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Original Paper

Dispersion of Latent Image Specks in Silver-salt Photographic Materials Formed by Exposure to an Electron Beam with Constant Energy

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Abstract: The system used to analyze the process of the formation of latent image specks (LISs) in silver-salt photographic materials due to radiation exposure to obtain the dispersion index, which is the number of LISs in a silver halide grain formed by one impact of high-energy particle, was improved. By using the electron beam from a transmission electron microscope, electrons of uniform energy could be produced for irradiation purposes. The number of electrons impacting a single silver halide grain along with the energy of each electron was controlled by varying the current density and the applied voltage. A new analysis method which included the probability that LIS may not be formed despite an impact of high-energy particle was proposed. The number of LISs was counted by the arrested development method. The probability and the dispersion index were estimated at approximately 0.5 and 1, respectively, for an unsensitized emulsion with cubic grains. This method will be useful in the design of new nuclear emulsion plates.

Key words: Silver-salt photography, Nuclear emulsion, Dispersion of latent image specks, Electron-beam exposure, Poisson distribution

1. Introduction

Nuclear emulsion plates incorporating silver-salt photographic materials have been used for the detection of radiation tracks in the search for elementary particles from the dawn of this form of research[1)~4)] up to the present day.[5)~11)] These plates possess several specific characteristics, such as high resolution and three-dimensional detector.

An understanding of the photosensitive process in silver-salt photographic materials, which corresponds to the formation process of latent image speck (LIS), is also important for radiation detection by using nuclear emulsion plates.[12)~14)] In the case of light exposure, LISs are formed by excited electrons generated by incident photons on a silver halide grain.[14)~16)] The excited electron is captured at a particular type of electron trap termed a sensitivity center and neutralized by an interstitial silver ion to form a silver atom. Repetition of this process produces an LIS consisting of silver atoms. These LISs act as a catalyst to reduce the silver halide grain to silver grain through the development process and an image consisting of silver grains appears.

This includes many stochastic processes, such as the time-interval distribution of excitation and the number distribution of excited electrons in each grain. Therefore, the analysis of the LIS formation process, including these distributions, becomes very complicated. An analysis of the sensitivity of these materials was carried out by measuring the number of incident photons and statistically analyzing the processes. Simulations were conducted by Hamilton[17)] and continued by Hailstone[18)] to analyze this stochastic process using Monte Carlo methods. Typically, because the first LIS acts as a strong electron trap and effectively gathers other excited electrons, only one LIS is formed in a grain. This is termed the concentration theory.[14), 19), 20)]

There are many other unknown factors associated with the formation process of LISs through radiation exposures, which have inspired attempts at analyzing the process.[21)~28)] A large number of electrons are excited by one impact of high-energy charged particle, and a plurality of LISs is often formed in one grain simultaneously in spite of the concentration theory. This phenomenon is called the dispersion of LISs[14), 20)]. Usually this brings about a decrease in sensitivity. Quantitative estimation of the degree of dispersion for each emulsion is very important because this characteristic is strongly affected by the properties of the emulsion.

As the former problem associated with the time interval distribution of excitation by photons is avoidable, radiation exposure would make the analysis of the formation process simple. In previous reports on the study of the LIS formation process due to radiation exposure[21)~24)], the distribution of the number of LISs formed on one silver halide grain was experimentally analyzed, and the mean number of high-energy charged-particles impacting one grain w was estimated using statistical analysis. The mean number of LISs v in one grain was obtained experimentally and the mean number of LISs z formed in one grain by one impact of charged particle was

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then calculated. This value z could be regarded as a dispersion index that indicates the degree of the dispersion of LISs.

The latter problem associated with the distribution of the number of excited electrons still remains because charged particles possess a distribution of energy and the number of electrons excited in each silver halide grain is different. This deteriorates the accuracy of the dispersion-index measurement. However, when electron-beam irradiation is used, the energy of each irradiated electron can be made uniform by adjusting the applied voltage. The energy transferred to each silver halide grain by one impact becomes nearly uniform, and the formation of LISs proceeds with an equivalent number of excited electrons in each grain. Consequently, the formation process becomes nearly identical, making the analysis simpler. A transmission electron microscope was used as a convenient apparatus for electron-beam irradiation and a new method was developed to analyze the LIS formation process. We analyzed the dispersion of LISs formed by the electron-beam irradiation and will report the results here.

