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Comments on the Statistical Distribution of Grain Sizes and Grain Density in G-5 Nuclear Emulsions

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Abstract

This work presents an experimental study of the statistical distribution of the size and density of grains along minimum ionizing π^- tracks in nuclear emulsions. Distribution parameters are computed, and an explanation is given for the departure of the size distribution curve from a normal curve. A quantitative explanation of the magnitude of the experimentally observed minimum grain density is also suggested. The experimental observations agree with the theory.

1. Introduction

Several workers (Von Ardenne and Angrew 1940; Hall and Schoen 1941; Knowles and Demers 1947; Baroni and Castagnoli 1950; Winnard 1951; Pickup 1953; Kumar 1959) have used electron micrographs of processed and unprocessed emulsion-grains of different types in order to study the shape of the grains. The grain-size distribution in unprocessed Ilford G-5 emulsions has also been examined by Baroni and Castagnoli (1950) and Barkas (1963) but, besides presenting a few statistical attributes of the emulsion grains, no-one has given details of the size and density distributions. Pickup described only the size histogram of the unprocessed G-5 emulsion grains, and it is quite clear from his work that the diameters of single grains fluctuate about a mean value, thus indicating that they do not all have equal diameters. A similar conclusion was reached by Della Corte *et al.* (1953*a*) with regard to processed single grain-groups. Kumar concluded that the size distribution for the silver grains in processed emulsion was less regular than that for the silver bromide grains in unprocessed emulsions. The reason for this may have been the inclusion of the smaller background grains together with those forming the tracks in the emulsion. The present work was thus undertaken to provide a detailed statistical study of both the undeveloped silver bromide grains and the developed silver grains along the paths of relativistic minimum-ionizing tracks in G-5 emulsions exposed to a 4 GeV/c π^- beam.

2. Experimental Details

(*a*) *Measurements of Grain Size in Processed G-5 Emulsions*

The microscope is an effective instrument for revealing the shapes as well as the size distribution of small particles. For the present measurements of silver grains, a Cooke optical microscope (40000 series) was used. The microscope has an oil immersion objective of 95× (numerical aperture $d_{NA} = 1{\cdot}3$) and an eyepiece of 15× carrying a fine-scale micrometer (goniometer) with a small drum having 100

divisions on its circular scale. The least count of the circular scale is 62·5 nm. The microscope is illuminated with filtered green light of wavelength $\lambda \approx 5200$ Å, and hence the minimum distance resolvable is 244 nm ($\approx 0{\cdot}61\lambda/d_{NA}$).

The grain-size measurements were made on π^- minimum-ionizing relativistic tracks of 4 GeV/c momentum (see Fig. 1). Several flat tracks (dip angle $\leqslant 5°$) were examined under a total magnification of 2138 ($95 \times 15 \times 1{\cdot}5$) by dividing them into successive segments of 50 μm length. Special precautions were taken in the measurements of the sizes of the grains in these segments. The segments were carefully aligned with the x motion of the movable crosswire of the micrometer scale.

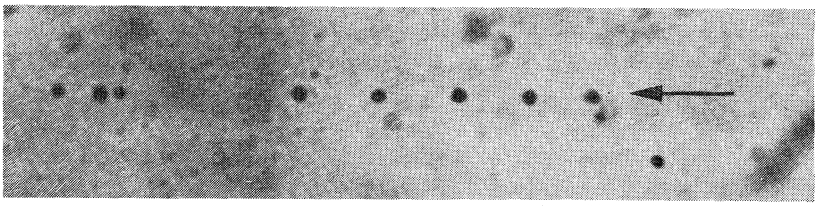

Fig. 1. Enlarged photomicrograph of developed silver grains along a π^- beam track.

