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Abstract

Statistical problems presented by the granularity of nuclear track emulsion have been treated. In concentrated emulsions a large fraction of the volume is occupied by solid silver halide crystals of various diameters. This excluded volume makes inapplicable considerations that presuppose the conditions leading to simple distributions. An exact treatment of the elementary track-cell is given. A corrected form of the Jdanov formula is derived. The effect of the emulsion granularity on the energy-loss straggling is calculated. A general formulation of the developability cross section of an emulsion grain is made. The mean primary grain density and the grain-density distribution are calculated. The gap-length distribution is derived. It is proved that this is exponential for gaps of all lengths, and that the gap-length coefficient is equal to the true grain density. The effect of secondary ionization is discussed.

Parameters have been introduced that can be evaluated experimentally. Knowledge of these quantities will presumably be useful in further resolving the problem of silver halide sensitivity. The statistical methods developed may be useful in other excluded-space problems.

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A. Introduction

When nuclear track emulsion intercepts a charged particle a series of developable grains is left in the emulsion along the particle trajectory. From an analysis of the developed track it may be required that the particle charge, velocity, and mass be determined with minimum error. To do this entails an understanding of how these particle quantities are translated by the emulsion instrument into measurable features of the track. From the track statistics, a particle quantity can be determined only within a certain confidence interval. It may often be crucial correctly to estimate what this interval is. On the other hand, sometimes it is desired to obtain the maximum amount of information from tracks with no more than a given amount of labor. An important respect in which emulsion can be improved as an instrument is in the "information density" contained in the track. One may ask how, and how much, this can be raised. These examples suggest that to attain mastery of emulsion as an instrument, an understanding of its statistical behavior is necessary. That is what is attempted in this paper.

Silver halide, which is the sensitive component of nuclear track emulsion, is present as crystals or grains of variable size that are randomly distributed throughout the gel matrix. The emulsions are normally concentrated, so that a large fraction of the volume is occupied by the crystals. This causes the statistical problems presented by emulsion to be complex.

Electron micrographs have shown, however, that the diversity of crystals can be reasonably well represented by a single parameter, the diameter. Only a few of the largest crystals do not appear to be spheres, and no appreciable error is introduced in treating them all as if they were.*

One always undertakes an analysis such as this with a number of postulates, stated or implicit. The assumption of a spherical shape for the crystals has been stated. Some others are also perhaps worth mentioning.

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*In principle, an arbitrary distribution of crystal shapes as well as sizes could be treated with additional parameters. If the crystal shape were described by a spherical harmonic analysis relative to the axes of its inertial ellipsoid, a set of coefficients would be obtained. A joint distribution function of these coefficients would exist describing the crystal population. For each crystal there would be in addition three space and two angle coordinates. These could be treated as random variables subject to the constraint of noninterpenetrability introduced later in this paper. Perhaps in certain cases such a treatment may become necessary and practical. It can also be seen that various results obtained in this paper do not depend critically on the grain shape, and are true for grain populations on which have been imposed only slight restraints.

It is assumed, of course, that the mixing is complete; that there is no stratification or marbling of the emulsion that might destroy the randomness. The randomness also could be affected in another way if the concentration of silver halide were so high that locally a further increase might sometimes be impossible. Were the crystals all of one size, the maximum volume concentration is $\pi/3\sqrt{2}$ or about 3/4. Because of the size distribution of grains present in the emulsion, the limiting concentration is considerably higher, and with a correctly chosen spectrum of grain sizes, the concentration could be made to approach unity.

Even if the average concentration were not near the maximum, if attractive forces between crystals caused them to aggregate, the assumed independence and randomness of the crystals would be compromised. The assumption has been made in this treatment that the fraction of the total volume consisting of such saturated volumes is negligible. The average volume concentration of silver halide in standard emulsion is about 1/2, and there is almost no evidence for the presence of crystal aggregates. The effect of crystal clumping on measured variances therefore is expected to be small.

Another assumption is that the volume into which no crystal can intrude is just the volume occupied by the other crystals, thus neglecting possible effects of the electrical double layer at crystal surfaces or the protective gel envelope surrounding each crystal. This is not a fundamental deficiency of the theory, and the statistics can be reformulated to permit the excluded volume to be enlarged if the increased complexity is warranted, and if these effects are physically important.

B. Statistical Geometry of the Grains

1. Expectation Values

In an emulsion, suppose the fraction of crystals with diameters exceeding D is $\int_D^\infty F(D)dD$. Let a crystal be penetrated by a particle, the segment of path in the grain being of length δ. The joint probability, $U(\delta, D)$, that the crystal diameter should be in the interval dD and that the path segment should be in the interval $d\delta$ is given by

$$U(\delta, D) = \frac{2\delta\, d\delta}{\langle D^2 \rangle} F(D)\, dD, \qquad (1)$$

with the employment of the following definitions:

$$\langle D \rangle = \int_0^\infty DF(D)dD,$$

$$\langle D^2 \rangle = \int_0^\infty D^2 F(D)\, dD = \langle D \rangle^2 + \sigma^2.$$

