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GRAY SCALE CHARACTERISTICS OF THE MICROENCAPSULATED ACRYLATE PROCESS OF IMAGING

by

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Abstract

A model of the imaging process involving both the "checkerboard" process and the analog process of imaging has been developed to describe both the granularity and the grey scale characteristics of the microencapsulated imaging process. The model is shown to provide a quantitative rationale of sensitometric and microdensitometric behavior of the system. The results of the analysis demonstrate that while the analog process governs the grey scale character of capsule images, there is a significant contribution from the digital process. This digital process manifests itself primarily in the granularity characteristics of the image and, except in special cases, contributes relatively little to the grey scale of the image.

INTRODUCTION

A distinguishing characteristic of the microencapsulated acrylate process of imaging is the way in which the fundamental imaging element, the microcapsule, governs image density. Each individual microcapsule provides a quantity of dye to the final image as a continuous function of the exposure energy received by the microcapsule\(^1\). This analog behavior of the microcapsule is in contrast to many processes of imaging in which image density varies with the number per unit area of imaging elements that appear in the final image. A midtone density in conventional silver and electrophotographic processes, for example, occurs because some fraction, per unit area, of the total number of grains or toner particles are developed or transferred to the final image. The microcapsule process, on the other hand, produces a midtone density when all of the capsules deliver a fraction of their available dye. Silver grains and toner particles may be considered to be "digital" imaging elements and are either "on" or "off" in the final image. The grey scale characteristics of these systems have been described quantitatively with a variety of "random dot" models\(^2-3\) in which the fraction of "on" and "off" elements composing the image is governed by exposure and by statistics. The grey scale of microcapsule images has also been described quantitatively but has been modeled as an "analog" process in which exposure drives the chemical kinetics of polymerization and crosslinking\(^4\).

Random dot models not only provide a description of the grey scale characteristics of systems with "digital" imaging elements, they also rationalize the attribute of graininess. The fraction of "on" versus "off" elements in the image governs image density, and the size and spatial distribution of the elements determines granularity. However, granularity has also been reported in images produced by the microcapsule process\(^5\). Microdensitometric examination of capsule images suggested that there is a random spatial distribution of photographic sensitivities among the capsules. Thus, while each capsule imaging element is capable of imaging in an analog fashion, the dye delivered to the final image was reported to vary spatially as a result of this sensitivity distribution. This suggests that the capsule imaging process, though primarily analog in nature, has some component of the digital mechanism superimposed upon it.

In the current study, we have attempted to combine a well known "checkerboard" model of
digital imaging with the analog nature of the capsule imaging elements. The resulting analog/digital model is then used to rationalize experimental data on the microcapsule imaging process and to measure, quantitatively, the relative contribution of the analog and the digital components of the mechanism to the overall grey scale characteristics of the system.

EXPERIMENTAL

Microcapsule Formulation: Microcapsules with walls formed from a condensation polymerization between urea and formaldehyde were prepared as described previously. The internal phase of the capsules is composed of trimethylolpropanetriacrylate (TMPTA). The monomer also contains a commercial carbonless type dye precursor which forms a visible image when brought into contact with a proprietary activator composition coated on the receiver sheet of the system, as described previously. Two capsule systems were prepared. The first employed 4-benzoyl-4'-methyldiphenyl sulphide (BMS) sensitizer at 0.652 molar with ethyl-p-dimethylaminobenzoate (EPD) as hydrogen donor at 0.228 molar. The second system employed benzophenone (0.24 molar) and Michler's Ketone (0.02 molar). In both systems the average capsule size was approximately 8μm, as determined with a Coulter Counter Model TAI. Both systems were coated on 1.0 mil polyethylene terephthalate film.