2. Theory

The calculation process for z was based on the following two hypotheses.[21)~24)]

Hypothesis 1:

The process involving charged particles impacting each silver halide grain is a random process.

Hypothesis 2:

Because the energy lost by an impacting charged particle is very large, many electrons are excited at the instant of the impact. Consequently, LISs are always formed in each silver halide grain absorbing the energy of the impacting charged particles. Some reports have showed that a silver halide grain became developable after one impact of high-energy electron.[29),30)]

The distribution $q(m)$ of impact times m at each grain will obey the Poisson distribution following from Hypothesis 1, and can be given as follows:

$$q(m) = \frac{e^{-w} \cdot w^m}{m!} \tag{1}$$

The probability that a charged particle will not impact a grain is given by Eq. (2):

$$q(0) = e^{-w} \tag{2}$$

This can be regarded as the ratio of grains which do not have any LISs following from Hypothesis 2.

When the distribution $f(n)$ is set for the number of LISs n in one silver halide grain obtained experimentally, the ratio of grains $f(0)$, which do not have any LISs, is equal to $q(0)$. A method that can be used to count the number of LISs in one grain, termed as the arrested development technique, has already been proposed by many researchers[21)~24),31)~36)].

$$f(0) = q(0) = e^{-w} \tag{3}$$

Consequently, the value w can be estimated experimentally from the value $f(0)$.

$$w = -\ln\{f(0)\} \tag{4}$$

The mean number v of LISs in a grain formed by w charged particles, which impacts on a grain, can be determined experimentally and be used to yield v/w, which can provide the mean number z of LISs formed by one impact of charged particle. This z value can be obtained experimentally from the slope of the straight line through the plotted points of w and v for different radiation exposure values.

Previously, it was noted that LISs were always formed by one impact of charged particle on a silver halide grain, as described in Hypothesis 2. However, the value $f(0)$ often changed for materials with different sensitization levels in spite of being exposed to the same radiation. This suggests a case in which an LIS is not formed even though a charged particle impacts a grain. In this case $f(0)$ does not agree with $q(0)$. This would occur when the energy loss is small, leading to only a small number of electrons being excited, or when the recombination process between excited electrons and holes proceeds vigorously.

Therefore, the case in which an LIS is not formed, despite an impact of charged particle, should be taken into consideration. The probability that an LIS is not formed even when a charged particle impacts a grain is assumed to be k.

The mean number of events in which LISs are formed on a silver halide grain when charged particles impact the grain w times is assumed to be w^*. This value w^* should be significantly correlated with the experimental results. The distribution $r(l)$ of the number of events l in which LISs are formed is then considered. When the probability k is constant, formation events for LISs after electron impacts proceed randomly, and $r(l)$ can be described by the Poisson distribution in the same way as Eq. (1).

$$r(l) = \frac{e^{-w*} \times w*^{l}}{l!} \tag{5}$$

The ratio of non-formation of an LIS is given as $r(0)$.

$$r(0) = e^{-w*} \tag{6}$$

This equals $f(0)$, obtained experimentally.

$$w *= -\ln\{f(0)\} \tag{7}$$

The probability that an LIS is not formed even if m charged particles impact in sequence is k^m. Therefore, $f(0)$ becomes the sum with m of the ratio of non-formation of an LIS despite m charged particles impacting the grain.

$$f(0) = q(0) + kq(1) + k^2 q(2) + \cdots = \sum_{m=0}^{\infty} k^m q(m) \tag{8}$$

Because $q(m)$ is given by Eq. (1), this can be substituted into Eq. (8) as follows:

$$f(0) = \sum_{m=0}^{\infty} k^m \frac{e^{-w} w^m}{m!} = e^{-w} \sum_{m=0}^{\infty} \frac{(k \cdot w)^m}{m!} = e^{-(1-k)w} \tag{9}$$

Taylor expansion can be used for e^x. As $f(0)$ is equal to $r(0)$, this leads to the correlation between w and w^*.