Then also

$$\langle A \rangle = \frac{\pi}{4}\langle D^2 \rangle = \frac{\pi}{4}\int_0^\infty D^2 F(D)\, dD = \frac{\pi}{4}\langle D \rangle^2 \left(1 + \frac{\sigma^2}{\langle D \rangle^2}\right),$$

the mean crystal cross section, and

$$\langle V \rangle = \frac{\pi}{6}\int_0^\infty D^3 F(D)\, dD \approx \frac{\pi}{6}\langle D \rangle^3 \left(1 + \frac{3\sigma^2}{\langle D \rangle^2} + \ldots\right),$$

the mean crystal volume. The mean length, $\langle \delta \rangle$, of path segments in crystals is

$$\langle \delta \rangle = \frac{2}{\langle D^2 \rangle}\int_0^\infty F(D)\, dD \int_0^D \delta^2\, d\delta = \frac{2}{3}\frac{\langle D^3 \rangle}{\langle D^2 \rangle} \qquad (2)$$

$$\approx \frac{2}{3}\langle D \rangle \left(1 + \frac{2\sigma^2}{\langle D \rangle^2}\right),$$

and their variance, $\langle \delta^2 \rangle - \langle \delta \rangle^2 \equiv \sigma_\delta^2$ is:

$$\sigma_\delta^2 = \frac{9 \langle D^2 \rangle \langle D^4 \rangle - 8 \langle D^3 \rangle^2}{18 \langle D^2 \rangle^2} \approx \frac{\langle D \rangle^2}{18} \left(1 + \frac{13 \sigma^2}{\langle D \rangle^2} \right). \tag{3}$$

The average number n of grains encountered in unit path is

$$n = N \langle A \rangle = \frac{\pi N \langle D^2 \rangle}{4} = \frac{\pi N}{4} \langle D \rangle^2 \left(1 + \frac{\sigma^2}{\langle D \rangle^2} \right), \tag{4}$$

where N, the average number of grains per unit volume, is given by

$$N = \frac{C}{\langle V \rangle} \approx \frac{6 C}{\pi \langle D \rangle^3} \left(1 - \frac{3\sigma^2}{\langle D \rangle^2} \right). \tag{5}$$

Here C is the volume concentration of silver halide in the emulsion.

A formula known as Jdanov's relation has frequently been used incorrectly to calculate n. Since $N \langle V \rangle = C$, and $n = N \langle A \rangle$, one can write

$$n = C \frac{\langle A \rangle}{\langle V \rangle} \approx \frac{3 C}{2 \langle D \rangle} \left(1 - \frac{2\sigma^2}{\langle D \rangle^2} \right). \tag{6}$$

The term $- 2 \sigma^2 / \langle D \rangle^2$ is not present in Jdanov's relation, as usually employed, and this may result in a considerable error. For Ilford G.5 emulsion it is about 7%. The reason for the discrepancy can be stated quite simply: The probability for encountering a grain is weighted by the square of its diameter; consequently the grains traversed by a particle are on the average larger than typical emulsion grains.

2. Distribution Functions

Let emulsion containing $N \int_D^\infty F(D)\, dD$ crystals per unit volume with diameters exceeding D be traversed by a particle moving in a straight line. The number of crystals whose centers lie at a distance between ρ and $\rho + d\rho$ from the particle trajectory, and whose diameters are contained in the interval dD, is

$$2 \pi N F(D) \rho \, dD \, d\rho \tag{7}$$

per unit path. A particular crystal of the above description is labeled "crystal C," and any other such crystal is of "type C."

Now one considers a different crystal, C', the diameter of which lies in the interval d D', and the center of which is removed from the trajectory a distance in the interval, d ρ'. The center of C' may be found anywhere with equal probability except in an excluded sphere of diameter $\frac{D + D'}{2}$ around crystal C. The probability that it be found in a volume element d V is $N F(D') d D' d V$.

In order that a segment of the particle path lie within crystal C, ρ must be less than D/2. Therefore, from Eq. (7), the mean number of crystals with diameters in the interval dD that are penetrated by the particle in unit path is

$$\frac{\pi N D^2}{4} F(D) d D,$$

and the probability that any particular crystal encountered belongs to class C is

$$\frac{8 \rho d \rho F(D) d D}{\langle D^2 \rangle} . \qquad (8)$$

One measures distance, μ, along the particle trajectory from the point defined by the projection of the center of C on the trajectory. The "distance" between crystals C and C' is the distance between the projections of their centers on the particle path.

The probability that a crystal satisfying the description of C' should be found at a distance between μ and μ + d μ from crystals C is

$$\frac{16 \pi N}{\langle D^2 \rangle} \rho \rho' F(D) F(D') d D d D' d \rho d \rho' d\mu . \qquad (9)$$

One now asks for the probability p that the particle reach a distance μ after traversing C <u>without</u> penetrating a crystal of type C'. This is formulated as follows:

$$\frac{1}{p} \frac{dp}{d\mu} = - \frac{16 \pi N}{\langle D^2 \rangle} \rho \rho' F(D) F(D') dD d D' d \rho d \rho' . \qquad (10)$$

The next step is to calculate the probability P that the particle may go a distance exceeding μ without traversing a crystal of any description. This is found from

$$\frac{1}{P}\frac{dP}{d\mu} = -\frac{16\pi N}{\langle D^2\rangle}\iiiint F(D)\,F(D')\,\rho\,\rho'\,d\rho\,d\rho'\,d\,D\,dD'. \tag{11}$$

The projected distance Δ, measured along the particle path, between centers of crystals successively traversed by a particle in emulsion is the elementary track-cell. Equation (11) provides the fundamental statistical law describing how this quantity varies. From Eq. (11), the probability that the length of the elementary cell exceed μ is

$$P = \exp\left[-\frac{16\pi N}{\langle D^2\rangle}\iiiint F(D)\,F(D')\,\rho\,\rho'\,d\rho\,d\rho'\,d\,D\,d\,D'\,d\,\mu\right]. \tag{12}$$