Sensitometry: Sensitometric exposures were carried out using a commercial step tablet, graded in 0.05 density steps, placed in direct contact with the capsule sheet. Exposures were carried out employing a 1000W xenon arc lamp with a grating monochromator set at a 20 nm slit width. Exposed capsule sheets were developed in face-to-face contact with a commercial Cycolor receiver sheet and processed with steel laboratory pressure rollers loaded to approximately 1000 N per cm of roller length. As described previously, the image density on the receiver sheet was found to be directly proportional to the quantity of dye, per unit area, on the sheet. The value of gamma at any point on the D-LogE curve was measured as the slope of the line drawn between the data points immediately before and after the selected point.

Microdensitometry: Microdensitometric measurements were performed with an image analysis system which utilized a monochrome video camera and a DT2851 frame grabber digitizing board. Once the image was digitized and converted to absolute density, a scanning aperture was synthesized in software and traversed across each step of the step tablet image to determine both the mean density and the variance in density (squared standard deviation). Each density scan was 3 mm long with a scanning aperture of 200 μm in the scan direction and 1000 μm in the orthogonal direction. Each scan was digitized to 128 discrete data points of density, and three different scans were run for each step of the step tablet image. The density, D, and density variance data, σ²D, reported in this paper are values averaged from the three scans.

A DIGITAL MODEL – THE CHECKERBOARD

The "checkerboard" model is, historically, one of the first approaches to a description of random dot images. It is also a relatively simple model. Although the checkerboard is conceptually less realistic than more complex random dot models, we have chosen to use the checkerboard as a simplifying approximation in our description of the combined digital/analog process. The essential features of the checkerboard, as originally described by O'Neill, are shown in Figure 1. The area, A, of the checkerboard is the aperture of the densitometric measurement (6 x 6 elements in this example). Each of the individual elements, area a, is assumed to be much smaller than the measuring aperture, A. The number of elements, N = A/a, is thus large, and the resultant reflectance of the image, Rr, measured through the aperture, A, is given as

\[ R_r = R_1 \cdot F + R_0 (1-F) \] (1)
where $F$ is the fraction of imaging elements that are "on", or at $D_{\text{min}}$ with a reflectance $R_1$, and $R_0$ is the reflectance of the "off", or $D_{\text{max}}$ elements. If $p$ is the probability that any randomly chosen element is "on", and if $A \gg a$, the binomial distribution function applies. Then, for any randomly chosen position of the measuring aperture, the average, or expectation value of $F$ (denoted $\bar{F}$) and the variance in $F$ (denoted $\sigma^2_F$), are given as follows.

$$\bar{F} = p$$

$$\sigma^2_F = \frac{\bar{F}(1-\bar{F})a}{A}$$

In traditional silver systems, the origin of the variability in $F$ is described by a Poisson distribution of photons on the imaging elements, each element requiring only a small number of photons in order to undergo an "on-off" transition. However, microcapsules are several orders of magnitude less sensitive than silver grains, and the photon flux required to expose the system is so great that the variance due to a Poisson distribution of the photons is assumed to make a negligible contribution to image variance. Rather, we will assume that the origin of the variance in $F$ is a reflection of a variance in the photographic sensitivity of the individual imaging elements. Experimental evidence for this sensitivity variance has been published.

We will assume that the photographic sensitivity of the individual imaging elements are normally distributed according to the following Gaussian function.

$$\Gamma_S(S_o) = \frac{1}{\sigma_S\sqrt{2\pi}} \exp\left[\frac{-(S_o - S_m)^2}{2\sigma_S^2}\right]$$

Here, $S_o$ is the exposure, in Log units, required to induce an "on-off" transition in an element, $S_m$ is the mean value of $S_o$ among the elements, and $\sigma_S^2$ is the variance in the $S_o$ sensitivity. We express the cumulative normal function, $C_{\text{norm}}$, as follows.

$$C_{\text{norm}}(x,\bar{x},\sigma_x) = \int_{-\infty}^{x} \frac{1}{\sigma_x\sqrt{2\pi}} \exp\left[\frac{-(x - \bar{x})^2}{2\sigma_x^2}\right] dx$$

Thus, for any given level of exposure, the average fraction of "on", or $D_{\text{min}}$, elements within a measuring aperture, $A$, is the $C_{\text{norm}}$ function.

$$\bar{F} = C_{\text{norm}}(S_o,S_m,\sigma_S)$$

By combining equations (1) and (6), we can generate a characteristic curve of the checkerboard imaging process, $R_r$ versus exposure in $S_o$ units. The midtone sensitivity of the system is the average sensitivity, $S_m$, of the elements, and the grey scale is governed by the sensitivity variance, $\sigma_S^2$.