$$w^* = (1 - k)w \tag{10}$$

In the case of irradiation from an electron beam with uniform applied voltage, each electron possesses a uniform energy. The energy of the electron E is given as $E = e \times V$, where V is applied voltage and e is the elementary charge. Furthermore, by adjusting the current density of the electron beam and arranging the grains with a mono-grain layer on the base, the mean number of electrons g impacting one silver halide grain can be controlled. The value g is given by Eq.(11), where I is the current density, S is the cross-section of the grain, and t is the irradiation time of electron beam.

$$g = \frac{I \times S \times t}{e} \tag{11}$$

The value g can be considered as being equal to w and the probability k can be estimated from the ratio of g and w^*. Both values can be obtained experimentally.

$$k = 1 - \frac{w*}{g} \tag{12}$$

This will provide information about the formation efficiency of LISs under exposure to electron-beam irradiation.

Because the dispersion index is given for the number of formed LISs, z^* is equal to the number of LISs when one event of the formation of some LISs proceeds.

$$z* = \frac{v}{w*} = \frac{v}{(1-k)g} \tag{13}$$

This z^* value expresses the real degree of the dispersion of LISs, even if an LIS is not formed with probability k at an impact of charged particle.

3. Experiment

The photographic emulsion used in this experiment was composed of pure silver bromide unsensitized cubic grains of 0.76 μm edge length. The emulsion was applied to a plastic film-base to form a mono-grain layer. Because there was no overlapping of the emulsion grains, each grain was exposed to the same amount of electron-beam irradiation. An image of this film surface taken using an atomic force microscope (AFM) is shown in Fig. 1. The grains do not overlap and the upper half of the grains projects from the gelatin layer, although the shape of the cube was not clear in this figure due to the thin, soft gelatin layer covering the grains which disturbed the scanning by AFM probe. Observation with a conventional optical microscope also indicated that all grains were arranged on the base with one face of the cube facing directly above. These images indicated that square density of the grains was 96000 grains/mm^2, and ratio of grains overlapping was only 0.004%. Consequently, the cross section of a grain was found to be $(0.76)^2$ μm^2 and the path length of an electron travelling through a grain was 0.76 μm.

Films coated the emulsion were set on film holders for a transmission electron microscope (JEOL, JEM 1200Ex) and the holders were set in the microscope. The emulsion grains were irradiated by an electron beam uniformly, because any observation sample was not set in the microscope. The applied voltages were 40, 60, or 80 kV, and one electron possessed an energy of 40, 60 or 80 keV. The variable g was adjusted by altering the current density of the electron beam. The irradiation characteristics of the electron beam are shown in Table 1.

LISs were detected using an arrested development method, performed with the arrested developer as described by Mifune[36]. Because the development rate was suppressed in this developer, only a small area around the LIS was developed to produce a developed silver cluster that was much smaller than the silver halide grain. The emulsion layer was dissolved after development to extract the silver halide grains, and carbon replicas of the grain were prepared. These replicas were observed with the same transmission electron microscope. The small, developed silver clusters in one silver halide grain were counted to obtain the number of LISs. An electron micrograph of the small developed silver clusters on the carbon replicas of the silver halide grains is shown in Fig. 2, where the black lumps are the developed silver clusters formed at each LIS. The clusters on one hundred or more grains were counted.

Table 1 Condition for the irradiation of electron beam

Current density (pA/cm^2)	3.5	17.7	17.7	17.7	17.7
Exposure period (second)	1	1	2	4	8
g	0.1	0.5	1	2	4

Cross section of the grain $=(0.76)^2$ μm^2

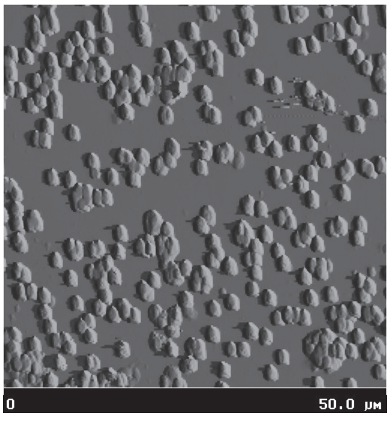

Fig. 1 AFM image of the surface of photographic film with mono-grain layer.

