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FIDAP simulation of the F color centers formation in alkali halides crystals using additive method of crystals coloring

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In this paper we have performed a numerical simulation of the additive coloring of the crystals by solving the time dependent diffusion equation. The finite element commercial code FIDAP has been used for the numerical modeling. The numerical results are in agreement with the experimental measurement and show that, after an exposure to the vapor of 30 minutes, the coloration of a KCl crystal, with the dimensions 2 mm × 2 mm × 20 mm and with the diffusion constant $D = 8 \times 10^{-6}$ cm^2/s at temperature T = 600 K, becomes very uniform. A numerical optimization of the coloration time for a two-dimensional (2D) and three-dimensional (3D) geometry is performed. The excellent agreement between the 3D numerical results and the experimental measurement shows that the 3D simulation is more realistic than the 2D case.

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1. Introduction

The color centers in ionic crystals are anionic (or cationic) vacancies of the crystal lattices in which electrons (or holes) have been captured. 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 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 (Fig. 1).

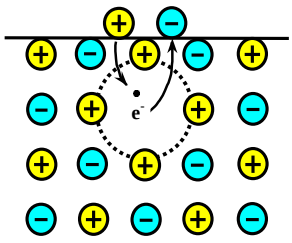


Fig. 1. The mechanism of additive crystals coloring.

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 depends of time and of the position in the crystal and it can be computed by solving the diffusion equation (with the convective term equal by zero):

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

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. As an example, for the KCl crystal, the diffusion constant is $D_0 = 1.22 \times 10^2$ 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 $D = 8 \times 10^{-6}$ cm^2/s.

In the follows, we present the numerical solution of (1) and (2), with appropriate initial and boundary conditions.

2. FIDAP 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 performed for a two-dimensional (2D) and a three-dimensional (3D) geometry. For the 2D case, we have considered a crystal with the thickness of 2 mm and the length of 20 mm. For the 3D geometry, a crystal with dimensions 2 mm × 2 mm × 20 mm has been considered. The crystal mesh has been generated by using the GAMBIT program, and contains 500 quadrilateral elements for the 2D case and 5000 brick elements for the 3D case. In the Fig. 2 is presented the crystal mesh for the 3D geometry.

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

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

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 the boundary condition at all the domain frontiers $N = 1$ and the initial concentration of the color center in the crystal has been considered $N = 0$.

So, by using the FIDAP numerical simulation, we have obtained the solution of the diffusion equation in all nodes of the mesh, at successive moments in time.

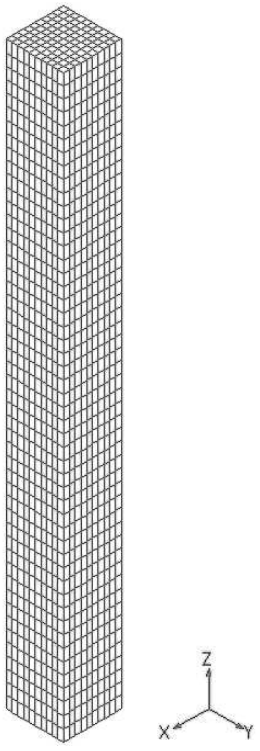


Fig. 2. Mesh with 5000 brick elements for the 3D simulation.

3. Results and discussions

We have analyzed the color centers distribution during the KCl crystal additive coloring. Fig. 3 shows the isoconcentration curves in the crystal obtained in the case of a 2D simulation. We observe that the coloration of the crystal become rather uniform after approximately 30 minutes.

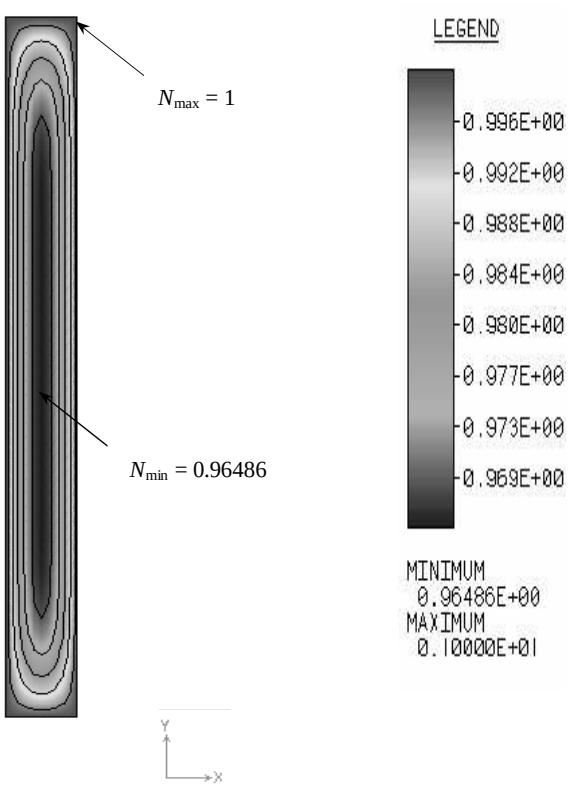


Fig. 3. Isoconcentration lines after 30 minutes of KCl crystal additive coloring. (2D geometry).

