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ON A NATURE OF CATHODOLUMINESCENCE CONTRAST OF FINE-DISPERSED STRUCTURES IN THE SCANNING ELECTRON MICROSCOPE

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Abstract

Fine-dispersed structures (FDS) consisting of a large number of microcrystalline or amorphous particles of different sizes and shapes were examined in cathodoluminescence (CL) mode scanning electron microscopy (SEM). Line dimension of each particle (about 10 - 100 μm) was larger than the electron beam diameter as well as electron scattering volume in material under investigation. An analysis of observed images showed the existence of some peculiarities in contrast which have not been observed in the CL-images for solid specimens. The FDS CL-image topographic contrast arises as a result of detection of CL-emission from an aggregate of FDS-elements surrounding an irradiated particle because of the bombardment of the elements by secondary electrons.

A model was created for the quantitative description of the secondary electron scattering processes. The model takes into account random distribution of microcrystals in FDS-volume, secondary electron emission, elastic and inelastic electron scattering, elastic and inelastic CL-emission photon scattering, and CL-collector angle aperture.

A computer model of the processes described above was made by the Monte-Carlo method to reveal a physical mechanism of FDS CL-image contrast formation. This allowed the calculation of a portion of topographic contrast of FDS CL-images and the dependence of that contrast on depth of a particle position in FDS-volume and on the incident angle of the electron beam on a particle surface. Comparison of the above results with real FDS CL-images shows a good agreement between the theoretical calculations and the experiments.

Key Words: Fine-dispersed structure, electron scattering, cathodoluminescence, random value, CL-image contrast.

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Introduction

In a previous investigation of the cathodoluminescence (CL) in a scanning electron microscope (SEM) on powdered structures in $\text{CaSO}_4 \cdot 0.5\text{H}_2\text{O}$ and BaO , Berdonosova *et al.* (1989) observed normal CL contrast, as well as aureole around the particles in CL-images (Figs. 1, 2 and 3). In this paper, our aim is to provide a theoretical explanation of these experimental observations. The electron beam penetrates deep into the volume of fine dispersed structure. The interaction of these primary electrons, as well as the scattered electrons within the sample lead to excitation of CL both in the crystals under investigation as well as the neighboring material in the fine-dispersed structure crystals of the scattering volume.

Scattering of Electrons in FDS-Volume

The traditional method of image formation in the SEM (progressive or digital scan) results, in some cases, in the formation of artifacts. For CL-images the main artifacts are topographic contrast (the aureoles mentioned above) and the mixed CL-spectrum displayed by particles in contact as shown in Fig. 1. Fig. 4 explains the mixed CL-spectrum image formation. The electron beam incidence angle ψ to the surface of the irradiated particle is varied from 0 to $\pi/2$. Obviously the CL-spectrum being detected for various incident angles will be dependent upon both the local luminescent properties of the irradiated particle and the luminescent properties of the FDS particles surrounding it. The CL-emission of the surrounding particles is excited by both scattered and secondary electrons. For two particles one can write an equation for the detected CL signal

$$I_{CL}(\psi, \lambda) = I_{CL}^A(\psi, \lambda_A) + I_{CL}^B(\psi, \lambda_B)$$

and for the common case

$$I_{CL}(\psi, \lambda) = I_{CL}^*(\psi, \lambda) + \sum_i I_{CL}^i(\psi, \lambda_i)$$