Because crystal C occupies a finite volume from which are excluded all other grains, a restriction on the limits of integration exists. The constraint can be written most simply as

$$(\rho + \rho')^2 > \left(\frac{D + D'}{2}\right)^2 - \mu^2 . \tag{13}$$

To evaluate Eq. (12) one distinguishes two regions of $\frac{D + D'}{2}$ $(\equiv \underline{D})$. When it is less than μ, the above inequality imposes no restriction on ρ and ρ', and the integral $\iint \rho\,\rho'\,d\rho\,d\rho'$ has the value $\frac{D^2}{16}\left(\underline{D} - \frac{D}{2}\right)^2$. The distances ρ and ρ' are constrained only to remain less than $D/2$ and $D'/2$ respectively.

When $\underline{D}$ is greater than μ three subregions must be distinguished.

For the moment one simply defines

$$\iint \rho\,\rho'\,d\rho\,d\rho' = \frac{1}{64}M(\underline{D}, D, \mu),$$

for $\underline{D} > \mu$, and Eq. (11) is written

$$\frac{1}{P}\frac{dP}{d\mu} = -\frac{\pi N}{4\langle D^2\rangle}\left[\int_0^{\mu} d\,\underline{D}\int_0^{2D} F(D)\,F(2\underline{D} - D)\,D^2(2\underline{D}-D)^2\,dD + \int_{\mu}^{\infty} d\underline{D}\int_0^{2D} F(D)F(2\underline{D} - D)M(\underline{D}, D, \mu)dD\right]. \tag{14}$$

The functions F(D) and F(2$\underline{D}$-D) fail to overlap where μ exceeds a certain length μ_0, and the second integral vanishes. For complete rigor, μ_0 must be set equal to the largest diameter occurring in the grain-size distribution.

In the first integral, when the maximum grain diameter is μ_0, and $\mu > \mu_0$, the integration over D and D' are independent and unrestricted. Then the right side of Eq. (14) becomes a constant:

$$-\frac{\pi N}{4\langle D^2\rangle}\int_0^{\mu_0} D^2F(D)dD\int_0^{\mu_0} D'^2F(D')dD'$$

$$=-\frac{\pi N\langle D^2\rangle}{4}=-n. \qquad (15)$$

The probability P that a particle traverse a distance exceeding μ ($\mu>\mu_0$) without encountering any crystal is therefore

$$P = W\exp(-n\mu), \qquad (16)$$

in which the constant factor W is given by

$$W=\exp\left[-\frac{\pi N}{4\langle D^2\rangle}\int_0^{\mu_0}d\mu\left\{\int_0^{\mu}d\underline{D}\int_0^{2D}F(D)\,F(2\underline{D}-D)D^2(2\underline{D}-D)^2\,dD\right.\right.$$

$$\left.\left.+\int_{\mu}^{\infty}d\underline{D}\int_0^{2D}F(D)\,F(2\underline{D}-D)M(\underline{D},D,\mu)\,dD\right\}+n\mu_0\right].$$

The function $M(\underline{D},D,\mu)$ has a different value in each of three subregions in the integration range of D. The three functions M_I, M_{II}, and M_{III} have been evaluated, and are as follows:

I $0<D<2(\underline{D}-p)$; $\quad p^2=\underline{D}^2-\mu^2$:

$$M_I=D^4-4\underline{D}D^3+4\underline{D}^2D^2=8/3\,p^4.$$

II $2(\underline{D}-p)<D<2p$:

$$M_{II}=\frac{D^4}{2}-8/3\,pD^3+(4\mu^2+16p\underline{D}-12\underline{D}^2)D^2$$
$$+(32\underline{D}^3-32p\underline{D}^2-16\underline{D}\mu^2)D \qquad (17)$$
$$+(16\underline{D}^2\mu^2-24\underline{D}^4+\frac{64}{3}p\underline{D}^3).$$

III $2p < D < 2\underline{D}$:

$$M_{III} = -8(\underline{D} - p)^2 D^2 + 16\underline{D}(\underline{D} - p)^2 D - 8/3(\underline{D} - p)^3(3\underline{D} + p) .$$

Then, in general, the probability P that the elementary cell length Δ exceed μ is

$$P = \exp\left\{ -\frac{\pi N}{4\langle D^2\rangle} \int_0^{\mu}\int_0^{\mu} d\underline{D} \int_0^{2\underline{D}} F(D)F(2\underline{D} - D)\, D^2(2\underline{D} - D)^2\, dD \right.$$

$$+ \int_{\mu}^{\infty} d\underline{D} \int_0^{2(\underline{D}-p)} M_I F(D)F(2\underline{D}-D)dD + \int_{\mu}^{\infty} d\underline{D} \int_{2(\underline{D}-p)}^{2p} M_{II}F(D)F(2\underline{D}-D)dD$$

$$\left. + \int_{\mu}^{\infty} d\underline{D} \int_{2p}^{2\underline{D}} M_{III}F(D)F(2\underline{D}-D)dD \right] d\mu \Bigg\} , \qquad (18)$$

with $p^2 = \underline{D}^2 - \mu^2$.