Measurements of the linear sizes of grains were then made along this direction and along the mutually perpendicular direction to determine the dimensions l_1 and l_2 respectively. These dimensions aided in estimating the percentage occurrence of 'blobs' (i.e. grain groups) among the data, by means of the following criteria:

$l_1/l_2 < 1{\cdot}5$, object is a single grain; $\quad l_1/l_2 \geqslant 1{\cdot}5$, object is a blob.

Confirmation of these identifications was made by displacing the focal plane in the z direction and noting the contours or attachments of the grains. It was found that nearly 17% of the data were comprised of blobs, while the remaining 83% were discrete single grains. The mean size of a grain or blob was taken to be $\frac{1}{2}(l_1+l_2)$.

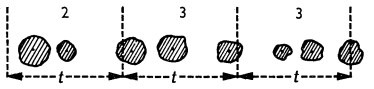

Fig. 2. Illustration of the assignment of grains (counts per cell are indicated) into cells of constant length t.

(*b*) *Measurement of Grain Density*

The grain density distribution was determined by counting the number of grains/groups per 50 μm segment, using the following conventions:

(1) A grain/group falling near the end of a segment was counted in that segment only if its centre is located in the segment (see Fig. 2).

(2) The grains/groups were counted irrespective of their sizes, most of them being single grains, with a few being groups of two, three or more grains.

In order to minimize errors, the measurements on each track were repeated several times.

(*c*) *Experimental Results*

Fig. 3*a* shows the size distribution for silver bromide grains in unprocessed G-5 emulsion as obtained by Pickup (1953), while Fig. 3*b* shows the corresponding

