Generalization of the Klosterboer-Rutledge Model

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Abstract

The Kosterboer-Rutledge model for silver-soap-based photothermographic systems has been modified to take into account a random distribution of silver-halide grains. This modified model is now general for all grain-number densities.

Introduction

In 1980, D.H. Klosterboer and R. Rutledge developed a theory to model the imaging mechanism of photothermographic systems consisting of silver-halide grains dispersed in silver soap, commercially known as *Dry Silver*.[1] This model is based on several assumptions, the weakest being the implicit assumption that the grains are far enough apart that they do not compete for the surrounding silver soap during the development process. Therefore, in cases where the silver halide grains are distributed in a near-random fashion, the Klosterboer-Rutledge (K-R) model is valid only in the regime of low grain-number density. I propose that this limitation can account for the discrepancies observed by Gisser[2] between experimental results and the K-R model. Gisser found that the dependence of the maximum image density D_{max} and speed on grain-number density deviated substantially from the theoretical predictions. In order to remove this limitation, the K-R model has been modified to take into account a random distribution of silver-halide grains. The modification effectively generalizes the K-R theory so that it is valid for the full range of grain-number densities. The generalized K-R model fits Gisser's data well and a value for the radius of the sphere of influence was extracted.

The Standard K-R Model [1]

The standard K-R model addresses a photo-thermographic system consisting of cubic silver-halide grains suspended in a mixture of silver soap and binder. The silver soap is assumed to be uniformly dispersed throughout the binder which includes other components such as developer and toner. However, the silver soap does not penetrate the silver-halide grains but is in *catalytic proximity* to the grains. The solid mixture of silver soap, binder and other constituents excluding the silver-halide grains will be referred to as the silver-soap mixture. The assumptions made in the standard K-R model are summarized in Table 1. The key concept in the Klosterboer-Rutledge (K-R) model is the *sphere of influence*. The main assumption is that all the silver soap within a sphere-of-influence radius from a latent image is converted into image silver upon thermal development. The optical density created is taken to be proportional to the amount of silver soap developed per unit area. Klosterboer and Rutledge calculated the relationship for image density by assuming that the silver-halide grains with at least one latent image will each contribute one sphere-of-influence volume of developed soap.

The standard K-R relationship for optical density D_{KR} as a function of exposure E in photons per unit area is given by,

$$D_{KR} = D_o \frac{W_{SS}}{W_o}\left(\frac{4}{3}\pi R^3 - L^3\right)\left(\frac{n}{1-nL^3}\right) f_{SH}(\alpha EL^2, A) \qquad (1)$$

$$\left(4\pi R^3/3 - L^3\right)n \le 1,$$

where n is the grain-number density

$$n = \frac{W_{SH}}{\rho_{SH} TL^3}\ , \qquad (2)$$

and

$$f_{SH}(\alpha EL^2, A) = \sum_{m=A}^{\infty} \left(\alpha EL^2\right)^m \frac{e^{-\alpha EL^2}}{m!}\ . \qquad (3)$$

D_o is a dimensionless constant, L is the AgBr grain size, R is the sphere of influence radius, W_{SH} is the silver-halide coating weight, W_{SS} is the silver-soap coating weight, W_o is a silver-soap coating weight for a reference or control sample, ρ_{SH} is the density of the silver halide, and T is the thickness of the film coating. W_o is introduced to make D_o dimensionless. All coating weights have units of mass per unit area. The summation $f_{SH}(\alpha EL^2,A)$ gives the fraction of grains that are developable after an exposure E for grains that require at least A absorbed photons to create a developable latent image.[3] α is the absorption efficiency of the grain.

The first term of Equation 1 in parenthesis represents the volume of silver-soap mixture in one sphere of influence. The second term in parenthesis is the number of silver-halide grains per unit volume of the silver soap mixture. Therefore, the product of these two terms represents the fraction of silver soap that is within a sphere of influence assuming no overlap between the spheres. The product of this term with $f_{SH}(\alpha EL^2,A)$ gives the fraction of silver soap that is within a sphere of influence containing a latent image assuming no overlap between the spheres. The portionality factors D_o and W_{SS}/W_o scales this fraction to obtain the optical density of the developed film.

Table 1. A summary of the main assumptions underlying the standard K-R model is given in the left column.[1] The assumptions that also apply to the generalized K-R model and the generalized D_{max} relationship are appropriately marked by an "X".