The mean value $\langle\Delta\rangle$ of Δ is found from

$$\langle\Delta\rangle = -\int_0^{\infty} \mu \frac{dP}{d\mu}\, d\mu , \qquad (19)$$

while the variance $\langle\Delta^2\rangle - \langle\Delta\rangle^2 \equiv \sigma_\Delta^2$ is

$$\sigma_\Delta^2 = \langle\Delta^2\rangle - \langle\Delta\rangle^2 = -\int_0^{\infty} \mu^2 \frac{dP}{d\mu}\, d\mu - \langle\Delta\rangle^2 . \qquad (20)$$

3. Heterogeneity Range Straggling

The range R of a particle is generally calculated from $R = \int_0^T \frac{dT}{\mathcal{J}}$, where T is the kinetic energy of the particle and $\mathcal{J}$ is a certain average energy loss per unit path. If $R_h = \int_0^T \frac{dT}{\mathcal{J}_h}$ is the range in pure halide, and $R_g = \int_0^T \frac{dT}{\mathcal{J}_g}$ is the range in pure gel, then $\mathcal{J} = \mathcal{J}_h C + \mathcal{J}_g (1 - C)$, where C is the volume concentration of halide in the emulsion. This insures that the range in emulsion will be the same as in a homogeneous material of the same average composition.

In addition to other causes of range straggling,[1] the ranges of a monoenergetic group of particles in emulsion fluctuate because of the granularity of the emulsion; the stopping power of the silver halide crystals is greater than that of the gel in which they are embedded.

If in a path length μ the sum of the halide paths is h, then the energy loss ϵ in this path is

$$\epsilon = h\mathcal{J}_h + (\mu - h)\mathcal{J}_g ,$$

$\mathcal{J}_h$ and $\mathcal{J}_g$ being energy losses in unit paths of halide and gel respectively.

The variance σ_ϵ^2 of the energy loss in a path μ is $(\mathcal{J}_h - \mathcal{J}_g)^2 \sigma_h^2$, σ_h^2 being the variance of h in path μ. The mean value of h in path μ is $\langle h \rangle = C\mu$.

The variance σ_h^2 of h is calculated as follows: The probability that at a distance in the interval $d\mu$ from crystal C, a crystal C' may be found in which the segment of path lies in the interval

$$d\delta' \left(= 4\rho' d\rho' / \left[D'^2 - (2\rho')^2 \right]^{\frac{1}{2}} \right)$$

is

$$\frac{4\pi N}{\langle D^2 \rangle} \rho \, d\rho \, \delta' \, d\delta'' \, F(D) \, F(D') \, d D \, d D' \, d\mu .$$

Then σ_h^2 may be calculated from

$$\sigma_h^2 = \frac{4\pi N}{\langle D^2 \rangle} \iiint (\delta'^2 - \langle \delta' \rangle^2)\, \delta' \, d\delta' \; \rho \, d\rho \; F(D)F(D')dD \, dD' \, d\mu. \tag{21}$$

As in calculating the distribution function for the elementary cell, $\rho + \rho'$ is subject to a constraint if very short paths μ, are of interest. Straggling of ranges that are less than the diameter of the largest crystal are of little importance. Although feasible by the above method, it is thought unnecessary to evaluate this case, and the integration is performed without restraints. The result is

$$\sigma_h^2 = \pi N \left[\frac{\langle D^4 \rangle}{8} - \frac{\langle D^3 \rangle^2}{9 \langle D^2 \rangle} \right] \mu. \tag{22}$$

Now, $\sigma_\epsilon^2 = \langle \epsilon^2 \rangle - \langle \epsilon \rangle^2$, so that

$$\sigma_\epsilon^2 = \pi N (\mathcal{J}_h - \mathcal{J}_g)^2 \left[\frac{\langle D^4 \rangle}{8} - \frac{\langle D^3 \rangle^2}{9 \langle D^2 \rangle} \right] \mu. \tag{23}$$

This is the variance of the energy loss in an element of path μ caused by the emulsion heterogeneity. If σ_R^2 is the variance of the residual range, and σ_T^2 is the variance of the residual energy of a particle from an originally monoenergetic population, then

$$d(\sigma_T^2) = \mathcal{J}^2 \, d(\sigma_R^2)$$

where $\mathcal{J} = (\mathcal{J}_h - \mathcal{J}_g) C + \mathcal{J}_g$ is the mean space-rate of energy loss. Therefore the range variance σ_R^2 is given by

$$\sigma_R^2 = \pi N \left[\frac{\langle D^4 \rangle}{8} - \frac{\langle D^3 \rangle^2}{9 \langle D^2 \rangle} \right] \int_0^{\langle R \rangle} \left(\frac{\mathcal{J}_h - \mathcal{J}_g}{\mathcal{J}} \right)^2 dR \,. \tag{24}$$

The ratio $\frac{\mathcal{J}_h - \mathcal{J}_g}{\mathcal{J}}$ varies slowly with velocity. If it is sufficiently accurate to consider it constant, the heterogeneity range variance is

$$\sigma_R^2 \approx \pi N \left[\frac{\langle D^4 \rangle}{8} - \frac{\langle D^3 \rangle^2}{9 \langle D^2 \rangle} \right] \left(\frac{\mathcal{I}_h - \mathcal{I}_g}{\mathcal{I}} \right)^2 \langle R \rangle, \tag{25}$$

where $\langle R \rangle$ is the mean range. If σ also is sufficiently small this may be further approximated by

$$\sigma_R^2 \approx 6 \pi \left(\frac{\mathcal{I}_h - \mathcal{I}_g}{\mathcal{I}} \right)^2 \left(1 + 11 \frac{\sigma^2}{\langle D \rangle^2} \right) \langle D \rangle \langle R \rangle. \tag{26}$$

This is to be added to other variance terms to obtain the total range variance of a monoenergetic group of particles in emulsion.

