Application of Random Dot Model-to-Fog Granularity Caused by High-Energy Radiation of Silver Halide Emulsions in Color Systems

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Abstract

The random dot model has long been used as a basis to understand microscopic density variations present as noise, or granularity, in the silver halide-based photographic system. The simplest representation of this model assumes a random distribution of monodispersed disks with both area and absorptance. Density, and granularity, the noise in density, as measured through a small aperture, can be modeled as a function of parameters associated with these disks.

This paper presents an application of the random dot model to explain radiation-induced fog granularity effects on silver halide grains in single-layer coatings processed in color developer with dye-forming chemistry. For radiation fog, the number of disks should be proportional to radiation exposure and silver level. The radiation fogging mechanism tends to cause multiple development sites on affected grains. This mechanism yields a grain-size dependence on the model's disk size and absorptance.

Introduction

A significant limitation of the image recording property of film is intrinsic noise. The origin and methods for characterization of this noise, the visual sensation of which is called graininess, has been studied by photographic scientists for more than nine decades. Detailed references to the body of this work appear in several texts.[1]

Random Dot Model.
Early work on film noise focused on measurement systems and characterization. Elements of the random dot model were developed near the beginning of the last century. In its simplest form, the model approximates image density because it is composed of monodispersed disks with both size and light absorption characteristics. The model was developed to correlate the metrics of image density and image noise with the sizes and numbers of imaging centers. The random dot model initially proved useful for characterizing black and white films, although much of the developed silver image is filamentary an/or "cloudlike" in nature rather than monodispersed and disk-like. Extensions of the random-dot model have evaluated effects of grain-size distributions, grain crowding, layer thickness, and other important phenomena. Some of these effects are observed with increasing image density. Generally, for a sample having low density with widely spaced imaging "disks," the simple random dot model proves adequate.

Color systems.
Color photographic systems, in which the silver is removed and colored-dye clouds form the image, can be thought of as giving something that looks more disk-like than the black-and-white filamentary product of silver development. In three dimensions, the dye clouds can approach spherical or globular shape, and the size of the dye clouds may be dependent on the types and activities of the chemistries, development time, conditions and kinetics, competitive reactions, and the effectiveness of the ballasts on the dye molecule. Trebka[2] and Saunders[3] have offered treatments of color granularity.

Multilayer color films systems usually incorporate a variety of grain sizes overlapping across three color records. The image is formed from simultaneous competitive reactions, coupler starvation, chemical inhibition, and consequent variations in partial grain development. As a result, the multilayer system exhibits very complicated characteristics and is difficult to evaluate using the simplicity of the random dot model. Multilayers will not be discussed here.

Single-layer color systems offer a simplified and more analytically tractable alternative to multilayer systems.

Even for simple single-layer color systems, the random dot model needs to be modified at higher densities. At higher density, at least two mechanisms interfere: dye-cloud overlap and coupler starvation. Dye-cloud overlap affects the statistics of the simple random dot model. Starvation is the tendency for incorporated coupling systems to merge dye clouds and trend towards a uniform dye density wherever the quantity of developed silver far exceeds the quantity of dye-forming molecules.

At low density in a single-layer format with an abundance of color-forming, effectively ballasted coupler dye, the dye clouds should form nonoverlapping disks, and the random dot model should be valid.

Radiation.

The effect of ionizing radiation has been noted to yield high levels of fog and disproportionate levels of granularity.[4,5] The increased granularity has been ascribed to two effects: correlated grain development, from a single high-energy photon exposing multiple grains, also called "grain yield"; and multiple development sites per grain, also called "latent image dispersity." Broadhead[4] has indicated that latent image dispersity dominates at large grain sizes, and high grain yield dominates at small grain sizes. Broadhead has also indicated that both mechanisms tend to disappear at low-silver coating weights.

This paper considers application of the simple random dot model to radiation effects on emulsions coated in single-layer format with color chemistry.

Experimental

Table 1 summarizes the pertinent physical characteristics for seven equally efficient tabular magenta-dyed green-light-sensitive emulsions. These tabular grains range from 0.47 to 2.90 µm in equivalent circular diameter (ECD) and range in volume over nearly two orders of magnitude.

Table 1. Emulsion grain sizes used in this experiment.