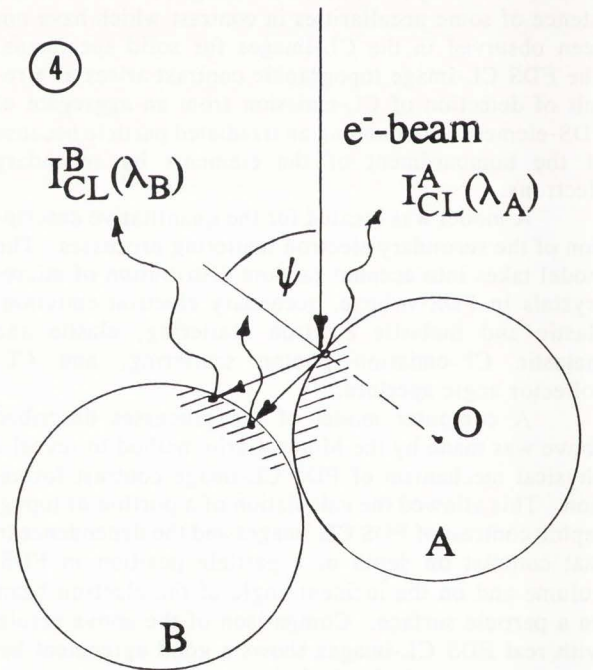
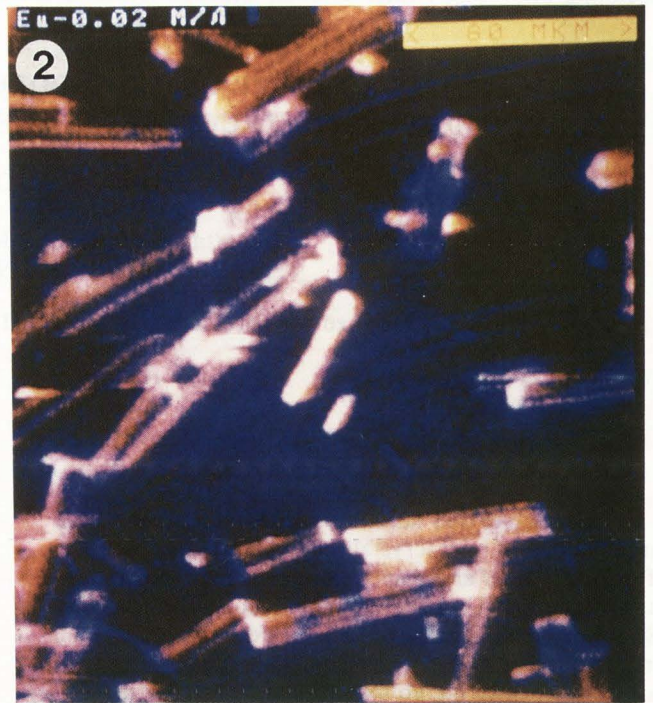
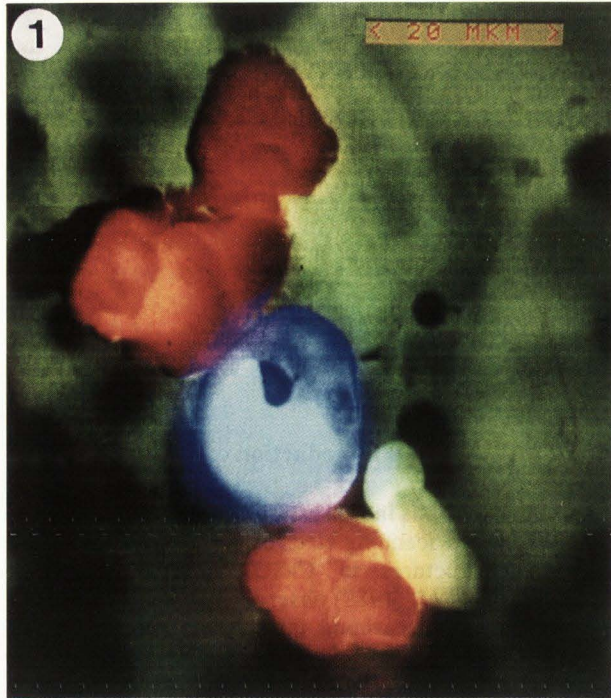


Figure 1. CL-image of from a sample of color television tube screen phosphor powder. The intensity variations from particle to particle, as well as within a particle, are small; but the CL-wavelength changes from particle to particle suggesting that different particles have different compositions. This is normal CL-contrast. Bar = 20 μm . **Figure 2.** CL-image of acicular crystals $\text{CaSO}_4 \cdot 0.5 \text{H}_2\text{O}$ in SEM. Acicular form of crystals is easily recognized because of the presence of light aureole around the particle perimeter. Bar = 80 μm . **Figure 3.** CL-image of the powdered BaO in SEM. All crystals have a light aureole at the particle perimeter. Bar = 150 μm .