C. Theory of Primary Grain Density

1. General Considerations

Many individuals have written on the theory of the emulsion grain density.[2-17] While some of these attempts are chiefly of historical interest, or contain unacceptable premises, several introduce important concepts and observations that throw light on the nature of the track-forming process. Their results cannot be ignored, and an improved theory must first of all find accord with what has already been established.

In this new treatment a determined effort has been made to start with fundamentals, and to avoid, as far as possible, artificial models the peculiarities of which will have unknown consequences, and which must more or less seriously destroy one's confidence in the treatment. Good fortune permitted the finding of means for solving all the mathematical difficulties encountered. This has enabled a seemingly over-ambitious undertaking to be surprisingly successful. It is too much to expect, however, that entire generality can ever be achieved, and one can point out several respects in which the present theory does not make provision for various complications.

In the title of this section the expression "primary grain density" is used because a full and general treatment is yet to be given for the additional grain density arising from energetic delta rays, and no provision has been made for the effect of photons produced in the gel. In the theory it is supposed that grains are rendered developable only by being penetrated by the moving particle. Certainly, the probability that a grain will develop falls very sharply with the distance from its surface to the particle trajectory. Only secondary effects, such as delta rays, can be of any importance in rendering nonpenetrated grains developable, as the range of the particle field is only about one atom diameter. Since the explicit form of this probability function is not known, one replaces this "boundary layer" by a mathematical surface. There is little evidence that this simplication leads to serious error for normal emulsions, but in very-fine-grain or hypersensitized emulsions it may be important.

A related limitation on the generality of this work is that it assumes that each crystal is completely independent of others. This is not true, for example, when depletion of developer restricts the degree of development of strongly exposed portions of emulsion. The theory makes no provision for proximity development induced by a conceivable emulsion additive which would provide conduction bands for low-energy electrons to migrate from one crystal to the next, or from gel to crystal. It is also possible that the enlargement of a grain on development may cause protuberances of silver to intrude on an adjacent grain and break through its gelatin envelope. This could cause the neighboring grain to develop. The influence on the grain density of infectious development has been considered by Ahmad.[15] The effect should be chiefly to reduce the number of short gaps.

Provision has been made in deriving expression (41) below for the enlargement of the diameter, ϵ, that takes place on development to be a function of the grain diameter, but the additional implicit postulate that the enlargement is symmetrical perhaps reduces further the generality of the calculations.

No attempt has been made to allow for the possibility of nonuniform development, etching of surface grains, fog background, mechanical distortion of the emulsion, and many other conceivable complications that might affect empirical data.

It is also supposed that the sensitivity is the same from grain to grain. To treat mixtures of two emulsion types, such as have been used by Vanderhaeghe[16] and his collaborators, would require additional terms and cross-product terms in the final equations. The case of a partial population of totally inert grains, as postulated by Baillard,[14] here would require that the emulsion be treated as if it contained a second group of grains that is peculiar only in that its sensitivity vanishes.

2. Creation of Developability

All current theory of the latent image contemplates the initial creation of a free electron-hole pair in the crystal lattice. This is the primary act, and it is immaterial for this treatment whether the electron is captured in an impurity center or whether imperfections in the crystal lattice or on the surface (as proposed by Mitchell[19]) serve as points where free silver atoms aggregate. The detailed mechanism will, however, affect the coefficients A_{ijk}, introduced below, and it is to be hoped that fundamental problems of silver halide sensitivity may be resolved by a study of the influence on the A_{ijk} of physical and chemical conditions, for example, the temperature.

In this treatment it is assumed that the effect of a moving charged particle is as follows: The virtual photons of the electric field of the particle disturb the electrons of a silver halide crystal and induce transitions between their energy states. This produces electron-hole pairs in the crystal lattice, as free photons do. Some of the electrons released are given sufficient energy to produce further electron-hole pairs in the crystal, just as an x-ray photoelectron is capable of doing.

Let $\frac{d\Sigma}{dw}\,dw$ be the cross section for a moving charged particle to transfer energy in the interval dw to the electron in the crystal, and let n_0 be the electron density. Then

$$\left\langle \vartheta \right\rangle_h = n_0 \int_0^{w_{max}} w \, \frac{d\Sigma}{dw} \, dw \qquad (27)$$

is the energy loss per unit path in the crystal. (For the purpose of this discussion one need not distinguish between the different classes of electrons—a separate integral over each class would in general be required.)

In the above formula w_{max} is the maximum energy that the electron can absorb from the field of the moving particle. This may be so large that the range of the electron far exceeds the dimensions of the crystal. For this

reason Messel and Ritson suggested that the upper limit of w be cut off at some value w_0, corresponding to the electron energy required for escape from the crystal.[20] Then

$$\mathcal{J}' = n_0 \int_0^{w_0} w \frac{d\Sigma}{dw} \, dw , \qquad (28)$$

the restricted rate of energy loss, was to be considered the appropriate variable for determining the effectiveness of a charged particle in creating developability.

This procedure does not yet seem correct. It is suggested that one cannot weight equally all transfers of equal amounts of energy irrespective of its form. There is certainly a quantity of energy, w_1, of optimum size, so that per unit of energy a highest probability of development occurs. A relative efficiency E (w) for energy utilization should then be defined. (E (w) may be calculable from the spectral sensitivity curve for the emulsion obtained by measuring its sensitivity to photons of all energies.) Then

$$\mathcal{J}'' = n_0 \int_0^{w_{max}} w \frac{d\Sigma}{dw} \, E(w) \, dw \qquad (29)$$

is the function affecting the probability of development of a crystal traversed by a charged particle. This one may call the effective rate of energy loss.