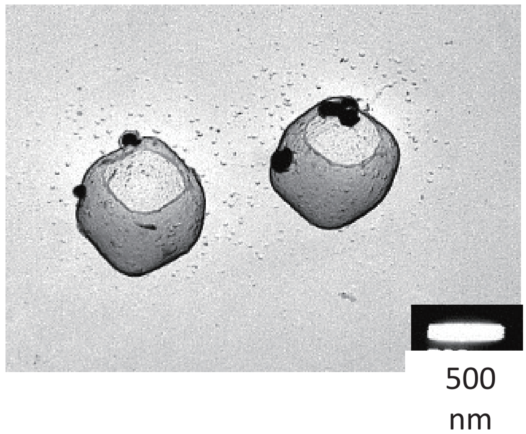

Fig. 2 Electron micrograph of small developed silver clusters on carbon replicas of silver halide grains. The black lumps are the developed silver clusters.

The mean number of LISs v on a grain and the distribution of the number $f(n)$ were obtained experimentally. The value w^* was then calculated from $f(0)$. The relationships between each w^* value and its corresponding v value were then plotted, and the value z^* was obtained from the slope of the straight line through these plotted points. The relationships between w^* and g obtained from the current density were also plotted, and the probability k was estimated from the slope of straight line through these plotted points.

4. Results

The distributions of the number of LISs in a grain $f(n)$ after exposure to an electron beam with an applied voltage of 40, 60 and 80 kV are shown in Fig. 3 for each g value. Values of v for each g are also shown in the figure. The distribution form increases in width along with an increase in g, thereby suggesting an increase in the number of LISs in one silver halide grain. In the case of previous radiation exposures[21)–24)], large numbers of LISs were formed by each impact of charged particle, and the distribution often showed two peaks at $n = 0$ and at $n > 1$. There was only one peak in this experiment and the shape of the distribution obeyed the Poisson distribution law with a high associated correlation coefficient.

The relationships between g and v are shown in Fig. 4 for each applied voltage. Plotting g versus v yielded a straight line, and the slopes of the straight-line graphs were all similar and were approximately 0.5 for all applied voltages, while the straight lines do not pass through the origin.

The relationships between g and w^* obtained from each $f(0)$ value are shown in Fig. 5 for each applied voltage. The plotted points formed a straight line but did not pass through the origin for any of the applied voltages. The gradient of the slope was approximately 0.5. These relationships were similar to those of g and v as shown in Fig. 4.

The relationships between w^* and v are shown in Fig. 6 for each applied voltage. The plotted points also formed a straight line. The correlation coefficients were all greater than 0.99, suggesting a good correlation. The line passed through the origin and v values were not saturated even when w^* increased above a value of one, while the v value found in previous results showed a tendency to be saturated with an increase in w^*.[21)–24)] The gradient of the slope, which represents the value z^*, was nearly unity for all applied voltages. This indicated that only one LIS was formed by an impact of single elec-