	Assumptions for the standard K-R model	Generalized K-R model	Generalized D_{max} relationship
1	The image density is proportional to the amount of silver soap developed.	X	X
2	The silver soap is evenly dispersed through everything in the film except for the silver-halide grains.	X	X
3	All silver soap that is in a spherical volume around a latent-image containing silver-halide grain is reduced to form image silver upon development (definition of the sphere of influence).	X	X
4	All spheres of influence are the same size.	X	X
5	The silver-halide grains are distributed in the film so that their spheres of influence do not overlap.	random grain distribution	random grain distribution
6	All silver-halide grains are cubes of equal size.	X	X
7	All silver-halide grains are equally sensitive requiring the same number of incident and absorbed photons to generate a latent image.	X	
8	The size or number of developable latent images on a silver-halide grain does not affect the sphere-of-influence size or developed density of the surrounding soap.	X	X
9	The distribution of latent images is determined by Poisson statistics.	X	
10	Fog centers are ignored	X	X

The Generalized K-R Model

The weakest point in the K-R model is the implicit assumption that the spheres of influence do not overlap. Near the maximum allowable grain-number density, this constraint would require the spheres of influence to be in a close-packed arrangement. More realistically, the AgX grains are arranged pseudo-randomly in the film. In a random distribution, two grains may be close enough so that their spheres of influence overlap. The resulting competition for the Ag soap in the overlap region, decreases the average amount of soap that can be developed per AgX grain. Therefore, in cases where the silver halide grains are distributed in a near-random fashion, the K-R model is valid only in the regime of low grain-number densities or low exposures. By allowing for overlap between spheres of influence, the K-R model can be generalized to all number densities. To make this generalization, the K-R model is modified by assuming a random distribution of AgX grains. The relationship for image density is calculated by taking into account the overlap between the spheres of influence.

Using assumptions 1, 3, 8 and 10 (Table 1) the basic form of the K-R relationship is given by

$$D = D_o \frac{W_{SS}}{W_o} f_{SS}, \qquad (4)$$

where f_{SS} is the fraction of silver soap that is within the sphere of influence of at least one grain with a latent image. D_o is equal to the optical density acheived when all of the silver soap in the reference sample is developed. For the standard K-R model, $f_{SS} = u_{SS} f_{SH} n/(1\text{-}nL^3)$, where u_{SS} is the average volume of silver-soap in one sphere of influence. Define this fraction to be f_{KR}. To generalize the model, a random distribution of the silver-halide grains in the film is assumed. A new relationship is obtained by recalculating f_{SS} to take into account overlapping spheres of influence that can occur for a random grain distribution.

The calculation is straight forward. First, arbitrarily divide the collection of silver-halide grains in the film into m subsets of equal size. m is chosen sufficiently large so that negligible overlap occurs between the spheres of influence within each subset. The grain-number density within each subset is equal to n_T/m, where n_T is the grain-number density of the entire system. If the grains in one subset are selectively processed, then the fraction of developed soap f_1 is given by the standard K-R model.

$$f_1 = u_{SS} f_{SH} \frac{n_T/m}{1 - n_T L^3} = \frac{1}{m} f_{KR} \qquad (5)$$

Note that n_T is still used in the denominator since the grains in the other subsets still displace soap.

Define u_i to be the fraction of undeveloped soap in the case where i subsets are selectively processed. Now if one additional subset of grains is selectively processed, the fraction of undeveloped soap is further decreased by a factor of u_1. This results in the following regressive relationship,

$$u_{i+1} = u_i u_1 . \qquad (6)$$

Therefore,

$$u_m = u_1^m = (1 - f_1)^m = \left(1 - \frac{1}{m} f_{KR}\right)^m \xrightarrow{m \to \infty} e^{-f_{KR}} . \qquad (7)$$

Finally, f_{SS} is simply 1- u_∞, so

$$f_{SS} = 1 - e^{-f_{KR}} . \qquad (8)$$

Therefore, the generalized K-R relationship is given by,

$$D = D_o \frac{W_{SS}}{W_o}\left(1 - e^{-f_{KR}}\right) \qquad (9)$$

$$= D_o \frac{W_{SS}}{W_o}\left(1 - \exp\left[-\left(\frac{4}{3}\pi R^3 - L^3\right)\left(\frac{n}{1-nL^3}\right) f_{SH}(\alpha E L^2, A)\right]\right).$$

In the case where f_{KR} is much less then one, the exponential can be approximated by 1-f_{KR} and the standard K-R model is easily recovered.

Evaluation of the Generalized K-R Model

The generalized K-R model can be tested by using the predicted relationship for the maximum image density D_{max} to fit experimental data. To obtain D_{max}, f_{SH} is set to unity in Eq. 9. The relationship for D_{max} does not depend on the sensitivity of the silver-halide grains or the statistics of latent image distribution (Table 1). Therefore, it is perfectly suited for testing the generalized K-R model since no changes in these areas were made.

Gisser has shown that the standard K-R model poorly fits experimental data for D_{max} as a function of grain-number density.[2] However, the generalized K-R model fits this data very well (Fig. 1). The two parameters that are allowed to vary are D_0 and R; best fit was achieved for D_0 = 3.64 and R = 0.32 µm. Clearly, more data points are desirable. Nevertheless, the fit shows that the generalized model can account for the anomalous data. Furthermore, the generalized and standard K-R models both have the same number of independent variables. Therefore, the generalization has not increased the ability to fit experimental data simply by increasing the number of fitting parameters.

