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NUMERICAL CALCULATION OF THE F COLOR CENTERS DISTRIBUTION IN ADDITIVELY COLORED ALKALI HALIDE CRYSTALS

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Abstract
In this paper, we have realized a numerical simulation of the additive coloring of crystals, by solving the diffusion equation in the time dependent case. The numerical modeling is performed by using the finite element commercial code FIDAP. Using two-dimensional (2D) FIDAP numerical simulations, we have computed the distribution of the F color centers, during the additive coloring of the KCl crystal. We have found that the coloration of a KCl crystal, with the dimensions 2 mm × 2 mm × 20 mm, the diffusion constant $D = 8 \cdot 10^{-6}$ cm^2/s at temperature $T = 600$ K and the diffusion characteristic time $\tau_D = 8.4$ minutes, becomes very uniform after approximately 30 minutes. This result is in a very good agreement with the experimental measurements. A numerical computation of the coloration time for KCl crystals with various thickness values (4 mm, 6 mm, 8 mm and 10 mm) has also been performed, using two-dimensional (2D) geometry. The dependence of the coloration time on the crystal thickness has been analyzed.
Keywords: Color centers; Alkali halides; Numerical simulation.

1. Introduction

The F color center in alkali halide crystals is the simplest color center and it consists of an anionic vacancy in which an electron has been captured. Using additive method of F color centers formation, an excess of alkali metal ions is introduced in the crystal by heating the crystal into a vapor atmosphere of the ions to be introduce in the crystal [1, 2]. The coloring temperature must be between the melting point of the crystal and the temperature of colloid formation [2]. As a result of this method, the alkali metal atoms (in the vapor state) are captured on the external surfaces of the crystal, where they become ionized, deferring the valence electron to the crystal lattice. Because all the states in the valence band are occupied, the deferred electron can go only into the conduction band. Because the coloring temperature is high, the mobility of the crystal ions is also high, thus the thermal move can bring a negative halogen ion, from the crystal lattice, near the positive ion "condensed" on the crystal

surface. Consequently, an anionic vacancy remains in the crystal and it can capture the electron from the conduction band, forming an F color center (figure 1). The equilibrium state is attained after the diffusion of the F color centers through the crystal [1]. The concentration n of the F color centers in the crystal depends on time and can be computed by solving the diffusion equation (with the convective term equal by zero):

$$\frac{\partial n}{\partial t} = D \cdot \Delta n \ , \tag{1}$$

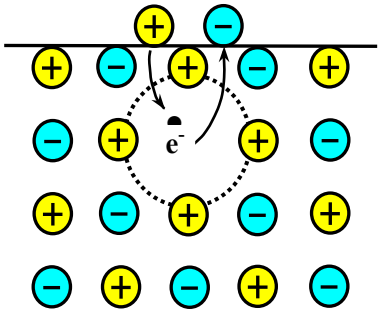

Fig. 1
The mecanism of additive crystals coloring.

where Δ is the Laplace's operator and D is the diffusion constant of the crystal, which depends on temperature according to the empirical law:

$$D(T) = D_0 \exp\left(-\frac{T_0}{T}\right) . \tag{2}$$

In relation (2), D_0 is the diffusion constant at temperature T_0. The diffusion constant for the KCl crystal is $D_0 = 1.22 \cdot 10^2$ cm^2/s at temperature $T_0 = 14430$ K [2]. Therefore, if the additive coloring temperature is $T = 600$ °C, the diffusion constant of the KCl crystal is $D = 8 \cdot 10^{-6}$ cm^2/s.

2. Numerical simulation

We have realized a numerical simulation of the additive coloring of KCl crystals, by solving the diffusion equation in the time dependent case. The numerical modeling is performed using the finite element commercial code FIDAP.

The simulation is realized for crystals with the length of 20 mm and different thicknesses: 2, 4, 6, 8 and 10 mm, using two-dimensional (2D) geometry, for the transversal quadratic section through the center of the crystal. Previous three-dimensional (3D) simulations show the same results for the optimal coloration time as this 2D simulation case. The simulation grids have been generated by using the GAMBIT program. They contain respectively 100, 400, 900, 1600 and 2500 quadrilateral elements, each element having the dimensions 0.2 mm × 0.2 mm.

The diffusion equation (1) is solved in the non-dimensional form:

$$\frac{\partial N}{\partial t} = D \cdot \Delta N , \tag{3}$$

where $N = n / n_0$ and n_0 is the equilibrium concentration of the F color centers at the crystal external surface.