CL Contrast of Fine-Dispersed Structures

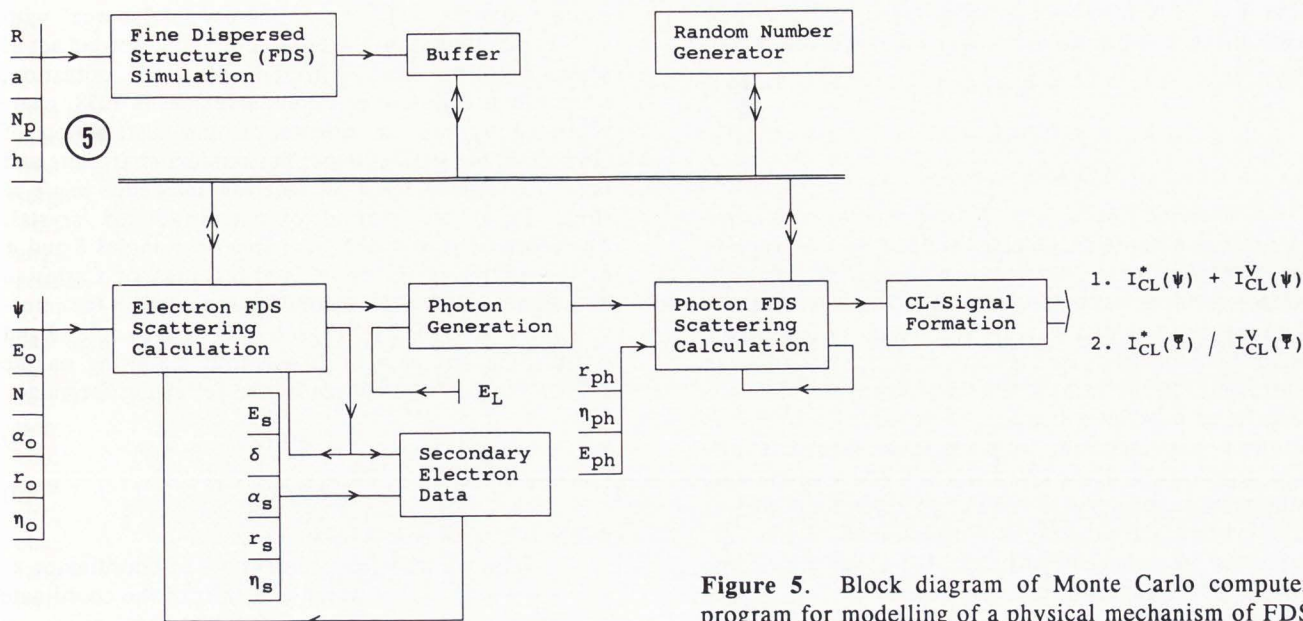


Figure 4 (at left on page 708). The excitation of two neighboring FDS particles. Local CL-spectrum depends on both the λ_A -wavelength of the irradiated crystal luminescence and λ_B - wavelength of the nearest neighboring crystal luminescence excited by the scattered electrons and secondary electrons. The integral CL-signal ascribed at the contact point of the electron beam and the surface particle (a) is equal to $I_{CL}^A(\psi, \lambda_A) + I_{CL}^B(\psi, \lambda_B)$. Shaded areas will be displayed with mixed CL-spectrum when the electron beam crosses particles A and B.

The value I_{CL} defines the brightness and spectral composition of the FDS CL-image displayed on the SEM screen. Fig. 1 illustrates the variation of the CL-spectrum of a line contact of TV-screen phosphor grains: red and blue, red and green, and blue and green particles.

Based on Monte-Carlo method (Sobol, 1968), we consider the basic principles and interactions of various program blocks (Fig. 5) for calculating electron beam scattering and formation of two CL signals: (1) CL from irradiated crystal at the point of impact of an electron beam (to be referred to as the true CL), and (2) CL excited by backscattered and secondary electrons of the FDS-volume (to be referred to as background CL).

The program starts with an ideal FDS consisting of a mixture of spherical crystals distributed randomly in the FDS-volume (Fig. 6). The radii, R , of all particles are assumed to be the same. This structure corresponds to the real FDS in which randomly placed crystals are assumed to have cross-section areas equal to R^2 for all directions chosen inside the FDS.

A three-dimensional system of Cartesian coordinates x, y, z is introduced. We assume that the area containing FDS-crystal centers is limited by the surface

Figure 5. Block diagram of Monte Carlo computer program for modelling of a physical mechanism of FDS CL-image contrast formation. R = particle radius; $2a$ = edge of cube; N_p = number of particles; h is depth of particle from FDS surface; ψ = impact angle; E_o, E_s, E_{ph} = energy of primary and secondary electrons and photons, respectively; N = number of beam electrons; α_o, α_s = CL efficiency; r_o, η_o = probability of elastic and inelastic scattering of primary electrons, respectively. $r_s, \eta_s, r_{ph}, \eta_{ph}$ = probability of elastic and inelastic scattering of secondary electrons and photons, respectively; δ = coefficient of secondary electron emission; $I_{CL}^*(\psi), I_{CL}^V(\psi)$ = CL intensity of the probed particle (true CL) and neighboring particles (background CL), respectively; and E_L = mean photon loss energy in inelastic interaction.

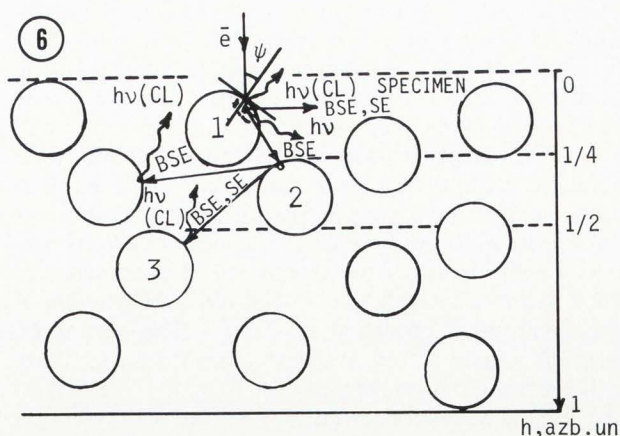


Figure 6. Vertical section of the ideal FDS. Particles 1, 2, 3 lie at depths $h_1 = 0, h_2 = 1/4, h_3 = 1/2$ arbitrary units, respectively. ψ - electron beam incidence angle; CL - local CL emission; SE - secondary electrons; BSE - backscattered electrons.