The results predict that if all of the silver soap were developed in the film samples, an optical density of 3.64 would be achieved, and a developable grain influences the soap within a 0.32 µm radius. These results seem reasonable, but should be tested experimentally. More data points can determine if the extracted D_0 is a good value. Some careful electron-microscopy studies may be able to determine if the extracted sphere-of-influence radius is reasonable. However, the sphere-of-influence construct has some simplifying assumptions that will make any real comparison between the extracted radius and actual radius difficult. The coating is not homogeneous at the scale of 0.3 µm, and the volume of influence may not necessarily be a sphere.

More relevant is the predictions the model makes on film parameters. The effect of grain-number density on contrast and speed for the generalized K-R model can be clearly seen in Eq. 9. The larger the coefficient in front of f_{SH}, the more sensitive the exponential term becomes, so both speed and contrast are predicted to increase with increasing grain-number density. This coefficient, $(4\pi R^3/3 - L^3)n/(1-nL^3)$, in the generalized model is no longer the fraction of soap within a sphere of influence. However, in either model, it is equal to the average number of grains in an arbitrary sphere of influence. This is the controlling parameter for contrast and speed in the generalized K-R model apart from the inherent contrast and speed of the silver-halide grains.

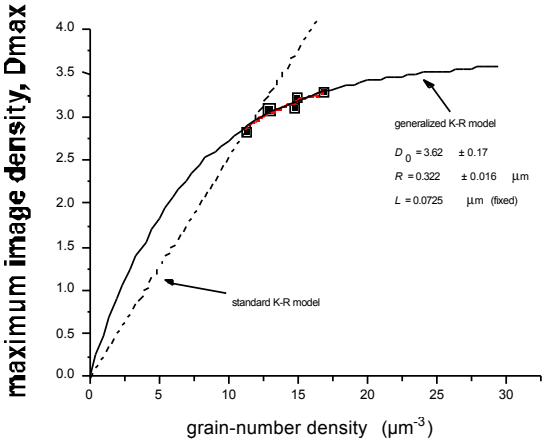

Figure 1. Experimental data documented by Gisser[2] is fit to the generalized K-R model. The results are compared with the predictions of the standard K-R model. The origin is an assumed data point. The slope of the line for the standard K-R model is chosen so that it passes through the origin and the first experimental data point.

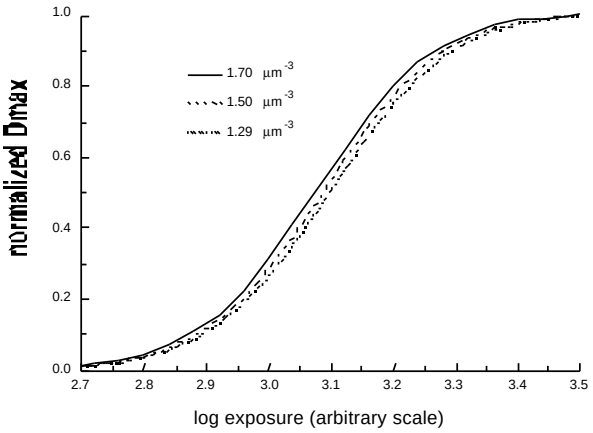

Figure 2. The normalized density versus log-exposure curves predicted by the generalized K-R model for systems with grain size of 71 nm, sphere of influence radius of 0.322 µm, and grain-number densities of 12.9, 15.0, and 17.0 grains per µm^3 are displayed. They correspond to the three respective data points in Fig. 1. The number of absorbed photons required to create a latent image was arbitrarily chosen to be 8.

The normalized image density versus log-exposure curves in Fig. 2 were generated by using Eq. 9. The increase in speed with increasing grain-number density is clearly evident; however, the speed change of about 0.01 between the points is much less than the speed change of 0.04 reported by Gisser. Possible explanations are an improper choice for the parameter A or the deficiencies of assumptions 6 and 7 which would make the predicted contrast too high and possibly affect the speed dependence on grain-number density. Nevertheless, the generalized K-R

model predicts a speed increase with increasing grain number density, which the standard K-R model cannot account for.

Conclusions

The limitation in the standard K-R model lies in the implicit assumption that the spheres of influence do not overlap, which restricts the validity of the model to low grain-number densities. The model was generalized by assuming a random distribution of silver-halide grains and taking into account the effect of the resulting overlap between spheres of influence. The generalized K-R model gives a good fit to the observed D_{max} dependence on grain-number. Enhancements in speed and contrast with increasing number of grains per sphere of influence volume is also predicted.

References

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3. J. C. Dainty and R. Shaw, Image Science, (Academic, London, 1974), Chap. 1.