# Defect Creation Under Reactor Neutron Irradiation in (Li,Na)F Based Crystals<sup>1</sup>

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Abstract – The neutron induced defects in activated LiF and NaF single crystals have been investigated by studying of the absorption and luminescence spectra. The results are presented and discussed.

## 1. Introduction

The problem of defects creation in alkali-halide crystals under various type of radiation (gamma-photon, electrons, ions and neutrons) is still very actual now as many years before [1–11]. In this paper the results of investigation of neutron induced defects in activated LiF and NaF single crystals by measuring the absorption and pulsed cathodoluminescence (PCL) spectra are presented and analyzed.

## 2. Specimens and methods

All of the (Li,Na)F based specimens have been grown in the Alybakov's laboratory in the Institute of Physics of Kyrgyzstan NAS by Kiropoulos method in platinum crucible in air. So, all of the specimens included the oxygen impurities. For experiments there were 8 specimens: LiF:Sc, LiF:U, NaF:Eu, NaF:Sr, NaF:Yb, NaF:0.01%U, NaF:0.1%U, NaF:U,Ti chosen. The specimens were 64640.7 mm<sup>3</sup> (in average) and 0.2–0.3 g in weight.

The irradiation has been made by reactor neutrons. The trial dose of irradiation (fluence) has been chosen as  $10^{16}$  and  $10^{18}$  cm<sup>-2</sup>. But as it was found before by A.A.Vorobiev [1] and E.L.Andronikashvili [2] the dose of  $10^{18}$  cm<sup>-2</sup> led to partial destruction and partial splitting of the materials and also to sufficient change of specimen density. So for systematic irradiation the dose of  $10^{16}$  cm<sup>-2</sup> was selected for all of the specimens. The specimens were wrapped by the aluminum foil and irradiated by reactor neutrons. The nuclide analysis in 10 days after irradiation has been made by gamma-spectrometer URS-2-SF. The <sup>24</sup>Na, <sup>152</sup>Eu and <sup>46</sup>Sc isotopes have been found. The absorption and PCL

spectra of specimens have been measured in 14 months after neutron irradiation. So the parameters only for time stable defects (more than one year) in (Li,Na)F based specimens have been investigated.

The optical absorption spectra were measured by Helois Alfa spectrophotometer ( $\lambda$  range from 190 to 1100 nm). The PCL spectra were measured by using KLAVI-type setup with HTD-polychromator (200-830 nm) and the CCD-camera. The PCL was excited by the electron beam (current density 700-1000 A/cm<sup>2</sup>, pulse duration 2 ns, energy 150-250 keV). All measurements have been made at room temperature.

#### 3. Results and discussion

The absorption and PCL spectra of the investigated samples are shown at Fig. 1 and Fig. 2 respectively.

## 3.1. Absorption spectra

For both LiF specimens there is the common position of F bands at 240 nm. There is the band at 195-200 nm connected with  $(O^{-}V_a)$  centers [5]. The band at 285–298 nm can be connected with Fisher  $O^{-}$  band [8]. For LiF:Sc the bands at 306 (F<sub>3</sub>(R<sub>1</sub>)), 377 (F<sub>3</sub>(R<sub>2</sub>)), 440-460 (F<sub>2</sub>+F<sub>3</sub><sup>+</sup>) and 520-545 nm (F<sub>3</sub><sup>+</sup><sub>A</sub>) were found. Identification of the bands was made using data from [6]. For LiF:U specimens there is total absorption in the whole visible range. The radiation stability of LiF:U is worse than that of LiF:Sc. So, uranium impurity decreases the radiation stability of LiF specimens. The total absorption at 190–700 nm for LiF:U is connected with large amount of Li colloidal particles and different color centers.

For all NaF specimens the common position of F bands at 340 nm is observed. The bands connected with oxygen ( $O^{--}V_a$ -centers) were found at 310, 210–218 and 194–202 nm. Earlier these bands have been found at ~ 309, 220 and 196 nm and described in [5].

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Fig. 1. Absorption spectra of LiF:Sc (*a*), LiF:U (*b*), NaF:Eu (*c*), NaF:Sr (*d*), NaF:Yb (*e*), NaF:0.01%U (*f*), NaF:0.1%U (*g*) and NaF:U,Ti (*h*) specimens irradiated by neutron (fluence  $10^{16}$  cm<sup>-2</sup>)



Fig. 2. PCL spectra of LiF:Sc (*a*), LiF:U (*b*), NaF:Eu (*c*), NaF:Sr (*d*), NaF:Yb (*e*), NaF:0.01%U (*f*), NaF:0.1%U (*g*) and NaF:U,Ti (*h*) specimens irradiated by neutron (fluence  $10^{16}$  cm<sup>-2</sup>)

So, the stable UV-bands of oxygen-vacancy  $(O^{-}V_a$ -centers) types have been found in all neutron irradiated (Li,Na)F based specimens.