having a cubic form with an edge equal to $2a$, coordinates of centers having been assumed as the values:

$$-a \leq x \leq a \quad (1)$$

$$-a \leq y \leq a \quad (2)$$

$$0 \leq z \leq 2a \quad (3)$$

Number $i = 1, 2, \dots, N$ is applied to each particle; this number is an address reserved by the program memory containing coordinates of a particle center, a counter of true secondary electrons and a counter of photons, emitted by crystal i . The crystal surface which receives an electron beam is numbered as $i = 1$ allotting coordinates $(0, 0, 2a-h)$ where variation of the FDS surface relief parameter h makes it possible to study the electron beam scattering from a particle, placed at various depths from the FDS-surface. When calculating centers of other crystals contact of crystal surfaces is allowed but their crossing is not permitted. Let us assume that we arranged i particles. Let us calculate coordinates of the center of crystal $j = i + 1$.

$$x_i = a(2\gamma_1 - 1) \quad (4)$$

$$y_i = a(2\gamma_2 - 1) \quad (5)$$

$$z_i = 2a\gamma_3 \quad (6)$$

where γ_i is a random number selected from the generator of random numbers.

Now we write the condition under which the crystal surface marked by j does not cross the surfaces of all crystals formed before:

$$(x_j - x_i)^2 + (y_j - y_i)^2 + (z_j - z_i)^2 \geq R^2 \quad (7)$$

for all $i = 1, j-1$.

In case of non-fulfillment of condition (3) let us repeat procedures (4), (5), (6), and (7) without change of the crystal address until condition (7) is fulfilled. Then, if $j < N$ let us go over to formation of coordinates of centers of the next crystal. The block for forming massive FDS-coordinates is implemented in a form of a separate program, feedback from the basic program of the electron above scattering be organized.

This makes it possible to carry on statistical calculations for FDS with fixed spatial configuration. Circulation from the basic program is also directed to another, perhaps the most important, block of the program. It is a subprogram of the calculation of coordinates of the a particle-"target". Let us consider an algorithm of the subprogram operation in order. The crystal on which the scattering of the electron takes place in future will be referred to as the particle-"source". The crystal accepting the scattered electron (i.e., where the electron takes part in a scattering event) will be called the particle-"target".

Let us consider the scattering of the electron on the crystal-"source", the address of which may take the value $i = 2 \dots N$. The case when the crystal-"source" is the probed crystal will be considered separately. It is

evident that the scattering on the crystal-"source" with $i > 1$ corresponds, at the minimum, to the second act of scattering of the electron in FDS, i.e., to the situation, when the direction of electron movement in FDS, characterized by random orientation and distribution of crystals in the FDS-volume, has random characters and does not depend upon an electron incidence angle ψ (Fig. 5) to the surface of the irradiated crystal. Therefore, it is assumed, that spherical angles θ and ϕ of the scattering direction in the system of Cartesian coordinates $x y z$ are randomly distributed in the intervals $[0, \pi]$ and $[0, 2\pi]$ respectively. Thus angles θ and ϕ from the direction of an electron scattering on the crystal-"source" are chosen by the following formulae:

$$\phi = 2\pi\gamma_4 \quad (8)$$

$$\theta = \pi\gamma_5 \quad (9)$$

where γ is a random number.

Then we transform the system of coordinates $x, y, z \longrightarrow x', y', z'$ which is a shift of the coordinate system to the center of the crystal-"source" x_i, y_i, z_i with two subsequent turns of the coordinate system, as a result of which axis z' of the new coordinate system coincides with the direction of electron scattering on the crystal-"source". Coordinates of the centers of all FDS crystals are transformed as follows:

$$x'_j = (x_j - x_i) \cos\phi - (y_j - y_i) \sin\phi \quad (10)$$

$$y'_j = [(x_j - x_i) \sin\phi + (y_j - y_i) \cos\phi] \cos\theta - (z_j - z_i) \sin\theta \quad (11)$$

$$z'_j = [(x_j - x_i) \sin\phi + (y_j - y_i) \cos\phi] \sin\theta + (z_j - z_i) \cos\theta \quad (12)$$

Furthermore, a uniform distance D_j from a particle-"source" to all particles in the new system of coordinates, having positive coordinate $z_j > 0$ is formed. A particle with number j for which the value D_j is minimum is chosen and a determination is made of whether getting a scattered electron attached to this particle is equivalent to satisfying the condition:

$$(x'_j)^2 + (y'_j)^2 < R^2 \quad (13)$$

If condition (13) is not fulfilled, D_j is given the value $4a$, which will be larger than the maximum distance between any two FDS-crystals. A new minimum distance D_k is chosen and the procedure of satisfying condition (13) is repeated.