THE ANALOG / DIGITAL MODEL

We will simulate the overall behavior of the microcapsule system by describing a checkerboard with analog imaging elements. Rather than undergoing a sharp "off-on" transition, the imaging elements will be assumed to change in a continuous way from $D_{\text{max}}$ to $D_{\text{min}}$ as the
level of exposure increases. The analog process by which the microcapsules control dye has been modeled based on the chemical kinetics of acrylate photopolymerization and the mechanics of pressure development\(^4\). This kinetic/mechanical model produced a sigmoidal D–LogE curve that fit well the observed behavior of the capsule system. However, rather than incorporate the complexity of the kinetic/mechanical model into a combined digital/analog description of the process, we will approximate the dye control characteristics of an individual imaging element with an empirical shape function.

\[
\Gamma_a(S, S_0) = \frac{1}{\sigma_a \sqrt{2\pi}} \exp \left[ -\frac{(S - S_0)^2}{2\sigma_a^2} \right] \tag{7}
\]

In this function, \(S\) is the exposure, in Log units, given to the image, \(S_0\) is an index of sensitivity of the imaging element, and \(\sigma_a^2\) is an empirical shape factor describing the grey scale of the analog imaging element. We now define \(R_0\) and \(R_1\) as the reflectance factors of a not exposed (Dmax) and of a thoroughly exposed (Dmin) element in a checkerboard. At an exposure level of \(S\), we can describe the reflectance of an individual element of sensitivity \(S_0\) as \(R_a(S, S_0)\).

\[
R_a(S, S_0) = R_d \cdot C_{norm}(S, S_0, \sigma_a) + R_0 \tag{8}
\]

where,

\[
R_d = R_1 - R_0
\]

In the limit as \(\sigma_a \to 0\), the imaging elements undergo a sharp "off–on" transition as in the traditional checkerboard model.

The overall reflectance, \(R_r\), of a checkerboard with analog elements, as measured through aperture \(A\), can now be described by an extension of equation (1). At a given level of exposure, \(S\), there will be some fraction of elements with a reflectance factor of \(R_a(S, S_0)\). The fraction of elements at \(R_a(S, S_0)\) is simply \(\Gamma_s(S_0)\) in equation (4). Thus, the overall reflectance, \(R_r(S)\), is the infinite sum of the products of the reflectances and fractions.

\[
R_r(S) = \int_{-\infty}^{\infty} \Gamma_s(S_0) \cdot R_a(S, S_0) \, dS_0 \tag{9}
\]

We define the slope of the curve, \(\gamma_r\) versus \(S\), as \(\gamma_r(S) = dR_r(S)/dS\). Taking the partial of equation (9) with respect to \(S\) gives,

\[
\gamma_r(S) = R_d \cdot \int_{-\infty}^{\infty} \Gamma_s(S_0) \cdot \Gamma_a(S, S_0) \, dS_0 \tag{10}
\]

We recognize the integral in this expression as a convolution of two Gaussian functions, and the result of this convolution is another Gaussian. Thus,

\[
\gamma_r(S) = R_d \cdot \Gamma_r(S) \tag{11}
\]

where,

\[
\Gamma_r(S) = \frac{1}{\sigma_r \sqrt{2\pi}} \exp \left[ -\frac{(S - S_m)^2}{2\sigma_r^2} \right] \tag{12}
\]
\[ \sigma_r^2 = \sigma_a^2 + \sigma_s^2 \]  

(13)

The standard deviation, \( \sigma_r \), of equations (12) and (13) is a measure of the grey scale of the digital/analog checkerboard system. As one might have anticipated, the variances of the digital and analog components are additive. We can now represent equation (9) as a Cnorm function.

\[ R_r(S) = R_0 + R_d \cdot \text{Cnorm}(S, S_m, \sigma_r) \]  

(14)

With image density defined as,

\[ D(S) = - \log[R_r(S)] \]  

(15)

we have the characteristic D–LogE behavior of the digital/analog model.