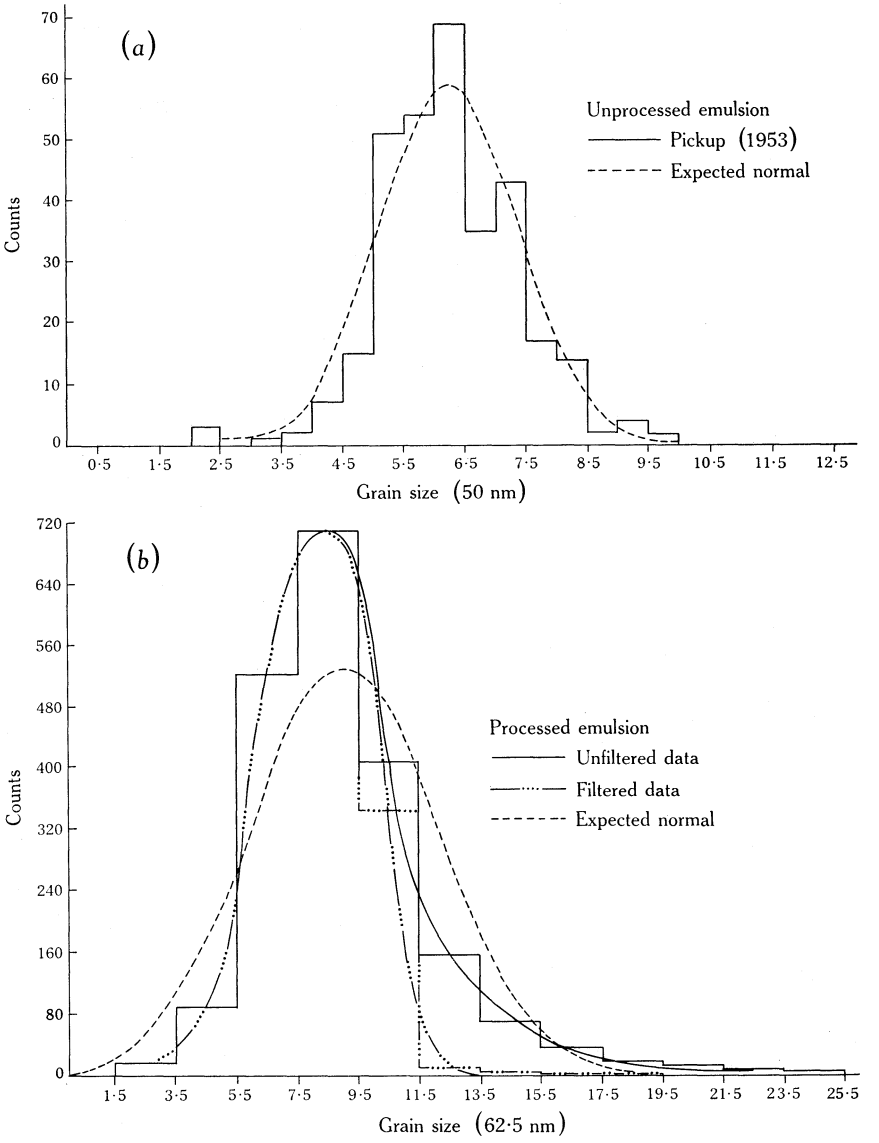

Fig. 3. Experimental size distributions for (*a*) unprocessed silver bromide grains and (*b*) processed silver grains in Ilford G-5 emulsions. The data are (*a*) those of Pickup (1953) and (*b*) the present results. The effect of filtration of blobs from the data is shown in (*b*).

distribution for processed grains as obtained in our measurements. In Fig. 3*b*, smoothed frequency curves corresponding to the histograms are also given, and in both figures a comparison is made with the expected normal distribution (dashed curve). Also, in Fig. 3*b*, the effect of filtration of the blobs (continuous lines, unfiltered data; dot–dash lines, filtered data) may be seen. Owing to a resolution limit for the microscope of about 244 nm, the size distributions in Fig. 3*b* are essentially invalid below about 4 divisions of the grain-size scale.

Measured values of the statistical parameters of the data plotted in Figs 3*a* and 3*b* are given in Table 1. For a normal distribution, the expected values of the mean, mode and median are identical, while the expected values of the coefficients of skewness and kurtosis are 0 and 3 respectively. From Table 1 we infer that the size distribution for the unprocessed grains follows a normal curve, while that for the processed grains does not and is leptokurtic.

Table 1. Comparison of size distributions for processed and processed grains

Statistical parameter	Unprocessed data	Processed data	
		Unfiltered	Filtered
Mean (μm)	0·31	0·566	0·51
Mode (μm)	0·315	0·517	0·511
Median (μm)	0·3101	0·539	0·505
Standard deviation (μm)	0·05615	0·187	0·119
Skewness	0·1361	1·575	0·187
Kurtosis	4·26	7·560	6·82

Analysis of the developed grain/group distribution along the π^- beam tracks yields the following results: a total of 2042 grains of mean diameter 566 nm were found in a total track length L of 10·4 mm, giving a mean number $\bar{N}$ of grains per 50 μm of 9·82 and a variance σ^2 for the density distribution of 8·32. The latter results correspond to the experimental grain density histogram shown in Fig. 4.

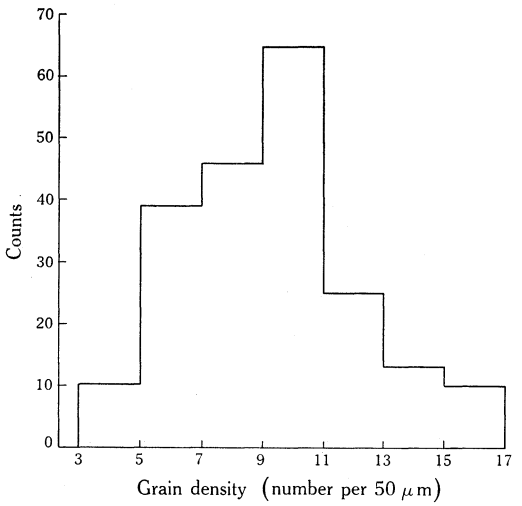

Fig. 4. Frequency histogram of the observed grain density in developed G-5 emulsion.