For the 3D geometry case, we have found that after 30 minutes, the distribution of the F color centers into the KCl crystal become very uniform. Figs. 4 a, b present the isoconcentration curves obtained in two perpendicular planes after 30 minutes of crystal coloration.

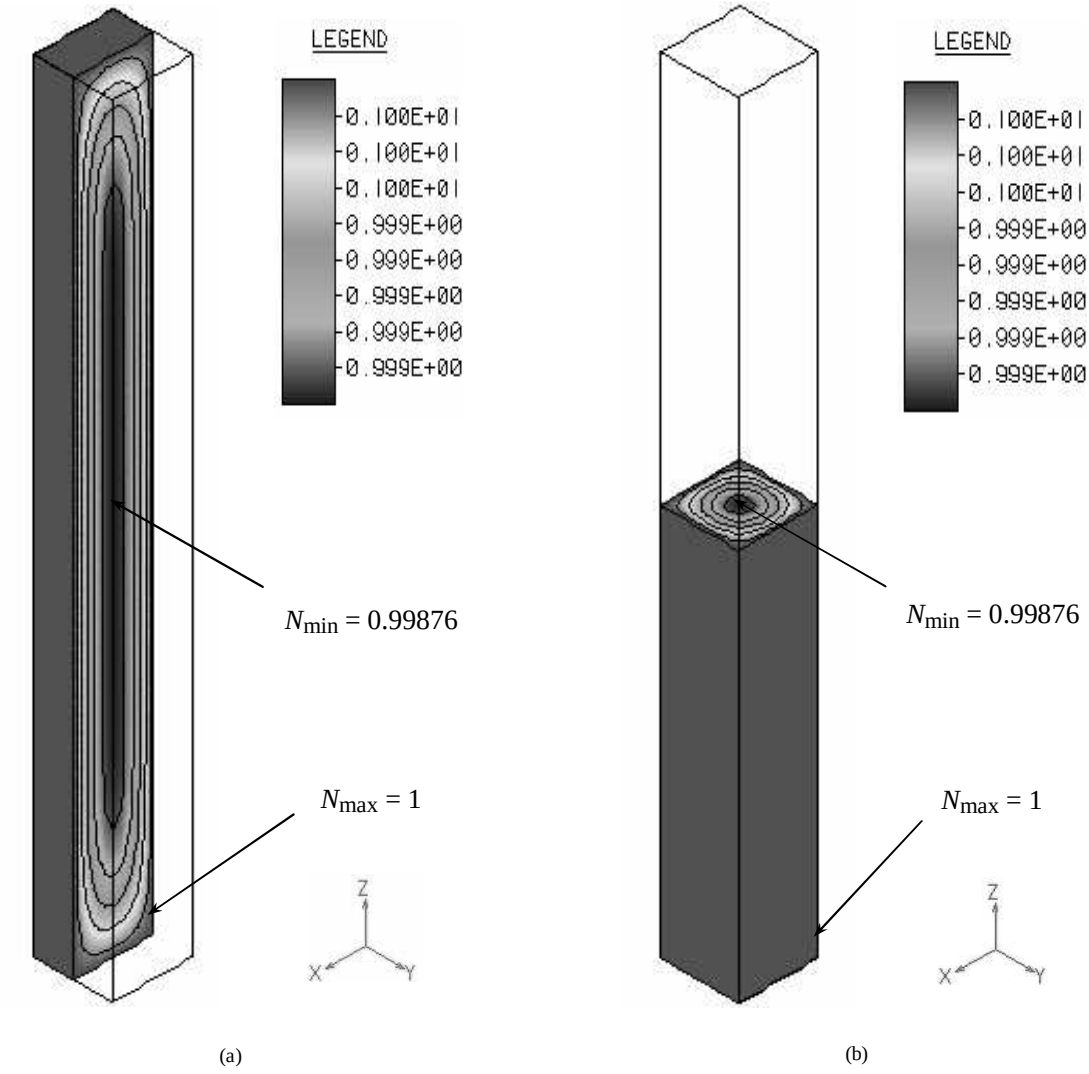
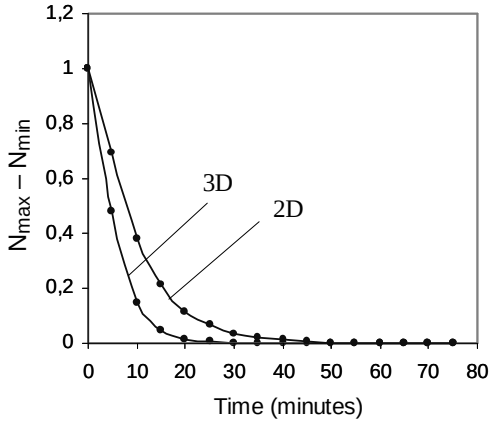


Fig. 4. Isoconcentration lines in two perpendicular planes after 30 minutes of additive coloration. (3D geometry).