ECD (µm)	Thickness (µm)	Volume (µm)^3
2.90	0.132	0.872
2.30	0.132	0.548
1.28	0.127	0.163
1.18	0.121	0.132
0.79	0.108	0.053
0.62	0.111	0.034
0.47	0.118	0.020
ECD= average equivalent circular diameter		

After sensitization, these emulsions were coated at a silver level equivalent to 90 mg/ft^2 laydown in a simple single-layer format. The coating format incorporate two imaging couplers C-1 at 1.4 mg/ft^2 and C-2 at 75 mg/ft^2. The total gel laydown integrated about 150 mg/ft^2 in an overcoat, 160 mg/ft^2 in the emulsion layer, and 454 mg/ft^2 as a sublayer. Several standard antifoggants were included to partially simulate a multilayer environment.

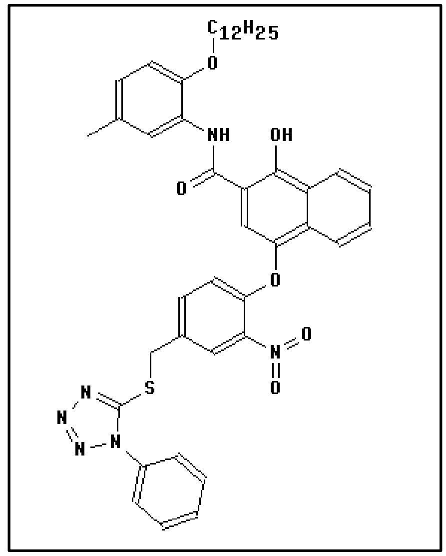

Figure 1. Coupler C-1

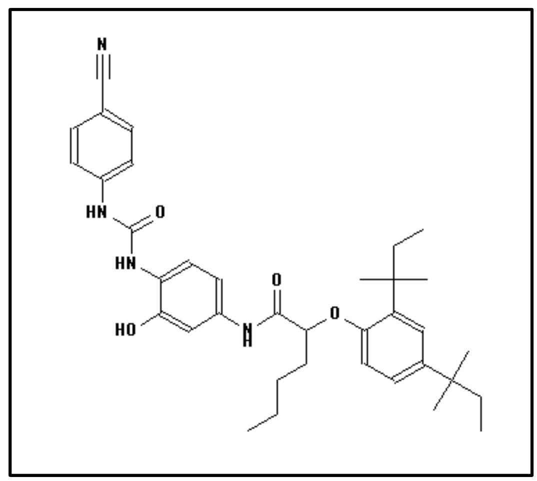

Figure 2. Coupler C-2

Coatings were developed at 1 min 30 s, 3 min 15 s, or 4 min and 30 s in standard color development chemistry. The coatings were treated to a similar regimen after exposure to 105 mRad and 210 mRad of ^{192}Ir radiation. For each of these coatings, granularity measurements were performed using standard techniques and the equivalent of a 48 µm aperture. Only the developed density produced without intentional spectral light exposure was analyzed.

Table 2 summarizes the range of experimental treatments discussed for each of the coatings.

Table 2. Experimental treatment

Radiation dosage (mRads)	Development time		
	1'30"	3'15"	4'30"
0	X	X	X
105	X	X	X
210	X	X	X

Analyses

The Random Dot Model. Mees and James[1a] outline a representation of the random dot model, defining density, D, and the granularity

$$D = agn\,(0.434) \qquad (1)$$

and,

$$\sigma_D = ag(n^{0.5})(0.434)(A^{-0.5}) \qquad (2)$$

and, for a given emulsion,

$$\sigma_D \,\alpha\, D^{1/2} \qquad (3)$$

where σ_D is the standard deviation of the density as read by a microdensitometer of aperture *A, a* is the area of a monodispersed disk*, g* is the absorptance of the disk, *n* is the average number of disks per unit area, and *0.434* is a constant.

For a constant aperture size the relationship can be simplified to:

$$\sigma_D = Kag(n^{0.5}), \qquad (4)$$

where the constant, *K*, captures all of the nonvariables.

For a circumstance in which there are two populations (1 and 2, and the total T) of disks, the contribution of each population to the overall noise in the system adds as the root mean square:

$$\sigma_{DT} = (\sigma_{D1}^{\ 2} + \sigma_{D2}^{\ 2})^{1/2}$$
$$= ((Ka_1g_1(n_1^{0.5})) + ((Ka_2g_2(n_2^{0.5}))^{1/2} \qquad (5)$$

Thus, in principal, for a radiated sample, the high-energy radiation-caused contribution to granularity can be calculated by the root mean square difference between the granularity of a nonradiated sample and a radiated sample. This report describes that calculation and discusses differences between the components of the two σ_D s, as interpreted using the random dot model

Prior to high-energy radiation exposure, the grains are likely to be fogged by sensitization or adventitious light fog. The number of fogged grains will be determined by sensitization conditions or the extent of light fogging. Harsher sensitization conditions should yield larger numbers of fogged grains. For practical applications, the sensitization is chosen to fit the grain's use. For an efficient emulsion of a particular size, there may be a wide range σ_D s, depending on sensitization conditions.