As a result of consecutive selection (by degree of closeness) of all crystals for which $z' > 0$, or at a certain stage, we determine the crystal-"target"; or in case of non-fulfillment of condition (13) for any one of the crystals, placed in direction of electron scattering, we come to the conclusion that the electron left the FDS volume without further interaction with crystals. This algorithm for calculating coordinates of the crystal-"target" center allows us to consider the "shadow" (created

by crystals, which are the nearest to the particle-"source" for FDS crystals).

Let us consider the electron scattering on the crystal exposed by the beam. The procedure of determining coordinates of the crystal-"target" does not differ from the above-mentioned one, except for the initial stage of choosing scattering direction angles θ and ϕ . This is explained by the fact that direction of probable angles of the first act of scattering for the crystal exposed to irradiation depends upon the angle of incidence ψ . For simplicity it is assumed that scattering direction has random uniform distribution in half-space, limited by the plane which is tangent to the irradiated crystal surface in the electron beam incidence point. Orientation of this plane in space is determined by angle ψ . Under the actual experiment conditions, in case of incidence of the electron beam of the final section on the surface with developed topography, it is a possible to change angle ψ for a fixed point of incidence in some range. Consequently, in this case, it is impossible to determine ψ unambiguously. Therefore, the previously mentioned procedure of choosing angles of incidence may be considered correct only for an infinitesimally thin beam, which under conditions of the real experiment (beam diameter $\approx 1 \mu\text{m}$) limits the minimum dimensions of FDS crystals to the value of $\approx 10 \mu\text{m}$.

While choosing spherical angles ϕ and θ of direction of scattering on the irradiated crystal for the first interaction of the beam with FDS there are two possibilities:

$$\phi = \pi\gamma_6 - \pi/2 \quad (14)$$

$$\theta = \gamma_7(\pi/2 + \psi) \quad (15)$$

or

$$\phi = \pi\gamma_6 + \pi/2 \quad (16)$$

$$\theta = \pi_7(\pi/2 - \psi) \quad (17)$$

The choice is performed by the analysis of fulfillment of the condition:

$$\gamma_8 = \psi/\pi + 1/2 \quad (18)$$

If condition (18) is fulfilled, angles ϕ and θ are selected by the formulae (14) and (15); otherwise equations (16) and (17) are used.

A characteristic feature of the subprogram organization is a built-in feedback system with numerous coordinates of the FDS-crystals centers, formed by the first program block, which makes the program slow.

The subprogram for calculating coordinates of the particle-"target" is used in case of modelling of the CL-photons scattering, since all the above-mentioned arguments do not require concrete definition of the nature of scattered particles.

Considering scattering of an electron or a photon on an FDS-crystal it is necessary, in the general case, to consider the following three possibilities: 1) elastic interaction; 2) inelastic interaction; 3) absorption. It is evident that by knowing probabilities of these processes,

it is not difficult to select a type of interaction by of the following two inequalities:

$$\gamma_9 < r \quad (19)$$

$$\gamma_9 < r + \eta \quad (20)$$

where r is the probability of elastic scattering, and η is the probability of inelastic scattering.

Fulfillment of the first inequality (19) corresponds to elastic interaction; fulfillment of only the second inequality (20) corresponds to inelastic scattering, and non-fulfillment of both inequalities corresponds to absorption. Coefficient r and η are unique to the type of flying incident particle.

A subprogram for choosing probabilities r and η is a part of the program. There is also a subprogram for choosing CL-efficiency α , depending upon the incident electron energy, since the electron energy between two subsequent collisions of electron with FDS-particles can change by an order of magnitude.

In this program, the generation of secondary electrons of medium energies (for example, Auger-electrons) is a result of absorption of a moving electron of high energy, this has to be taken into account while choosing values r and η . Generation and scattering of secondary electrons of excited FDS-crystals is considered in the program separately, since the coefficient of the real secondary emission may be more than one. Specific values r , η , and α are chosen for calculation of true secondary electron scattering.

The probability of inelastic scattering of photons of visible field of spectrum is extremely low and it may even be zero. However, this value can be derived from the program, which is therefore applicable for calculation of signals in the short-wave area (ultraviolet, X-ray) of the excited FDS-spectrum.

Formation of CL-Signal in FDS

Now let us consider the algorithm of the basic program operation. All scattering events on FDS crystals are considered for each electron (or photon) separately. During the first interaction a beam of electrons is scattered in the exposed crystal. During energy loss, the number of photons (N_{ph}) of CL-emission of an excited probed crystal is calculated.