3. Theory

The process which renders a grain developable is strictly related to the specific ionization of the charged particle and the intrinsic characteristics of the AgBr grains. The calculation of grain density involves a parameter Π called 'the probability of development of a grain'. Many workers (Demers 1947; Della Corte *et al.* 1953*b*; Fowler and Perkin 1955; Herz and Davis 1955; Sharma and Gill 1962; Sharma

and Gaur 1968) have calculated this parameter in different ways. Demers (1947) gave the following relation:

$$\Pi = 1 - 2\lambda^{-2}\{1 - \exp(-\lambda) - \lambda \exp(-\lambda)\}, \tag{1}$$

where $\lambda = \alpha' x_0$, with x_0 defined as the total number of electrons produced within the grain due to the passage of an ionizing particle, and α' a constant characteristic of the type of the emulsion. The modified form of the relation (1) given by Della Corte *et al.* (1953*b*) is

$$\Pi = 1 - 2\lambda_0^{-2}\{1 - \exp(-\lambda_0) - \lambda_0 \exp(-\lambda_0)\}, \tag{2}$$

where

$$\lambda_0 = 4{\cdot}18 \times 10^{-3} n_t(\infty), \tag{3}$$

with $n_t(\infty)$ being the effective number of electrons produced within the grain due to the traversal of the charged particle through it. This number is always less than the total number of electrons produced by the charged particle within a grain, owing to

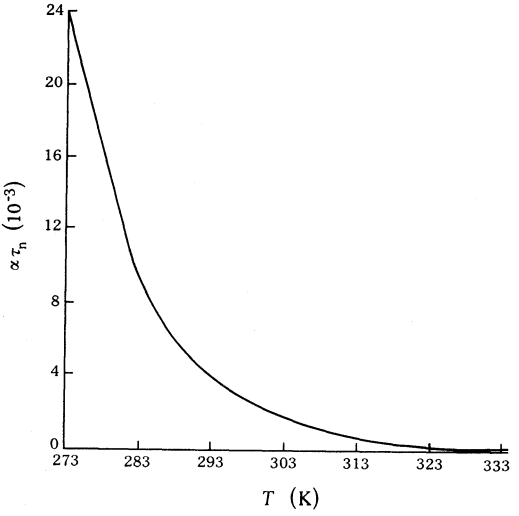

Fig. 5. Variation of $\alpha\tau_n$ with temperature T in the vicinity of room temperature.

neutralization through the recombination process. Singh and Sharma (1973) calculated $n_t(\infty)$ in terms of n_0, the maximum number of electrons produced in the grain, from the following relation:

$$n_t(\infty) = n_0 \exp\{-\alpha\tau_n n_t(\infty)\}, \tag{4}$$

where

$$n_0 = 46{\cdot}86\, dE/dR\,. \tag{5}$$

Here α is the rate constant for the recombination of electrons and holes present in the grain at any instant just after the exposure of the emulsion (at room temperature (300 K) we have $\alpha = 7{\cdot}7 \times 10^4\,s^{-1}$), τ_n is the mean ionic neutralization time, i.e. the time for the neutralization of an electron and Ag^+ ion at the trapping centre, and dE/dR is the specific ionization (the energy loss per path length) in GeV m^{-1}.

The temperature dependence of $\alpha\tau_n$ may be expressed as

$$\alpha\tau_n = \frac{\alpha}{C_e} = \frac{B_0 T \exp\{-(6{\cdot}43 - 0{\cdot}34/kT)\}}{\sqrt{2}\exp(-G_i/kT) + \exp(-G_F/2kT)}, \tag{6}$$

where $B_0 = 5{\cdot}336 \times 10^{-14}\,\mathrm{K}^{-1}$, G_i and G_F are the free formation energies for an interstitial and a Frenkel defect respectively, k is the Boltzmann constant in $\mathrm{eV\,K}^{-1}$, and C_e is a constant which depends on the concentration of interstitial Ag^+ ions and is given by $C_e = \tau_n^{-1}$ (Hamilton and Brady 1962; Y. T. Tan, personal communication).