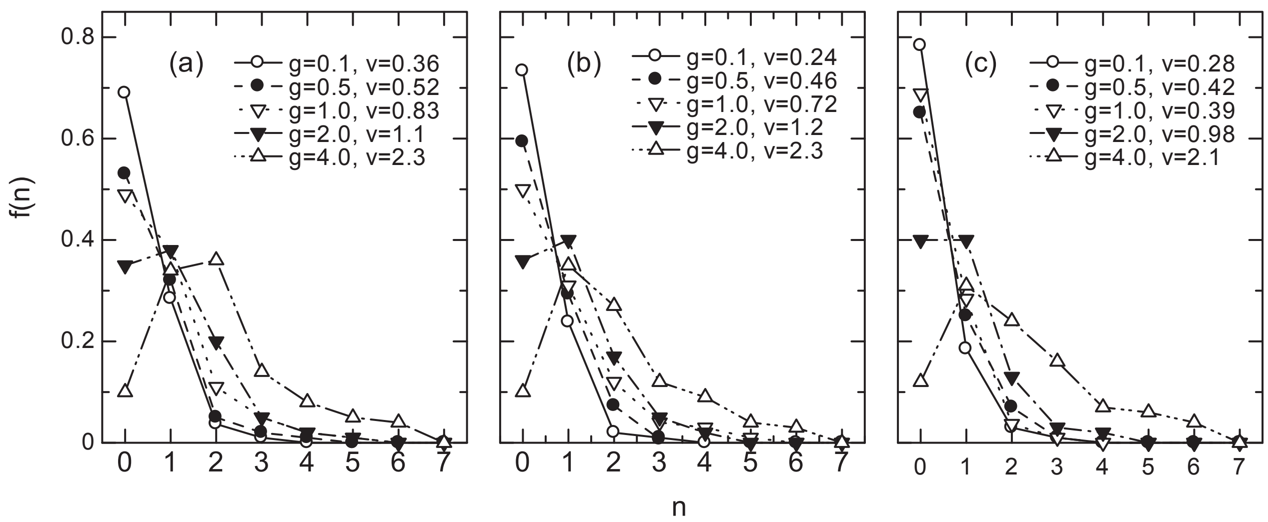

Fig. 3 The distribution of the number of LISs n on a silver halide grain for different g values, where g is the mean number of high-energy electrons impacting a grain, obtained from the current density of the electron beam, and v is the mean number of LISs on one grain. The applied voltage of electron beam was (a) 40 kV, (b) 60 kV, and (c) 80 kV.

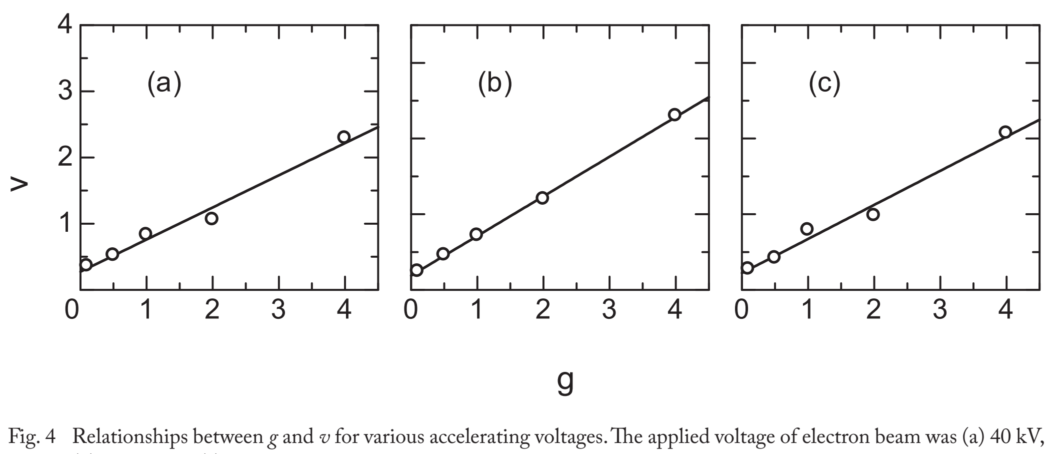

Fig. 4 Relationships between g and v for various accelerating voltages. The applied voltage of electron beam was (a) 40 kV, (b) 60 kV, and (c) 80 kV.

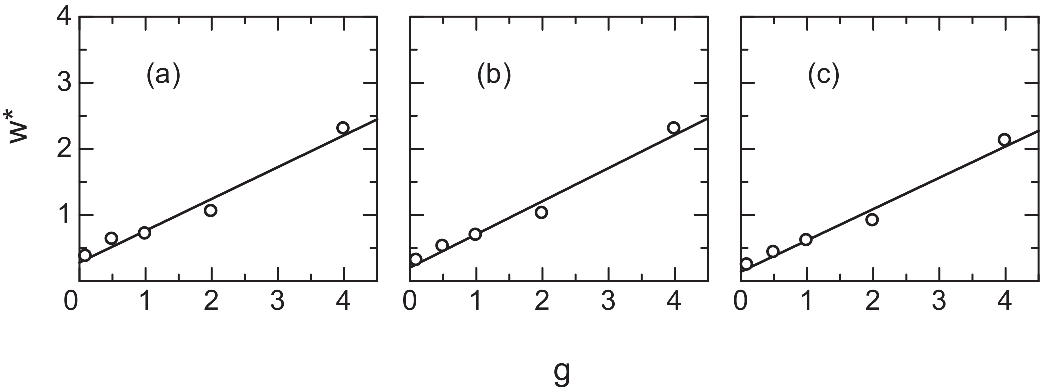

Fig. 5 Relationships between *g* and *w** for various accelerating voltages. The applied voltage of electron beam was (a) 40 kV, (b) 60 kV, and (c) 80 kV.