**GRANULARITY OF THE DIGITAL/ANALOG MODEL**

The only difference between the traditional checkerboard model and the digital/analog model is the nature of the "off–on" transition. In the current model, we can understand "off" to mean an element for which the exposure sensitivity is greater than or equal to the given exposure, \((S_0 \geq S)\) regardless of the particular reflectance, \(R_a(S, S_0)\), of the element. Thus we can maintain the concept of an "on" fraction, \(F\), so the binomial distribution function still applies. This in turn means, equation (3), (where \(F\) is given by equation (6)) describes an intrinsic variance in \(F\) in the combined digital/analog checkerboard.

Experimentally, we measure the variance in the image as a density fluctuation, \(\sigma_D^2\). However, for a given exposure, \(S\), the image density is a function of the mean sensitivity of the imaging elements. As shown previously, error propagation relates the value of \(\sigma_D^2\) to an observable variance, \(\sigma_{S_0}^2\), in sensitivity as follows.5

\[ \sigma_D^2(S) = \sigma_{S_0}^2 \cdot \gamma_D^2(S) + \sigma_B^2 \]  

(16)

Here, \( \gamma_D^2(S) \) is the square of the point slope on the D–LogE curve, and \( \sigma_B^2 \) is a background variance in density caused by, for example, non uniformities in the optical properties of the receiver to which the image is applied. By plotting measured values of \( \sigma_D^2 \) against the point slope on the D–LogE curve, an estimate of the sensitivity variance, \( \sigma_{S_0}^2 \), can be made. Figure 2 shows an example for exposures made on the BMS/EPD microcapsule system. The dependence of \( \sigma_{S_0}^2 \) on the wavelength of exposure will be discussed subsequently.

The numerical value of \( \sigma_{S_0}^2 \) obtained in this way is, of course, dependent on the experimental aperture, \(A\), of the measurement of \( \sigma_D^2 \). Thus, \( \sigma_{S_0}^2 \) is not the same as \( \sigma_S^2 \) in equation (4), which describes the intrinsic variance in sensitivity among the imaging elements. The relationship between the two can be obtained from equation (3), as follows. First, we relate the observed variance, \( \sigma_{S_0}^2 \), to the variance in the "on" fraction, \( \sigma_F^2 \), by error propagation.
Substituting equation (3) for $\sigma_F^2$, and identifying the partial, $\partial S_0 / \partial F = 1/\Gamma_S(S_0)$, in equation (4), we obtain the following.

$$
\sigma^2_{S_0} = \sigma_F^2 \cdot \left[ \frac{\partial S_0}{\partial F} \right]^2
$$

A special case of this function can be written when the exposure given the system is exactly equal to $S_m$. Then, $F = 1/2$ and $\Gamma_S(S_m) = 1/(\sigma_S \cdot \sqrt{2\pi})$. Thus, equation (18) becomes

$$
\sigma^2_{S_0} = \sigma_S^2 \cdot \frac{\pi \cdot a}{2A}
$$

This expression suggests that the observed variance in image density, $\sigma_D^2$, should be inversely proportional to the aperture of measurement. As with most imaging systems, this is indeed the case, as shown, for example, in Figure 3 for measurements made on images generated from the BMS/EPD capsule system exposed at 360 nm.