Because the crystal is introduced into a vapor atmosphere of alkali metal atoms, these atoms are captured on all the external surfaces of the crystal. Thus, the F color centers concentration is almost constant on the all crystal external surfaces. We have considered as boundary conditions $N = 1$ at all domain frontiers. The initial concentration of the color centers in the crystal has been considered $N = 0$. The transient numerical analysis shows the evolution in time of the F color centers distribution in the crystal.

3. Results and discussions

We have analyzed the color centers distribution during the additive coloring of KCl crystals with different thicknesses. Figure 2 shows the isoconcentration curves in the crystal with the thickness of 2 mm, after 30 minutes of additive coloration. The legend of the figure 2 shows the dimensionless concentration values, N. The minimum concentration value (N_{min}) is obtained in the center of the crystal and the maximum concentration is always constant ($N_{max} = 1$) on the domain frontiers. We can observe that the coloration of this crystal becomes rather uniform after approximately 30 minutes. This result is in excellent agreement with the Mollenauer's experimental measurements [2].

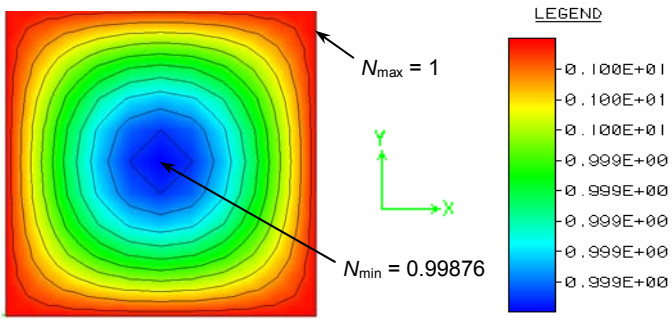

Fig. 2
Isoconcentration lines of the KCl crystal with 2 mm thickness, after 30 minutes of additive coloring.

In figure 3 the evolution in time of the difference between the maximum value of the concentration, N_{max} , and the minimum value, N_{min} , is represented for KCl crystals with different thickness values: 2 mm, 4 mm, 6 mm, 8 mm and 10 mm.

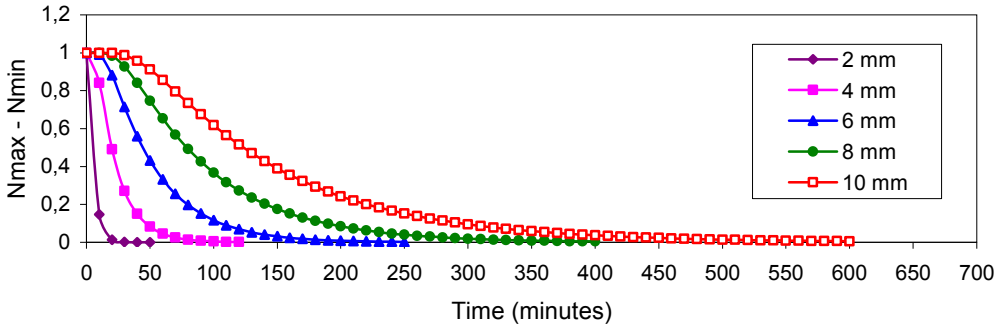

Fig. 3 Evolution in time of the difference between the maximum and the minimum concentration of the color centers in KCl crystals with various thickness values.

Figure 4 presents the dependence of the coloration time (after which the color centers concentration becomes rather uniform: N_{max} - N_{min} = 0.01) on the KCl crystal thickness. It can be observed a significant increase of the optimal coloration time as function of crystal thickness (570 minutes for a 10 mm crystal thickness).

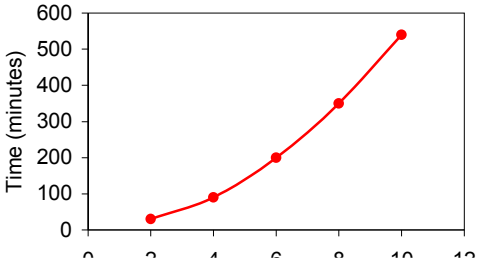

Fig. 4 The dependence of the coloration time on the KCl crystal thickness.

The FIDAP code has been used in order to compute the distribution of the F color centers during the additive coloring of the KCl crystals. It is found that the coloration of a KCl crystal with the thickness of 2 mm become rather uniform after about 30 minutes. This result is in a good agreement with the experimental measurements. The optimal coloration time of KCl crystals with various thickness values (4 mm, 6 mm, 8 mm and 10 mm) has been numerical computed. The numerical results show a significant increase of the optimal coloration time when the crystal thickness increases. This kind of simulation can be used in order to optimize the coloration time for the alkali halide crystals.

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