The function E(w), however, is probably peaked well below w_0, and since the velocity dependence of $\frac{d\Sigma}{dw}$ occurs chiefly as a factor, one may expect a large range of high velocities over which $\mathcal{J}''$ is proportional to $\mathcal{J}'$. The distinction between them nevertheless should be retained.

A theory that does not take account of the statistical nature of the energy-loss process can have no fundamental validity. The loss of energy when a charged particle traverses a grain, as has been emphasized by Barkas,[21] Brown,[10] Fowler and Perkins,[13] and Bogomolov,[11] is a highly stochastic process. The mean rate of energy loss has little direct connection with the energy loss in a particular grain. Suppose one considers a 100-micron path of a 3-Bev proton in an emulsion of standard composition with a root-mean-square grain diameter of about 0.17 micron (Ilford L-type emulsion). The average energy loss in a grain will be about 100 ev, and the proton will encounter about 440 grains in this element of path. Now, whereas in about 190 such traversals the proton will lose more than 100 ev, in about 19 the energy loss will exceed 1000 ev. Therefore, at least in fine-grain emulsions, a large fraction of the grains rendered developable at the minimum of ionization can be attributed to the relatively rare traversals with large energy transfers.

3. Developability Coefficients and the Grain Density

When a charged particle penetrates a silver halide crystal it transfers energy to electrons until, after a certain such collision, the crystal may for the first time be in a developable condition. This point may be called the conception point.

We assume that in any element of path the probability for such an act that of itself is sufficient to render the crystal developable depends on the product of three factors: s, a measure of the local sensitivity and the degree of development; $\mathcal{J}''$, the effective rate of energy loss; and dy, the element of path length. First one treats the simplest model that may have some validity: A probability ψ is defined that a crystal of diameter D remain undevelopable after being penetrated a distance y by a charged particle with an effective rate of energy loss $\mathcal{J}''$. One assumes

$$d\psi = -\psi s \mathcal{J}'' \, dy$$

or

$$\psi = \exp(-S\mathcal{J}'' y). \tag{30}$$

The probability G that the crystal be completely traversed without being rendered developable then is

$$G = \exp(-s\mathcal{J}'' \delta), \tag{31}$$

where δ is the length of the track segment in the crystal.

In effect, this model assumes that there is a mean free path, $(s\mathcal{J}'')^{-1}$, for developability. It is unlikely, however, that matters are that simple. A mean free path for developability will exist only if the rendering of a crystal developable by an electron collision is solely determined by this event, and is not influenced by a cumulative conditioning effect in the crystal brought about by the prior passage of the charged particle through parts of that crystal or surrounding matter.

It is also unlikely that s can be independent of position in the crystal. Bogomolov has put forward evidence that the sensitive volume of a crystal is a thin surface layer.[11] Certainly surface sensitivity centers are more accessible to the reducing action of the developing agent, and such centers probably are more abundant on the surface. The migration distance of electrons and silver ions is not known.

The true physical situation cannot be stated precisely at present, and in a general theory one must simply make appropriate allowance for any reasonable form the facts ultimately may be found to assume. This is done by the introduction of free parameters, the presence of which in the theory will offer the possibility of their experimental evaluation.

If a radial variation of sensitivity is to be taken into account, then the traversal of a crystal of diameter D with a path segment in the crystal of length δ would lead to a probability $\exp\left[-\mathcal{J}'' s \delta f\left(\frac{D-\delta}{b}\right)\right]$ that the crystal would not be rendered developable. Here b is a characteristic length describing how rapidly the sensitivity varies with radius, and the function f is unknown

except that it is expected to increase with an increasing argument. Since one cannot in general overlook the possibility of cumulative effects, as mentioned above, the quantity $\mathcal{J}''$ s $\delta f(\frac{D-\delta}{b})$ must be multiplied by still another function of δ.

One must also now introduce a second characteristic length, a, which has the significance that the effect of the passage of a charged particle through a segment of the crystal will have little effect on conception points more remote than the distance a.

Then to allow for all these possibilities one expresses the probability of nondevelopability, G, by the following series:

$$G = 1 - \sum_{i=1}^{\infty} \sum_{j=0}^{\infty} \sum_{k=0}^{\infty} A_{ijk} \left(\frac{D}{b}\right)^{j} \left(\frac{\delta}{a}\right)^{k} (\delta s \mathcal{J}'')^{i} . \qquad (32)$$

This may be compared with the simpler form, Eq. (31), obtained by using the mean-free-path model. This is

$$G = \exp(-\delta s \mathcal{J}'') = 1 - \sum_{i=1}^{\infty} \frac{(-1)^{i-1}}{i!} (\delta s \mathcal{J}'')^{i} .$$

From Eq. (32) the total cross section, Ω, for developability of a crystal can be calculated:

$$\Omega = \pi/2 \int_0^D (1-G)\, \delta d\delta \qquad (33)$$

or

$$\Omega = \frac{\pi}{2} \sum_{i=1}^{\infty} \sum_{j=0}^{\infty} \sum_{k=0}^{\infty} \frac{A_{ijk}}{i+k+2} D^{L+j+k+2} b^{-j} a^{-k} (s\mathcal{J}'')^{i} . \qquad (34)$$