In case of inelastic scattering:

$$N_{ph} = \alpha \cdot (\gamma_{10} \cdot E_0/E_{ph}) \quad (21)$$

In case of absorption

$$N_{ph} = \alpha \cdot (E_0/E_{ph}) \quad (22)$$

where α is the quantum efficiency of CL; γ_i is a random number; E_0 is the initial energy of the electron; $\gamma_i E_0$ represents the losses of energy in case of an inelastic scattering; E_{ph} is the energy of the CL-photon.

Then a cascade of scattering is considered for each photon, taking into account incidence angle ψ . As

a result of these scattering events a photon will be either absorbed or will leave the boundary of FDS. In the latter case one should check the condition:

$$\theta < \pi/3 \quad (23)$$

where θ is the spherical angle of direction of a photon movement after the last scattering event on FDS-crystal.

If condition (23) is fulfilled, we consider the collection of a photon by light detector. All CL-photons of the irradiated crystal which are excited by an incident beam and get in the collector form "true" signal of FDS-CL which is local CL of the probed crystal.

Now let us return to the type of interaction between a beam of electrons and the irradiated crystal. In case of absorption, the program chooses the next electron of the beam. If elastic or inelastic scattering has taken place, the program calculates a cascade of collisions within the FDS-crystals taking into account the initial angle of incidence till the electron is either absorbed or it leaves FDS.

For each event of inelastic electron interaction, the CL-photon scattering is considered in accordance with the above-mentioned procedure, but without taking into account the angle ψ . The CL-background signal is formed by photons of excited FDS-crystals, as well as photons knocked-on by backscattered electrons getting into the irradiated crystal, subject to fulfillment of condition (23). The value of the "true" CL-signal is accomplished when the program completes calculation of scattering for the last of the specified number of beam electrons. The background CL-signal value continues to increase during the second part of the program which is devoted to scattering of real secondary electrons. Each FDS-crystal is covered by the hit counter, which increases by one with each new collision of an incident or scattered electron with a crystal. The counter stops reading with either the absorption or emission of the electron from the FDS.

By multiplying the final reading of the counter for a FDS-crystal with the "true" secondary electron emission coefficient, δ , we obtain the number of "true" secondary electrons emitted from the crystal. In some case this number can obviously be zero.

The program selects all FDS-crystals ($i = 2 \dots N$) by addresses, and for each crystal, it calculates cascade of collisions of each secondary electron, thereby, if necessary, increasing the value of the CL-background signal. We assume that the trajectory of scattering does not depend on angle ψ , and the directions of movement of a "true" secondary electron is uniformly distributed in the full solid angle 4π . An exclusion is the direction of emission of the first δN_0 "true" secondary electrons from the irradiated crystal, knocked out by N_0 primary electrons. In this last case, we take into account the initial angle of incidence of electron beam (ψ) while calculating the spherical angles θ and ϕ .

The rest of the procedure of calculation of "true" secondary electrons scattering repeats the above procedure of the electron beam scattering.

Functional Potentialities of the Program

The model consists of a packet of programs in "Turbobasic" and is intended for estimation of real CL- and a part of topographical CL-contrast useful in the study of powdery objects in the SEM-CL mode. The program takes account of: 1) random character of the distribution of crystals in the FDS-volume; 2) cases of elastic and inelastic interaction, as well as absorption, of the electrons scattered from the primary beam during the interaction with FDS-crystals; 3) secondary electron emission: the generation, elastic and inelastic scattering, and absorption of "true" secondary electrons during interactions with FDS-crystals; 4) a change of monochromatic CL-radiation of each of the FDS-crystals for a probable case of the inelastic scattering and absorption of photons of radiation on the FDS-crystals; and 5) an angular aperture of the CL-radiation collector. As a result, for a fixed angle of electron beam incidence, this program calculates the ratio of the intensity of the CL-signal of radiated crystal to the total intensity of CL-signal for a complex of crystals, excited by the electron beam scattered in the FDS-volume (i.e., the ratio of the "true" to background signals), as well as the sum of these two intensities (the integral intensity of FDS CL).

This theory allows us to obtain: 1) spatial distribution of the area of excitation of the FDS crystals by the electron beam; 2) spectrum of FDS CL; 3) dependence of these characteristics upon the closeness and dimensions of crystals; 4) dependence of the integral CL-spectrum of FDS upon the electron beam incidence angle, which makes it possible to explain and to calculate a part of the topographical contrast of CL-image.

The disadvantage of this model is the insufficiently strict requirement of uniform random distribution within the specified boundaries of spherical angles θ and ϕ , of directions of scattering of high energy and real secondary electrons during each interaction with the FDS-crystals. On one hand, this model gives an approximate character to the calculations, taking into account the random character of scattering of an electron beam in the FDS. On the other hand, the calculations as well as the time for achieving the program aims are considerably reduced. This model is a good approximation for calculating electron scattering on crystals of non-spherical form and random orientation, which is the case for the majority of powdery materials.