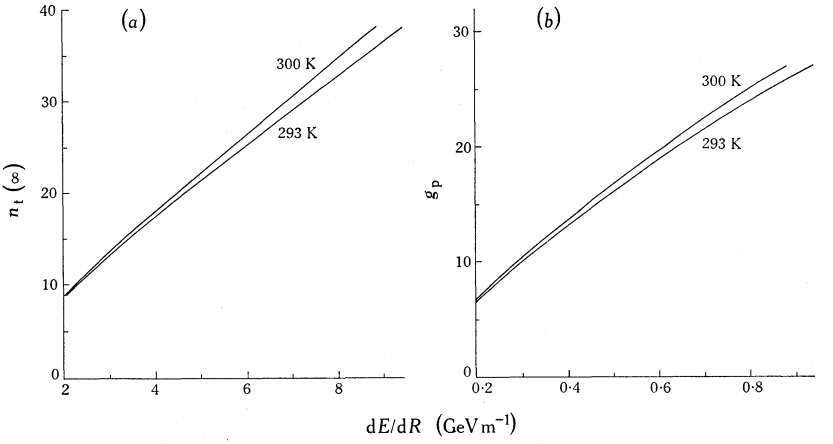

Fig. 6. Variation of (*a*) the effective number of electrons $n_t(\infty)$ and (*b*) the primary grain density g_p as functions of the specific ionization dE/dR (note that $\mathrm{GeV\,m^{-1}} \equiv \mathrm{keV}\,\mu\mathrm{m}^{-1}$).

A plot of $\alpha\tau_n$ as a function of temperature T in the vicinity of room temperature is shown in Fig. 5. For the present work we consider only room temperature calculations, as the emulsion plates used by us were exposed under that condition. Representative values for $\alpha\tau_n$ are $2{\cdot}229 \times 10^{-3}$ and $3{\cdot}982 \times 10^{-3}$ at 300 and 293 K respectively. Thus, from equations (4) and (5) we obtain

$$n_t(\infty) = 46{\cdot}86(dE/dR)\exp\{-2{\cdot}229 \times 10^{-3} n.(\infty)\} \quad \text{for} \quad T = 300\ \mathrm{K}, \tag{7a}$$

$$= 46{\cdot}86(dE/dR)\exp\{-3{\cdot}982 \times 10^{-3} n_t(\infty)\} \quad \text{for} \quad T = 293\ \mathrm{K}. \tag{7b}$$

Plots of these equations are shown in Fig. 6*a*. A graphical solution for $n_t(\infty)$ at a particular value of dE/dR may be read from either of these curves. Substituting the resulting value of $n_t(\infty)$ into equation (3) yields the corresponding value of λ_0, and hence the value of the probability of development Π from equation (2).

The number N of AgBr grains per 100 μm path length in G-5 unprocessed emulsion is 275 (Voyvodic 1950; Sharma and Gill 1962; Gaur and Sharma 1970). This gives the primary grain density g_p as

$$g_p = N\Pi = 275[1 - 2\lambda_0^{-2}\{1 - \exp(-\lambda_0) - \lambda_0 \exp(-\lambda_0)\}]. \tag{8}$$

Values of g_p, computed from equation (8), are plotted in Fig. 6*b* against corresponding values of dE/dR for both $T = 300$ and 293 K.

Apart from the primary grains (those directly affected by the charged particle), a few other grains (secondary grains) are also developed due to outgoing δ-rays. This secondary grain density g_s also contributes to the total grain density g, so that we have

$$g = g_p + g_s .$$

We are interested here only in grain density values at minimum ionization. Gaur (1973) concluded that the contribution of secondary grains was $\sim 10\%$, which is in agreement with the results of Nicoletta *et al.* (1967), while according to Patrick and Barkas (1962) this contribution is $\sim 12{\cdot}5\%$*. Therefore, we have

$$g = g_p + 0{\cdot}125 g$$

for the observed mean value of g. Thus our experimental value of 19·64 grains per 100 μm gives $g_p = 17{\cdot}2$ grains per 100 μm. For a primary grain density at minimum ionization of this size, the corresponding values of dE/dR (as obtained from Fig. 6*b*) are 0·52 and 0·545 GeV m^{-1} at 300 and 293 K respectively. These are very reasonable values of energy losses associated with the production of an observable grain density of around 20 grains per 100 μm. Conversely, given the value of dE/dR, the corresponding value of the experimental grain density can easily be calculated.