The true mean grain density, g, in a track then is also calculable:

$$g = N \int_0^{\infty} \Omega\, F(D)\, dD = \frac{\pi N}{2} \int_0^{\infty} F(D) dD \int_0^D \delta(1-G) d\delta \qquad (35)$$

or

$$g = \frac{3C}{\langle D^3 \rangle} \sum_{i=1}^{\infty} \sum_{j=0}^{\infty} \sum_{k=0}^{\infty} \frac{A_{ijk} \langle D^{i+j+k+2} \rangle a^{-k} b^{-j} (s\mathcal{J}'')^i}{i + k + 2} . \qquad (36)$$

For a particular emulsion type, the moments $\langle D^\mu \rangle$ of the diameter distribution may be established by a grain-diameter analysis. The coefficients A_{i00} can be identified with the quantities $\frac{(-1)^{i-1}}{i!}$. The product $s\mathcal{J}''$ must be expressed in reciprocal length units; s is therefore a reciprocal energy.

The remainder of the coefficients is to be determined from experiment, but they are dimensionless and assumed to be independent of D and $\mathcal{J}''$; s and $\mathcal{J}''$ appear only in the product relationship. The coefficients can be studied by measuring g while varying $\mathcal{J}''$, s, and the $\langle D^\mu \rangle$ separately. The sensitivity s is maintained constant when the crystal precipitation, sensitization, and development procedures are fixed. It is not known how varying these procedures affects the quantities a, b, and A_{ijk}. In the Ilford G, K, and L series of emulsions the standard composition remains the same in all, and the grain-size distribution in each series is kept constant, but is different for each series. The sensitivity varies through each series, being maximum for sensitivity 5-emulsion and decreasing as one goes through the K series, for example, from K.5 to K.4 to K.3, etc. The concentration of each emulsion type can also be varied. It appears that these emulsions might be suitable for investigations on the cross section for developability of a silver halide crystal. The variable $\mathcal{J}''$ is of course under the control of the investigator, as is the physical and chemical environment.

4. Structure of the Developed Track

Whatever its form, one can symbolize by

$$G = G(\delta, D, b, a, s\mathcal{J}'')$$

the probability that crystal C will not develop when traversed by a charged particle. The quantity

$$\frac{\pi N (1 - G) \delta \, d\delta \, F(D) \, dD}{2g} , \qquad (36)$$

then, is the probability that any particular crystal encountered will be of class C and will be rendered developable by the charged particle.

Now the probability that a crystal of class C' in the interval of particle path $d\mu$ will also be rendered developable is

$$\frac{\pi^2 N^2}{4g}\ \delta\,\delta'\,d\delta\,d\delta'\,(1-G)\,(1-G')\,F(D)\,F(D')\,d\,D\,d\,D'\,d\mu\,.$$

Then the probability is q that the particle render crystal C developable and traverse the emulsion a distance exceeding μ without rendering developable a crystal of class C'. The probability is found from

$$\frac{1}{q}\frac{dq}{d\mu} = -\frac{\pi^2 N_0^2}{4g}\ \delta\,\delta'\,d\delta\,d\delta'\,(1-G)\,(1-G')\,F(D)\,F(D')\,d\,D\,d\,D'. \tag{37}$$

One now is in a position to calculate the probability Q, that the particle, after rendering a crystal developable, may go a distance exceeding μ without creating developability in any other crystal. This is formulated

$$\frac{1}{Q}\frac{dQ}{d\mu} = -\frac{\pi^2 N^2}{4g}\iint \delta\,\delta'\,d\delta\,d\delta'\,(1-G)\,(1-G')\,F(D)\,F(D')\,dD\,dD', \tag{38}$$

so that, as P was calculated in Eq. 18:

$$Q = \exp\left\{-\frac{\pi^2 N^2}{4g}\iiint \delta\,\delta'\,d\delta\,d\delta'\,(1-G)\,(1-G')\,F(D)\,F(D')\,dD\,dD'\,d\mu\right\}. \tag{39}$$

The variance of the spacing of developed grains then is found from

$$-\int_0^\infty \mu^2\,\frac{dQ}{d\mu}\,d\mu - \left[\int_0^\infty \mu\,\frac{dQ}{d\mu}\,d\mu\right]^2. \tag{40}$$

This quantity, like g, is not directly observable because indefinitely small grain spacings are included, and many of the grains cannot be separately resolved with a microscope. A more useful approach is to study the gaps in the track.

Suppose that when the developed grains are projected on the particle trajectory a gap exists between C and C' only for $\mu > \frac{D}{2} + \frac{D'}{2} + \frac{\epsilon}{2} + \frac{\epsilon'}{2}$; where ϵ and ϵ' (the enlargements of grains C and C') are functions of their respective grain diameters D and D', but also may contain additive terms which vary with the optical conditions. This differs from the rule introduced by Fowler and Perkins[13] chiefly by allowing for a distribution of grain sizes.

After development of crystals C and C' a gap of length $\ell = \mu - \frac{D}{2} - \frac{D'}{2} - \frac{\epsilon}{2} - \frac{\epsilon'}{2}$ is left between them. Therefore, on integrating Eq. (37) with respect to μ, one has, for the probability that a gap with a length exceeding ℓ will be found between C and C',

$$\exp\left\{-\frac{\pi^2 N^2}{4g}\left(\ell + \frac{D}{2} + \frac{D'}{2} + \frac{\epsilon}{2} + \frac{\epsilon'}{2}\right)\left[\delta\,(1-G)\,d\delta \cdot \delta'\,(1-G')\,d\delta'\right]\right.$$

$$\left. \cdot\, F(D)\, F(D')\, d\,D\, d\,D' \right\} . \qquad (41)$$

(In order to facilitate comparisons, we shall use the notation of Fowler and Perkins[13] for important gap-density quantities).