RELATING SLOPE TO DYNAMIC RANGE

The grey scale of an imaging system may be described quantitatively by either the slope, "gamma", of the characteristic D—LogE curve, or by the dynamic range, in LogE units, over which image density changes. For the Gaussian model of equation (12), $\sigma_r$ is an index of the dynamic range, and the relationship between dynamic range and the peak slope, $\Gamma_p$, of the $R_r$—LogE curve is given by equation (12) with $S = S_m$.

$$
\Gamma_p \sigma_r = \frac{1}{\sqrt{2\pi}}
$$

The contribution of the digital component of the imaging process, represented by $\sigma_S^2$, to the overall grey scale of the system, represented by $\Gamma_p$, can be seen by combining equations (13) and (20).

$$
\frac{1}{\Gamma_p^2} = (\sigma_S^2 + \sigma_a^2) \sqrt{2\pi}
$$

Thus, an increased variance in photographic sensitivity in the checkerboard system will decrease the gamma of the image.

An experimental test of equation (21) can be obtained by analysis of the behavior of the BMS/EPD capsule system. As shown in Figure (2), the magnitude of $\sigma_{S_0}^2$ depends on the wavelength to which the coated capsule sheet is exposed. This phenomenon was shown to derive from coating irregularities coupled with the optical density of the BMS sensitizer in the microcapsules. The molar extinction coefficient of the sensitizer increases as the exposure wavelength decreases from 380 nm to 330 nm. This causes the optical density of the capsules to vary with wavelength. At higher optical densities, small coating fluctuations induce a fluctuation.
in sensitivity, and the reader is referred to our earlier report for mechanistic details of this phenomenon. That $\sigma_{S_0}^2$ can be altered experimentally simply by selecting the exposure wavelength provides a convenient test of equation (21). Indeed, as shown in Figure (4), the grey scale characteristics of the BMS/EPD system are wavelength dependent in subjective agreement with equation (21).

In order to examine quantitatively the relationship between $\sigma_{S_0}^2$ and grey scale, we need to recast equation (21) in terms the slope of the D–LogE curve rather than the slope of the $R_r$–LogE curve. We define $\gamma_D(S)$ as the slope of the D–LogE curve.

$$\gamma_D(S) = \frac{dD}{dS}$$  \hspace{1cm} (22)

Taking this derivative, with equations (15) and (14), we obtain,

$$\gamma_D(S) = -K_1R_d \cdot \frac{\Gamma_r(S)}{R_r(S)}$$  \hspace{1cm} (23)

where $K_1 = \log_{10}(e)$. Inspection of this function shows that $\gamma_D(S)$ does not reach a maximum at the same value of $S$ at which $\Gamma_r(S)$ reaches a maximum. Thus, to find the peak value of $\gamma_D(S)$, we take the derivative of equation (23) with respect to $S$ and set it equal to zero. Through algebraic manipulation, this leads to the following

$$\gamma_p\sigma_r = \frac{-K_1R_d}{R_p \sqrt{2\pi}} \cdot \exp\left[\frac{-\gamma_p^2 \sigma_r^2}{2K_1^2 R_d^2}\right]$$  \hspace{1cm} (24)

where $\gamma_p$ and $R_p$ are the values of the slope and reflectance at the point on the D–LogE curve where $\gamma_D(S)$ is at a maximum. This function is similar to equation (20) describing the relationship between slope and $\sigma_r$ on the $R_r$ versus $S$ curve. Since $\gamma_p$ and $\sigma_r$ appear always as a product in equation (24), it is possible conceptually to solve equation (24) for the product $\gamma_p\sigma_r$ and thus to obtain an expression equivalent to equation (20)

$$\gamma_p\sigma_r = K_2$$  \hspace{1cm} (25)

Unlike the product $\Gamma_p\sigma_r$, the value of $\gamma_p\sigma_r$ is dependent on the reflectance values, $R_d = R_1 - R_0$ and $R_p$. Experimental estimates for these reflectances were readily obtained from D–LogE curves of the BMS/EPD system, and equation (24) was then solved numerically to obtain $K_2 = -0.292$.