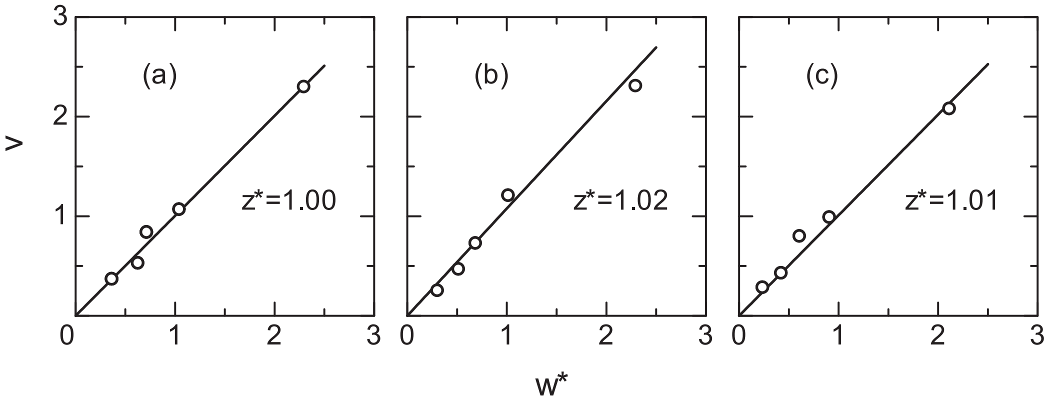

Fig. 6 Relationships between *w** and *v* for various accelerating voltages. The applied voltage of electron beam was (a) 40 kV, (b) 60 kV, and (c) 80 kV.

Table 2 Relations of g to v and w*, and z values for each applied voltage

Applied voltage (kV)	40	60	80
Relations of *g* to *v*	v = 0.49g+0.27	$v = 0.52g+0.19$	$v = 0.45g+0.22$
Relations of *g* to *w**	w* = 0.48g+0.28	w* = 0.50g+0.21	w* = 0.47g+0.14
z^*	1.0	1.0	1.0

tron. The relationships of *g* to *v* and *w**, and *z* values for each applied voltage are summarized in Table 2.

5. Discussion

The mean number of LIS formation events *w** in one silver halide grain due to impacts of high-energy electron was derived experimentally from $f(0)$ values at the distribution of LISs, which was a similar process used in previous experiments[21)–24)] involving radiation exposure. In this experiment, the mean number of electrons impacting one silver halide grain *g* was newly derived from the current density of the irradiating electron beam.

If LISs are always formed by an electron impact, the probability *k* becomes 0, and *w** agrees with *g*. However, the gradient of the slope of the straight line through the plotted points of *w** and *g* was approximately 0.5. The probability *k* was calculated to be approximately 0.5 from Eq. (12), and this indicated that approximately half of the high-energy electrons impacting a grain did not form an LIS.

The number of excited electrons in one silver halide grain was calculated from the line energy-loss in silver halide, the electron-pass in the grain, and the energy required to excite one electron in silver halide. The energy loss for an electron passing through a silver bromide cubic grain of 0.76 μm edge length as obtained from the Bethe' Equation[37] was 2.2 keV for an electron of energy of 40 keV, 1.7 keV for that of 60 keV, and 1.4 keV for that of 80 keV.

Ihama used an energy of 5.8 eV as the energy required to produce one excited electron due to the passage of a high-energy electron through silver bromide.[38] Using this value, the number of electrons generated by excitation was found to be approximately 380 at 40 keV, 290 at 60 keV, and 240 at 80 keV. Ihama estimated that the number of electrons excited by an electron of 30 keV was 500 while an electron of 1 MeV would excite 100 electrons in an octahedral grain whose circle-equivalent diameter was 0.8 μm. These estimations were similar to the results of this study.

There were some reports which indicated that the minimum of four silver atoms were required to make one developable LIS.[14),20),39)] The estimated number of excited electrons was sufficient to form developable LISs in all grains impacted by the electron beam. Consequently, there must be a large loss process associated with the excited electrons. The same number of holes is always generated at excitation, and recombination is the most probable loss process in the absence of any effective process to remove these holes. The emulsion used in this experiment was an unsensitized one, therefore, many of the formed holes would not be removed, and vigorous recombination would proceed in each grain, leading to half of the grains not being able to form any LIS. Prevention of the recombination would drastically increase the sensitivity to radiation exposure.