For all neutron irradiated NaF:Me specimens the stable aggregate color center have been found at ~ 430–435 ( $F_3(R_1)$ ), 480–490 ( $F_2$ ), 520 ( $F_3^+$ ) and 615–625 nm ( $F_3^-(F_4)$ ). The availability of  $F_3^-(F_4)$  color center in all irradiated NaF:Me specimens points to higher degree of aggregation of centers up to dimer formation. The results are in a good agreement with data [5]. The search of  $F_2^+$ -centers (laser active centers, absorption at 725–740 nm) at neutron irradiated NaF:Me specimens was negative.  $F_2^+$ -centers have not been found in our experiments in 14 months after irradiation of NaF:Me crystals due to their unstable.

## 3.2. Luminescence spectra

For LiF:Sc specimen in luminescence spectrum the band of  $F_3^+$  (536 nm) and  $F_2$  (642 nm) color centers

are dominated (Fig. 2*a*). For LiF:U the main band at 523 nm is connected with  $U^{6+}$  centers (Fig. 2*b*).

For NaF:Me without uranium impurity (Fig. 2 *c*, *d*, *e*) the  $F_3^+$  (540 nm) and  $F_2$  (640 nm) bands in form of common band are dominated. In the specimens with uranium impurity the bands of U<sup>6+</sup>-centers (~550 nm) are dominated. The influence of  $F_2$ -centers on luminescence spectra of NaF:U (Fig. 2 *f*, *g*, *h*) at 640 nm region is noticeable.

So, the aggregate  $F_2$  and  $F_3^+$  color centers give the main contribution to the intrinsic luminescence bands of (Li,Na)F neutron irradiated specimens. The stable weak bands at 700–740 nm in PCL spectra of neutron irradiated (Li,Na)F:U based crystals (Fig. 2) can be determined (according to [9, 10]) by  $F_2Me^{n+}V_c$  or  $F_2Me^{n+}$ -centers. The formation of F,  $F_2$  and  $F_3^+$  color centers in (Li,Na)F:Me and their luminescence can be describe by Lisitsina reactions [11] modified by taking into account the influence of impurities (Sc, U, ...):

$$\begin{split} \mathcal{O}^{--}V_{a}^{+} &\to \mathcal{O}^{-} + F; \quad V_{a}^{+} + (F,F^{-}) \to (F_{2}^{+},F_{2}); \quad F + F \to F_{2}(S_{0}); \\ F_{2}^{+} + e \xrightarrow{S_{c}} F_{2}(S_{j}) \cdots F_{2}(S_{1}) \xrightarrow{\tau_{0}} F_{2}(S_{0}) + h\nu(F_{2}); \\ F_{2}^{-} + V_{k} \xrightarrow{\tau_{\nu}} F_{2} + e_{im}^{0} \Big| \xrightarrow{S_{c}} F_{2}(S_{1}) \xrightarrow{\tau_{0}} F_{2}(S_{0}) + h\nu(F_{2}); \\ F_{2}(S_{0}) + p \to F_{2}^{+} + e_{im}^{0} \to (F_{2}^{+})^{*} \xrightarrow{\tau_{0}} F_{2}^{+} + h\nu(F_{2}^{+}); \\ F_{3} + p \Big| \to F_{3}^{+} + e_{im}^{0} \Big| \xrightarrow{U^{n+}} (F_{3}^{+})^{*} \xrightarrow{\tau_{0}} F_{3}^{+} + h\nu(F_{3}^{+}) + h\nu(U^{n+}), \end{split}$$

where  $S_j$  is *j*-excited state of luminescence centers;  $e_{im}^0$  is exciton localized about impurity

### 4. Conclusion

The some parameters of stable (during 1 year and more) aggregate color centers in (Li,Na)F:Me compounds irradiated by reactor neutron are presented. The important role of colloidal particles in absorption spectra of LiF:(Li,Sc) compound are marked. It is found that impurity of *f*-elements (U<sup>6+</sup> and Eu<sup>3+</sup>) leads to increasing of radiation sensivity of LiF and NaF specimens. The F<sub>2</sub>, F<sub>3</sub><sup>+</sup> and U<sup>6+</sup> centers are dominated at PCL spectra.

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