Variation in σ_Ds also occur as a function of grain size because, at constant silver coverage, grain size affects the number of grains coated in a given area, *n*. Grain size also affects the developability of the grains and all of these affect density*, D.*

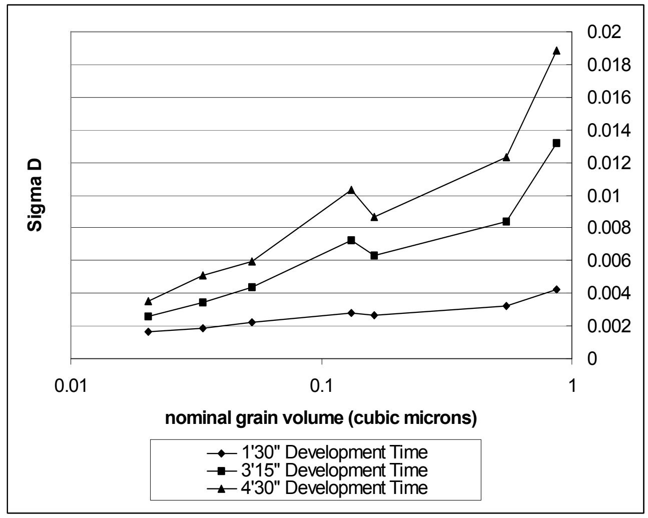

Figure 3. σ_Ds at three different development times for the experimental grains without high-energy radiation exposures

Figure 3 shows some of the variation alluded to in previous paragraphs. The plots look "jagged" because grains having nearly the same volume can give very different granularities, depending, in this case, on the harshness of the sensitization. The largest variations occur because of differences in time of development. Relating this data to the random dot model: *n* for a given emulsion is set by the sensitization and, for that emulsion, is identical across development times, but the size and absorptivity of the dye cloud, *ag*, increases with development time. These σ_Ds are proportional to the square root of *D*, which is not shown here.

After high-energy radiation exposure, the grains fogged by sensitization are still present. Some of these fogged grains a very small fraction from statistical arguments interact with high-energy radiation and accumulate additional development sites. But a number of grains that were not previously fogged will be impacted by radiation. The number of radiation-fogged grains will be determined entirely by the level and energy of radiation. For example, interaction with 25 KeV radiation is so catastrophic that efficiency of sensitization, within the bounds of practicality, is irrelevant, and all grains absorbing these photons will be rendered developable. But for a given level of radiation, the number of exposed grains, grain size, and development time will determine σ_D. Therefore, unlike the Fig. 1, which shows variations as a result of sensitization, the σ_Ds calculated, through the use of Eq. (5) should be smooth and well-behaved across grain sizes.

Figures 4 and 5 show the result of calculations extracting the σ_D contribution from 105 and 210 mRad of radiation.

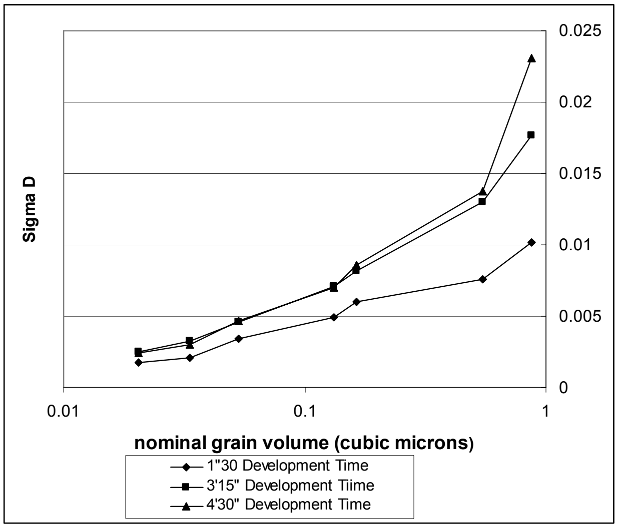

Figure 4. Calculated σ_Ds contributed by radiation for the experimental grains with 105 mRad high-energy radiation exposure at three development times

Both Figs. 4 and 5 exhibit the kind of behavior suggested by random dot theory. Unlike the data in Fig. 3, which is jagged across grain sizes, the calculated σ_Ds in Figs. 4 and 5 show a smooth monotonic increase across grain size and increasing σ_Ds with grain size and development time.