By combining equations (13), (19), and (25), we obtain,

$$\frac{1}{\gamma_p^2} = \sigma_{S_0}^2 \alpha + \beta$$  \hspace{1cm} (26)

where,

$$\alpha = \frac{2A}{K_2^2 \pi a}$$  \hspace{1cm} (27)
Values of $1/\gamma_p^2$ and $\sigma_{S_0}^2$, obtained from exposures of the BMS/EPD system at different wavelengths, were plotted as shown in Figure (5), and the results fit reasonably a straight line. Moreover, the unitless slope of the line, $\alpha = 1333$, coupled with our estimate of $K_2 = -0.292$ and a measuring aperture of $A = 200,000 \mu m^2$ in the granularity analysis, leads to an estimate of $\sqrt{\alpha} = 33 \mu m$ from equation (27). This is an estimate of the linear dimension of the imaging element of the checkerboard model approximating the behavior of the BMS/EPD system. Figure (6) shows a photomicrograph of a midtone image of the BMS/EPD system along with a 33 $\mu m$ checkerboard element. While the system is clearly not an ideal checkerboard, an element size of 33 $\mu m$ appears to provide a reasonable first order approximation of the system.

An overall fit of the model to experimental D–LogE data can now be obtained. The value of $\beta$ in Figure (5), with $K_2 = -0.292$, provides an estimate $\sigma_a^2 = 0.0017$ (in units of Log E squared). Values of $\sigma_{S_0}^2$, from Figure (2), coupled with equation (19), provide values of $\sigma_S^2$ at any wavelength of exposure. Thus, equations (13) through (16) may be combined, and a fit to experimental D–LogE data can be made by varying only the arbitrary sensitivity index, $S_m$. Figure (7) shows fits for the BMS/EPD system exposed at 330nm and 360nm. The agreement between the curves and the data in Figure (7) suggests that the assumed Gaussian shape functions, and the checkerboard model, provide a reasonable approximation to the grey scale characteristics of the microcapsule system.

CONCLUSIONS

The results of this study indicate that the overall grey scale of the microencapsulated acrylate process of imaging is governed by two factors. The first is the intrinsic analog nature of the individual microcapsules. The second is a random spatial distribution of photographic sensitivity across the coating of microcapsules. These two factors have been labeled the "analog" and the "digital" components of the imaging process and have been modeled as a checkerboard of analog imaging elements with a random distribution of sensitivities among the elements. The size of the imaging elements, approximately 33$\mu m$, is larger than the size of the microcapsules and corresponds to the size of the coating microfluctuations discussed previously. The model provides a means of characterizing the relative contribution of the analog and digital components as $\sigma_a^2$ and $\sigma_S^2$ respectively. From experimental behavior observed in figure (5), it appears that the analog process dominates the grey scale character of the system when the optical density of the system is relatively low. At high optical densities, the digital component of the process induces significant granularity and increases grey scale.

Acknowledgement

The authors express their sincere appreciation to Dr. L. Feldman and Dr. E. Saccocio for their continued insistence that we "look at spots". Thanks also to P. Engeldrum for technical guidance and for not asking "what's a chemist like you doing on a project like this?" Thanks also to Dan Dillard for assisting with the microdensitometric measurements.
REFERENCES


FIGURES

Figure 1: A checkerboard image.
Figure 2: Variance in image density versus gamma^2 for the BMS/EPD system.

Figure 3: Variance in image density versus 1/aperture for the BMS/EPD system.
Figure 4: D–LogE data for the BMS/EPD system exposed at 330 nm and 360 nm.

Figure 5: Relationship between the peak value of gamma and the measured variance in sensitivity.
Figure 6: Photomicrograph of a midtone image of the BMS/EPD system exposed at 360 nm. Arrows indicate a 33 μm checkerboard element.

Figure 7: D–LogE data and model for the BMS/EPD system at 330 nm and 360 nm.