The value z^*, including the probability k, is, however, also useful as an index for the degree of dispersion of LISs. The result which showed that z^* was approximately equal to one suggested that only one latent image speck was formed when one electron impacted the grain despite a large amount of energy being provided to the grain. Previous results involving radiation exposure for emulsions with sulfur, sulfur-plus-gold, or reduction sensitization[21)~24)] showed that z yielded a value greater than one, depending on the type and level of sensitization, and a plurality of LISs was formed. On the other hand, z was found to be nearly equal to one for unsensitized emulsions with cubic grains. Similarly, the dispersion of LISs did not appear and only one LIS was formed by each impact of high-energy electron in this experiment, because the emulsion used was an unsensitized one composed of cubic grains. When an LIS is first formed, it attracts all excited electrons in the grain because there is no effective electron trap which can compete against the capture of excited electrons to form LIS. It is regarded as a characteristic phenomenon of unsensitized emulsions containing cubic grains that only one LIS is formed in a grain by one impact event. The concentration theory[14),19),20)] is consistent even for radiation exposure.

On the other hand, the plotted points of w^*-v in Fig. 6 were always on a straight line, even when the value w^* was larger than one. This suggested that new LISs were formed even after the second or third high-energy electron impacted the grain and even though LISs already existed in the grain. The formation of new LISs by subsequent impacts of electron proceeded in spite of the existence of some LISs. The concentration theory was therefore not effective in this case, and this leads to the following consideration. The working area of the concentration theory in a grain has certain limitations. When the grain size is larger than this area, new LISs can be formed in sites outside of this area, as previously proposed.[34] Therefore other LISs could be formed in other areas of the grains of 0.76 μm size as used in this experiment, which were larger than the 0.2 μm grain sizes used in the previous reports.[21)~24)] The distribution of $f(n)$ in Fig. 3 would obey the Poisson distribution, because the formation of LISs within specific areas in the grain was a random process. The validity of this consideration will be inspected as part of future investigations through the use of an emulsion with a smaller grain size.

Although the applied voltage was altered from 40 to 80 kV, the relationship between w^* and g for each voltage showed only a small variation. It was expected that w^* would depend on the applied voltage because the number of excited electrons changed as described above. The absence of such a phenomenon was considered as follows. The same number of holes as electrons is always formed by excitation and the total number of electron-hole pairs varies with varying applied voltage. On the other hand, there will be a limitation in terms of hole scavenging capacity to prevent recombination in silver halide grains. The number of electrons surviving recombination will become the same as the number of scavenged holes and be kept at the same level, even though the number of electron-hole pairs increases. This also explains similar values for the probability k for each applied voltage.

The slopes of the straight-lines in Fig. 4 or Fig. 5 do not pass through the origin. One possible reason is fog formation, because fog would increase v and w^* values uniformly and the line would deviate from the origin. However, the fog level of this emulsion was not so high due to the unsensitized one. Another reason is competition of surface and internal sensitivities. The size of this emulsion grain is enough large to have internal sensitivity, and the unsensitized emulsions often have high internal sensitivity.[40)~42)] Competition of excited electrons would occur between surface and internal sites. When the surface sensitivity is slightly dominant, LISs are formed on the surface preferentially at small g values, and the probability k becomes high. However, when internal LISs are formed following the increase of g value, the probability k decreases because the development method used this experiment cannot detect the internal LIS. Then, the slope becomes small and the line seems to deviate from the origin. This internal sensitivity could also affect to make the probability k smaller.

Due to the use of the electron-beam and the method described in this paper, handling of radioactive materials is not required, and the experimentation is more convenient than that using previously reported methods. Moreover, the precision of the LIS-dispersion-index measurement was higher, and the accuracy was improved in comparison with the previous radiation exposures as shown in the plots of w^*-v, which were well set on a straight line along with a high correlation coefficient.

These considerations suggest that the measurement of z^* values will be useful for the analysis of the LIS formation process. The characteristic differences in the emulsions can be estimated by comparing z^* values, and this will provide useful information for preparing nuclear emulsions with specific properties.

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