4. Results and Conclusions

We have seen in Section 2*c* that the size distribution for the unprocessed grains corresponds to a normal distribution, while that for the processed grains shows some departure from a normal distribution. Using the results listed in Table 1, we are led to the following conclusions:

(i) For our measurements we have used the developed grains created by the passage of a minimum ionizing (relativistic) charged particle of very high momentum. As the specific energy loss in these grains is very small, secondary ionization is least expected for such exposures and the clustering of grains due to secondary ionization is considered to be extremely small. There may also be a possibility that the background grains may contribute significantly to the observed clustering. As noted in Section 2*c*, the variance of the density distribution σ^2 is seen to be slightly smaller than $\bar{N}$, the mean number of grains per 50 μm. This also indicates the small percentage of grain groups that are present. Since the clustered grains have been filtered out, the size distribution (dot–dash curve in Fig. 3*b*) may in turn be considered to arise from single grains (within the limits of microscope resolution).

(ii) The grains in the unprocessed emulsions have a random distribution and are of varying sizes. Hence the developed grains in the emulsion are also found to have varying sizes with lesser variance. Values for the development or growth factor γ, defined as the ratio of the mean size of developed grains to the mean size of undeveloped grains for G-5 emulsions, are compared in Table 2.

* Patrick and Barkas (1962) made a numerical error in obtaining their expression for A. The coefficient of the $W^{7/6}$ term in equation (9) of their paper should be $6{\cdot}6\times10^{-2}$ rather than $1{\cdot}1\times10^{-2}$. The numerical value of A then becomes 1·9 instead of 3·9. This reduces their quoted value for the number of secondary grains at minimum ionization by $\frac{1}{2}$, that is, from 25% to 12·5%.

If the growth factor remained constant irrespective of the size of the undeveloped grains then we definitely would expect a normal distribution for the developed grains which is similar to that for unprocessed grains in nuclear emulsions. However, Fig. 3*b* clearly indicates that neither experimental distribution follows a normal distribution and that both have a common characteristic of high kurtosis. In order to explain this high kurtosis we conclude that the magnitude of γ varies for grains of different sizes, i.e. that the value of γ is less for large grains than for small. Consequently, most of the large grains contributing to the right-hand wing of the normal distribution have shifted toward the left after development. Similarly, the smaller grains contributing to the left-hand wing of the distribution have shifted toward the right after development.

Table 2. Comparison of growth factor measurements for G-5 emulsion grains

Data	Assumed grain size (unprocessed)	Estimated growth factor[a]
Della Corte *et al.* (1953*a*)	0·20 μm	3–4
Bizzetti and Della Corte (1959)	0·27 μm[b]	1·85
Skjeggestad (1958)	0·27 μm	2·2
Present work		
Filtered	0·27 μm	1·92
Unfiltered	0·27 μm	2·09
	0·31 μm[c]	1·83

[a] Generally accepted as $\gamma = 2$ by most workers.
[b] As stated by the manufacturers, Ilford.
[c] Based on data of Pickup (1953).

It is also evident from Fig. 3*b* that more than 80% of the total grains are always aggregated in a narrow size interval of from 5·5 to 11·5 divisions and that the wings of both distributions have been highly depressed by comparison with the those of the expected normal curve. It is also clear that the theoretically computed value for the grain density agrees well with our experimental observations.

Acknowledgments

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