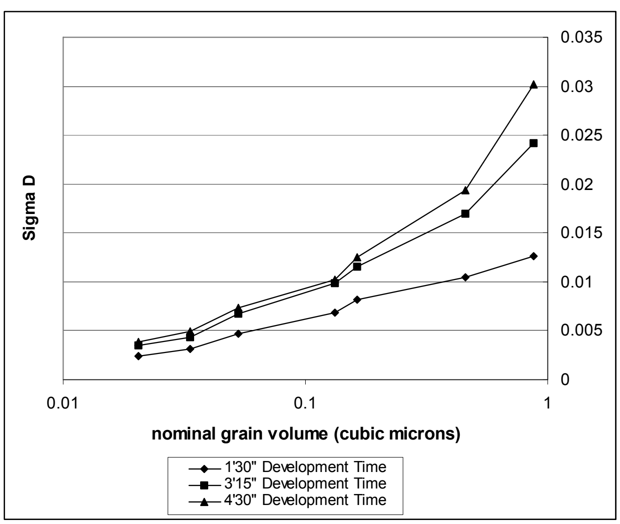

Figure 5:calculated σ_Ds contributed by radiation for the experimental grains with 210 mRad high-energy radiation exposure at three development times.

An additional feature that can be examined is the relationship between the data shown on the two figures. The treatment that generates the difference between the two figures is the level of radiation: Fig. 5 data has twice the radiation relative to Fig. 4 data. With double the radiation dosage, the data shown in Fig. 5, relative to Fig. 4, should have very close to twice the change in *n*, and, correspondingly, twice the increase in *D,* which is not shown here. The increase in *D* should boost the calculated σ_Ds shown in Fig. 5 by a factor of the square root of 2, relative to those shown on Fig. 4. Figure 6 shows the result of dividing the data for each point on Fig. 5 by the data for the corresponding point on Fig. 4.

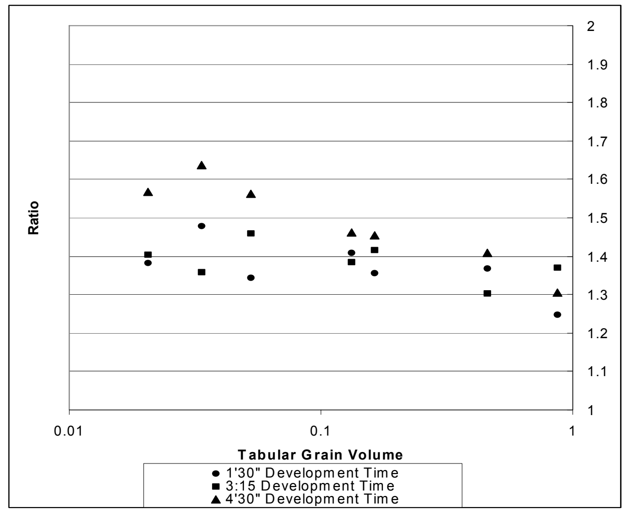

Figure 6. The result of dividing Fig. 5 data by the corresponding data in Fig. 4

Figure 6 demonstrates the relationship between the calculated σ_D s for the two radiation exposures comes close to the expected 1.41 relationship. All of the 1:30 and 3:15 data are within 10% of the expected ratio and only the 4:30 development time data for the smallest grains lies outside of 10 percent. The average of all 21 points on the plot is 1.413. This supports the suggestion that the calculated σ_D s may be meaningfully related to the number of development centers generated by high-energy radiation.

It has been noted by a number of investigators that fogged grains, whether fogged by adventitious light or fogged through the process of sensitization, tend to exhibit a single development site. It has also been argued that an important difference between these fogged grains and high-energy radiation fogged grains is the presence of multiple development sites on the high-energy radiation-fogged grains. Multiple development sites should tend to enhance early development of the radiated grains relative to the nonradiated grains. This suggests that the effect of longer development should be less pronounced on the grains exposed to high-energy radiation because more of the grain's volume is likely to be developed earlier. Using the random-dot model to interpret this: for a given radiation exposure, *n* does not change over the range of development times, but *ag*, the disk size, might be expected to achieve the largest growth early in the development time for radiated grains with much reduced growth at longer times of development, as the grain's silver is exhausted. A visual comparison of Fig. 3 with Figs. 4 and 5 demonstrates that difference. Figure 3 shows a relatively large difference between the σ_Ds for the middle development time and the longer development time. Figures 4 and 5 show σ_D's that virtually collapse together for the middle and longer time of development. More analytical comparisons are shown in the figures below.