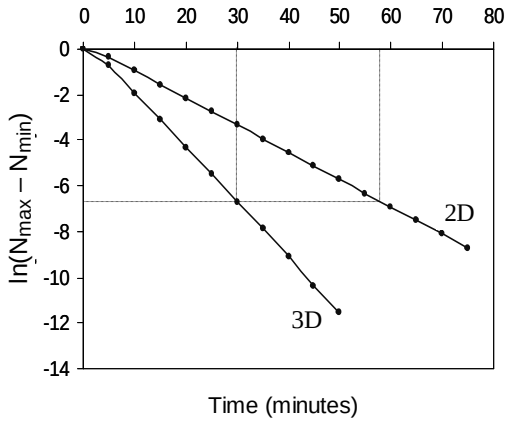
The legends of the Figs. 3 and 4 present the dimensionless concentration values N. The minimum concentration value (N_{min}) is obtained in the center of the crystal and the maximum concentration is always constant (N_{max} = 1) on the domain frontiers. We observe that, after 30 minutes of the coloring, the value N_{min} = 0.96486 obtained in the 2D case is less than the value N_{min} = 0.99876 computed in the 3D case. So, the numerical results show that the color centers distribution becomes uniform more rapidly in 3D case than in the 2D case.

The results are in good agreement with the Mollenauer's experimental measurements [2], which show that for a slab of KCl 2 mm thick, at 600 °C, an exposure to the vapor of 30 minutes or more duration should result in a coloration more than sufficiently uniform for most purposes.

In Fig. 5 a, b, 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 the both 2D and 3D cases.



(a)

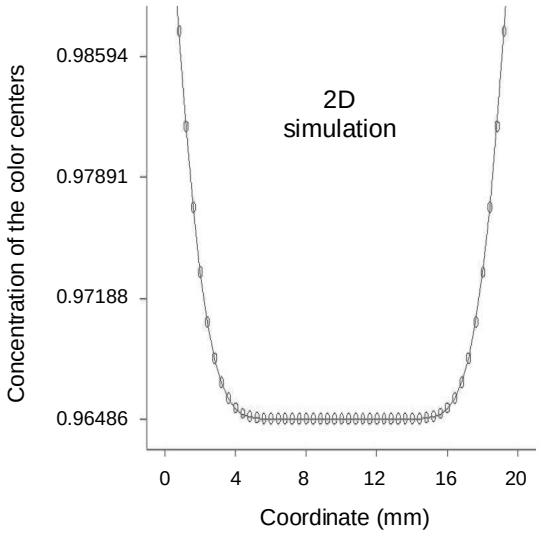


(b)

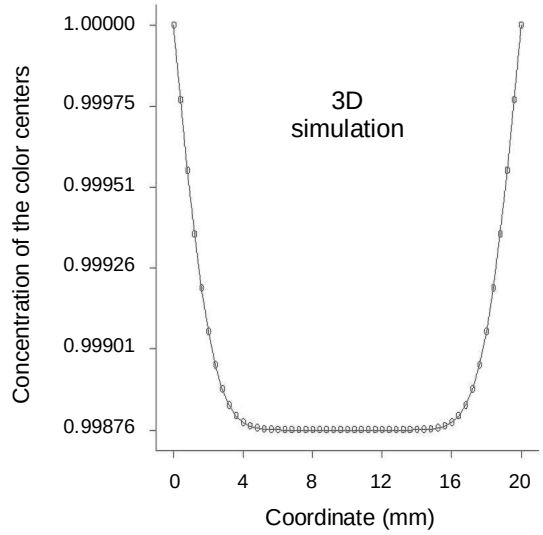
Fig. 5. Evolution in time of the difference between the maximum and the minimum of the color centers concentration in KCl crystal, in the 2D and 3D cases.

In Fig. 5 b, we can observe that the coloration of the crystal obtained after approximately 30 minutes in the 3D simulation is attained after about 58 minutes in the case of 2D simulation. Thus, the 3D simulation more realistic describes this process than the 2D modeling.

Fig. 6 presents the distribution of the color centers on the vertical symmetry axis of the crystal, obtained from a 2D simulation (a) and 3D simulation (b).



(a)



(b)

Fig. 6. Distribution of the color centers on the vertical symmetry axis of the crystal (a) – the 2D case, (b) – the 3D case.

We observe that, in the both 2D and 3D cases, the F centers concentration varies on the 4 mm length near the crystal external surface and remains almost constant on 12 mm length near the center of the crystal.

4. Conclusions

Using bi-dimensional (2D) and tri-dimensional (3D) FIDAP numerical simulations, we have computed the

distribution of the F color centers during the additive coloring of a KCl crystal.

- We have found that, the coloration of the KCl crystal become rather uniform after about 58 minutes in the 2D case and after approximately 30 minutes in the 3D case. This is an expected result, since the 3D model accounts for the diffusion through all six lateral faces, while the 2D model considers only four of them.
- The 3D simulation results are in a good agreement with the experimental measurements.
- Analyzing the F color centers distribution along the KCl crystal axis, we have found that, in the both 2D and 3D cases, the F centers concentration varies on the 4 mm length near the crystal external surface and remains almost constant on 12 mm length near the center of the crystal.
- This kind of simulation can be used in order to optimize the coloration time for the alkali halides crystals.

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