ions to render it developable, the number of sensitivity centres (S'_i) which may become developable from this group will be given by the following relation

$$S_i' = S_i \sum_{i}^{n_i} p(x) \qquad \ldots (1)$$

where $p(x)$ is the probability that one of the S_i sensitivity centres gets x of these n ions. $p(x)$ can mathematically be defined as

$$p(x) = C_x^{n_i} \frac{(S_i - 1)^{n_i - x}}{S_i^{n_i}} \qquad \ldots (2)$$

Sharma and Gill (1962) in earlier work have considered a similar distribution of electrons in various sensitivity centres. They have considered the distribution of number of electrons over the sensitivity centres of grain groups present in a certain path length which does not seem to be much feasible in comparison to the present distribution of positive ions in individual grains. The reason is that positive ions produced in one grain remain confined to that grain and cannot

migrate to other neighbouring grains. Thus the present model has more fundamental assumptions in comparison to the previous one and is based on prevelent Mitchell and Motts' positive hole theory of latent image formation in nuclear emulsions.

In this random distribution various groups will be possible depending on the various values of i i.e., $i = 1$ to $i = B$, a limiting value of positive ions to be utilized for latent image formation. As a result of this grouping, let S' be the mumber of sensitivity centres becoming developable per grain, then S' can mathematically be represented as

$$S' = \sum_{i=1}^{B} S'_i$$

$$S' = \sum_{i=1}^{B} S_i \sum_{x=i}^{n_i} C_x^{n_i} \frac{(S_i-1)^{n_i-x}}{S_i^{n_i}} \qquad .. \quad (3)$$

The solution of the above equation is as

$$S' = \gamma e^{-\alpha}(1-\beta)^{-1}[1+(\alpha-\beta)(1-\beta)^{-1}+\tfrac{1}{2}(\alpha-\beta)(\alpha-2\beta)(1-\beta)^{-2}+$$

$$+\tfrac{1}{6}(\alpha-\beta)(\alpha-2\beta)(\alpha-3\beta)(1-\beta)^{-3}+\tfrac{1}{24}(\alpha-\beta)(\alpha-2\beta)(\alpha-3\beta)$$

$$(\alpha-4\beta)(1-\beta)^{-4}+..+.. \text{ negligible terms}] \qquad .. \quad (4)$$

where $\alpha = \frac{n}{S}$, ratio of effective number of positive holes and total number of sensitivity centres in a grain.

$\beta = \frac{B}{S}$, ratio of limiting number of positive holes and total number of sensitivity centres.

and $\gamma = \frac{\alpha}{\beta} = \frac{n}{B}$

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