With reference to Eq. (4), for a particular emulsion, *n* should be constant over different development times, and dividing the σ_D from one development time by the σ_D from another development time should give a measure of the change or growth in size and absorptivity, *ag,* of the dye cloud "disks," as a function of development time. This division operation should yield a "ratio" larger than unity for a growing disk.

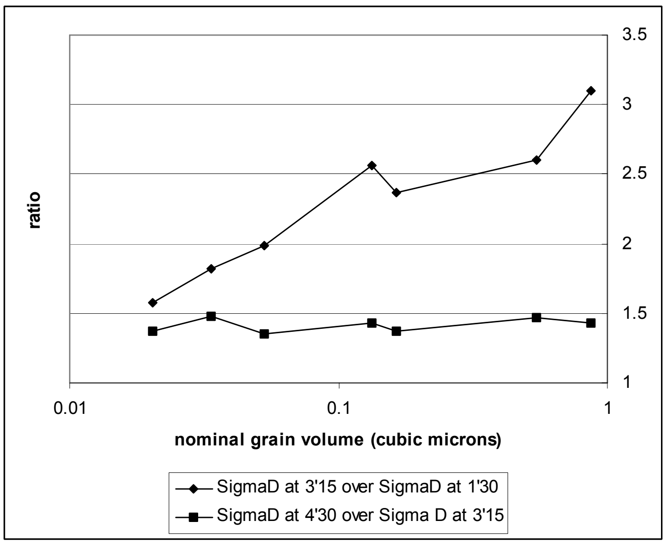

Figure 7. The "ratio" of for different development times, which should be a measure of the relative size of "ag," the disk size and light absorption, for data from Figure 3, no radiation.

As shown in Figs. 3, 4, and 5, the σ_Ds increase with longer development time. Dividing σ_D, from a long development time, by a σ_D from a shorter development time, should always yield a result larger than unity. All of the numbers plotted on Figs. 7 and 8 fall between about 1.1 and about 3.1.

As expected, Figs. 7 and 8 show some similarities. The largest growth appears to occur earlier in the development, as shown in the figures, between the σ_D at 3'15" and the σ_D at 1'30" development time. The largest growth also occurs for the largest grains. Both plots show a nearly constant growth across grains sizes for the longest development time comparison

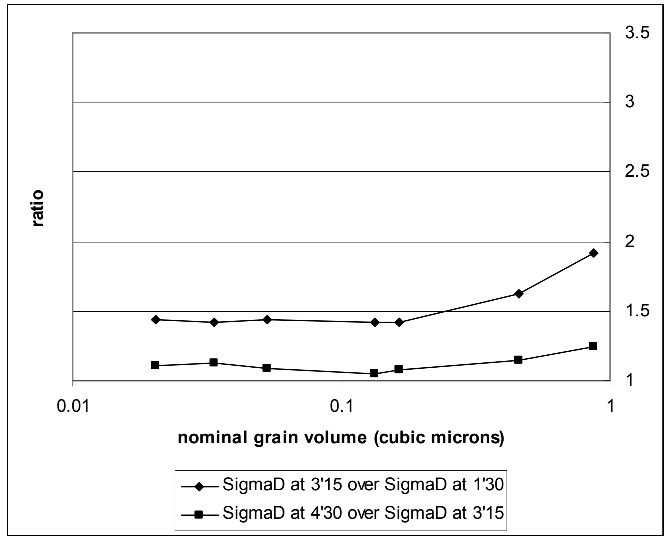

Figure 8. The "ratio" of σ_Ds for different development times, which should be a measure of the relative size of "ag"the disk size, and light absorption, for data from figure 5, 205 mRad exposure.

The data in Figs. 7 and 8 do, however, differ significantly in magnitude. Figure 7, which evaluates the σ_Ds for grains before radiation, shows much larger growth for all comparisons relative to Fig. 8. This difference is consistent with Fig. 7, illustrating the behavior of a population of grains having predominantly a single development site, whereas the Fig. 8 grain population exhibits predominantly multiple development sites.

At the longest development time comparison, Fig.8 σ_Ds show only about 10% growth for all but the largest grain size. This suggests that the grains have achieved almost complete development by 3'15".

Summary

This paper has shown that the contribution to σ_D from high-energy radiation can be analyzed using the random dot model and can be related to radiation level. The σ_D contribution from radiation is fundamentally different from other fog, probably because of the presence of multiple development sites on radiated emulsion grains. In terms offered by the random-dot model, radiation-fogged grains yield dye-cloud "disks" that are larger and tend toward whole-grain development early in the development process.

References

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