Considering crystals of all classes, the density H of gaps with lengths exceeding ℓ is

$$H = g \exp\left\{-\frac{\pi^2 N^2}{4g}\left[\int_0^\infty (D+\epsilon)\,F(D)\,d\,D \int_0^D \delta(1-G)d\delta\right]\left[\int_0^\infty F(D)dD \int_0^D \delta(1-G)d\delta\right]\right.$$

$$\left. -\frac{\pi^2 N^2 \ell}{4g}\left[\int_0^\infty F(D)\,d\,D \int_0^D \delta\,(1-G)\,d\,\delta\right]^2\right\} . \qquad (42)$$

This can be written

$$H = B \exp(-g\ell) , \qquad (43)$$

with

$$B = g \exp\left\{-\frac{\pi^2 N^2}{4g}\left[\int_0^\infty (D+\epsilon)F(D)\,d\,D \int_0^D \delta\,(1-G)\,d\delta\right]\left[\int_0^\infty F(D)dD \int_0^D \delta(1-G)d\delta\right]\right.$$

and

$$g = \frac{\pi N}{2} \int_0^\infty F(D)\, d\, D \int_0^D \delta\, (1 - G)\, d\delta \ ,$$

the expression giving the true grain density.

The form of these equations displays two important new results. The gap-length distribution is exponential, and the true grain density and the gap-length coefficient are the same quantity.

The first result was shown empirically by O'Ceallaigh to be true over a range of gap lengths.[12] Fowler and Perkins extended the interval over which it has been tested, but the complete validity of the exponential gap distribution has not previously been demonstrated nor has the gap-length coefficient been shown to be the true grain density.

The quantity B is also known as the "blob" density because it is the density of clusters of unresolved grains in the track. It is of course numerically equal to the gap density.

If

$$\overline{D} = \frac{\int_0^\infty (D+\epsilon)F(D)\, d\, D \int_0^D \delta\, (1 - G)\, d\, \delta}{\int_0^\infty F(D)\, d\, D \int_0^D \delta\, (1 - G)\, d\, \delta}$$

is the average diameter of a developed grain, then the blob density B can be written

$$B = g \exp (- g \overline{D}) . \qquad (44)$$

$\overline{D}$ is therefore to be identified with the quantity a defined by Fowler and Perkins. It should be noted, however, that $\overline{D}$ has not a purely geometric meaning unless the grain-size distribution is very narrow. In general it will be slightly dependent on g. The mean gap length, $\langle G \rangle$, is found from $\langle G \rangle = g^{-1}$.

The lacunarity L of a track segment is the mean linear fraction of it that is occupied by gaps. From Eq. (44) this is

$$L = \exp (- g \overline{D}) . \qquad (45)$$

It may be noted that this is a very sensitive function of g, and that it remains useful when g is large.

The variance σ_B^2 of the blob density, B, can also be calculated in good approximation. It is

$$\sigma_B^2 = B\left[(1 - L + L\ell nL)^2 + L^2\right] . \qquad (46)$$

Some remarks on the effects of secondary ionization are required because under extreme conditions the secondary ionization influences the gap-length coefficient. Consider a cylindrical volume, the axis of which is the particle path and the radius of which is defined by the consideration that any grain whose center lies within this distance from the particle path is considered a track grain. Now, the linear density of developed grains within this cylinder, irrespective of whether they are grains traversed by the moving particle or grains rendered developable by secondary effects, will be the observed gap-length coefficient. Consequently, if the particle velocity is high enough for delta rays to be produced, and the emulsion is of a type that is sensitive to electrons, the saturation value of the gap-length coefficient can be expected to exceed n, the total linear density of grains traversed. An interesting example of this effect was reported by Fowler and Perkins,[12] who from the tracks of relativistic magnesium and silicon nuclei in G.5 emulsion found a saturation value of the gap-length coefficient equal to 5 per micron, whereas n in G.5 emulsion is only about 3 per micron.

A further remark on experimental results should also be made. Whereas all observers find an exponential distribution of long gap lengths, a number of investigators[22] report deviations at short gap lengths contrary to the data of Fowler and Perkins and the general theoretical result obtained here. In the published cases it seems as if the log frequency vs. gap-length curves might be straight lines if a few tenths of a micron were to be subtracted from all gap lengths (exclusive of zero which corresponds to the blob density). The possibility that the effect is an optical one suggests itself. What one sees is the superposition of grain diffraction patterns, and the measured length of short gaps can be strongly affected by the microscope numerical aperature, as well as light wave length and intensity. In addition, when the length of the gap is measured by the length of time that a key is depressed, as in many automatic instruments, the observer reaction time, the width of the hair-line in the eyepiece and the instrument reaction time also may introduce an additive constant to the gap lengths. If the effect is optical, it may be simply that a gap between C and C' is not recognized as such unless the distance between centers exceeds $\frac{D+D' + \epsilon + \epsilon'}{2} + \ell_0$, but when the distance is greater than this, say $\frac{D + D' + \epsilon + \epsilon'}{2} + \ell$, the gap length is measured to be ℓ. Then the gap (blob) density is really the density of gaps whose lengths exceed ℓ_0.

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