Topographical Contrast of CL-images

The above model of electron beam scattering in the FDS was used for quantitative estimation of the topographical contrast. The program computes the scattering of a focussed electron beam of 20 keV impinging on the ideal FDS (consisting of 64 particles of spherical form, Fig. 6) and is a mathematical approximation of the real finely-dispersed materials.

The dependence of the normalized integral CL-intensity, $I_n(\psi)$ upon the electron beam incidence angle ψ

CL Contrast of Fine-Dispersed Structures

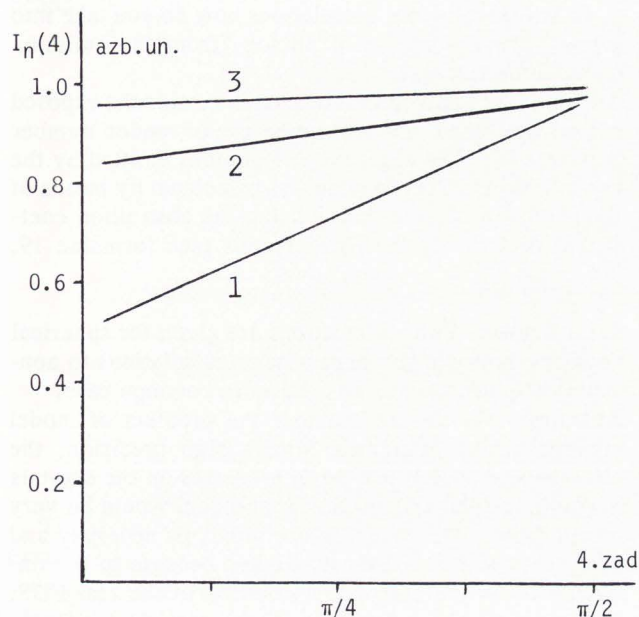


Figure 7. Function $I_n(\psi)$ in arbitrary units for $h_1 = 0$, $h_2 = 1/4$, $h_3 = 1/2$ (curves 1, 2 and 3 respectively), where I_n is the normalized intensity of integral CL of the FDS.

about the probed crystal surface was calculated and plotted (Fig. 7). Calculations were carried out for three crystals (1, 2 and 3) of the ideal FDS located at different depths $h_1 = 0$, $h_2 = 1/4$ and $h_3 = 1/2$ arbitrary units respectively. Function $I_n(\psi)$ are linear with the value of inclination angle depending upon parameter h as follows:

$$\text{for } h_1 \quad I_{n1} = 0.56 + 0.28 \psi \text{ arb. units} \quad (24)$$

$$\text{for } h_2 \quad I_{n2} = 0.88 + 0.08 \psi \text{ arb. units} \quad (25)$$

$$\text{for } h_3 \quad I_{n3} = 0.98 + 0.01 \psi \text{ arb. units} \quad (26)$$

On the basis of these results the angular contrast $C(\psi)$ was calculated:

$$C(\psi) = (1/I_n) (dI_n/d\psi) d\psi \quad (27)$$

where $d\psi = 1^\circ$ for the full interval $0 - \pi/2$.

Fig. 8 shows $C(\psi)$ corresponding to the three values of parameter h ; these curves have a form of hyperbolas:

$$\text{for } h_1 \quad C_1 = [0.5 / (0.56 + 0.28 \psi)] \% \quad (28)$$

$$\text{for } h_2 \quad C_2 = [0.14 / (0.88 + 0.08 \psi)] \% \quad (29)$$

$$\text{for } h_3 \quad C_3 = [0.02 / (0.98 + 0.01 \psi)] \% \quad (30)$$

For angles ψ close to $\pi/4$ and for value $h_1 = 0$, the value of topographical contrast of FDS CL is 2-2.5 times smaller and for the case of normal incidence

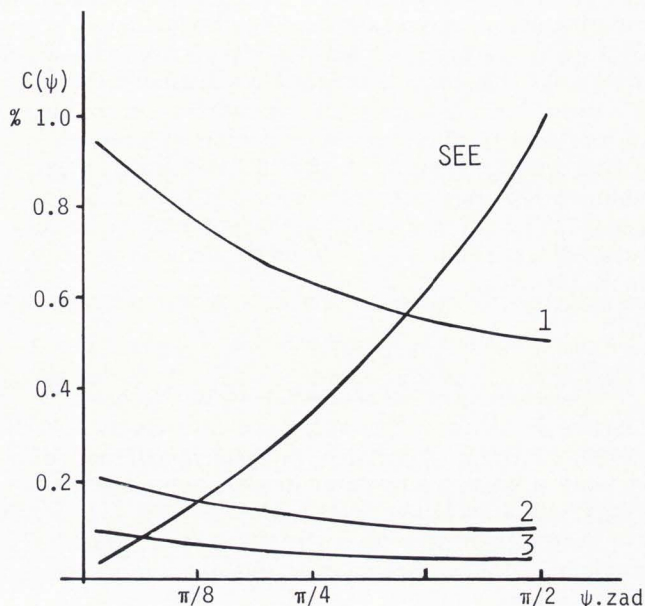


Figure 8. Function $C(\psi)$ in percentage for $h_1 = 0$, $h_2 = 1/4$, $h_3 = 1/2$ (curves 1, 2 and 3 respectively for CL-mode); SEE - for secondary electron emission mode. $C(\psi)$ is topographical component of contrast of the FDS CL-image.

($\psi \rightarrow 0$) it may exceed by several orders of magnitude the topographical contrast value calculated for the secondary electron emission (SEE) mode for solid monochromatic objects (Goldstein and Yakowitz, 1975). The curve corresponding to angular contrast in SEE is also shown in Fig. 8.

Comparing dependence of the topographical contrast values on h , the depth of the crystal, for the fixed angle of incidence ψ , Fig. 8 shows that the value of topographical contrast of CL-image of FDS-particles decreases with an increase in depth h . Therefore, it is possible to estimate the relative depth of location of FDS-crystals by the angular contrast.

Conclusions

Fine-dispersed structures cover a wide class of objects having a certain specificity while forming contrast in various modes of SEM operation. SEM CL-mode studies of $\text{CaSO}_4 \cdot 0.5\text{H}_2\text{O}$ and BaO with typical dimensions of 10-100 micrometers showed a new type of contrast. Understanding of the mechanism of origin of this contrast is very important for the interpretation of CL-images, particularly because of the presence of a number of artifacts during formation of the video signal:

- 1). Contrast due to the topography of particle surface exposed to electron beam.
- 2). Influence of the particle position in the FDS-volume upon the contrast.
- 3). Spectral changes (in intensity and wavelength) in the integral CL-signal due to various

contributions of several surrounding FDS-particles.

Physical model and calculations carried out have shown that the above-mentioned peculiarities of FDS CL-images in SEM are significant and can be recognized. Topographical contrast of particle under investigation depends on the depth of particle position in FDS-volume. Spectrum variation of detected CL-emission depends on the quantum yield and spectral composition of local CL-emission of particle under study as well as those surrounding it.

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Discussion with Reviewers

S. Myhajlenko: Have the authors considered the geometric aspects of light extraction as perhaps another source of the CL-aureoles, see the edges of the acicular crystals in Fig.3?

Authors: The CL-images of FDS-crystals have regular geometric shapes (circle, lines etc.). The authors conclude from this observation that the main reason of topographic contrast formation (in particular, the aureoles around the crystals) is the electron beam scattering in the FDS-volume. But this fact does not exclude the action of crystal geometrical shape for light emission from edges of the acicular crystals. This problem must be considered in additional work.

S. Myhajlenko: The authors comment on the depth resolution possible from measurements of angular contrast of FDS-CL. What assumptions have been made about the medium/volume containing the FDS?

Authors: We believe that it is possible to estimate (qualitative estimation only) the relative depth of location of FDS-crystals. This is not depth resolution.

J. F. Bresse: In your calculations how do you take into account the absorption of photons from the emission point to the surface?

Authors: The absorptions of photons inside the exposed crystal was taken into account by use of random number parameter γ . The absorption of photons emitted by the crystal into the FDS-volume was calculated by means of the photon scattering theory using the absorption coefficient of light by the FDS-crystals (see formulae 19, 20).

J. F. Bresse: Your calculations are given for spherical powders, how can you expand your calculations to non-spherical powders which is the more common case?

Authors: We do not consider the problem of model creation which calculates, with a high precision, the electron and photon scattering processes on the crystals having non-spherical shape. That model would be very complicated. Moreover, to our mind, its necessity and usefulness will be extremely limited because of the impossibility of the computer modelling of the real FDS. Consequently the calculation of the particle scattering processes on the non-spherical crystals carried out with the higher precision will have an approximate character. Our model allows us to explain the observed topographic contrast on the CL-images of FDS-crystals which have any geometrical shape. Furthermore, both random distribution and orientation in FDS-volume of the non-spherical shape and statistical characteristics of the electron scattering allow, in common case, to use the spherical crystal sizes as parameters defining the average efficient cross-section of electron scattering process. In this case it takes into account the dependence of this parameters on the scattered particle energy.

G. Remond: Besides the role of backscattered electrons could the observed contrasts (aureoles) result from the sum of CL-intensities originating from adjacent grains characterized by large differences in the decay times of their CL. Such a possibility is suggested by the contrast shown in Fig. 1. In the center of Fig. 1 the particle emitting blue is surrounded by blue lines parallel to the direction of the line-scan. Could you comment on possible phosphorescence effect?

Authors: Such effect could be observed for long persistence materials and fast scan rate.