

## Peculiarities of doping KBr-2SrBr<sub>2</sub> melt with europium at 973 K

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The processes of Eu<sub>2</sub>O<sub>3</sub> solubilization in molten KBr-2SrBr<sub>2</sub> at 973 K were studied from the viewpoint of introducing rare-earth dopants in melts in situ for obtaining activated halide scintillators. The control of the solubilization was performed on the basis of current oxide-ion molality ( $m_{O^{2-}}$ ) which was detected by the potentiometric method. A membrane oxygen electrode Pt(O<sub>2</sub>)|YSZ was used as an indicator one. The process of Eu<sub>2</sub>O<sub>3</sub> solubilization in KBr-2SrBr<sub>2</sub> melt with the formation of Eu<sup>2+</sup> does not occur due to slight solubility of EuO or EuOBr. The solubility product of EuO in the studied melt is estimated as  $2.8 \cdot 10^{-7} \text{ mol}^2 \cdot \text{kg}^{-2}$ . The carbohalogenation process (treatment with ‘C+Br<sub>2</sub>’ red-ox couple) provides not only the dissolution of oxide, but also complete Eu<sup>3+</sup>-Eu<sup>2+</sup> reduction. The kinetic study of the carbohalogenation process in KBr-2SrBr<sub>2</sub> melt containing suspension of Eu oxocompounds show that in this case the plateau-like section arises in ‘pO-time’ dependences due to participation of two solid substances (carbon and Eu oxocompounds) in the reaction. The data corresponding to the end of this plateau-like section give another option to estimate the solubility product of EuO as  $1.6 \cdot 10^{-7} \text{ mol}^2 \cdot \text{kg}^{-2}$  that is in a good agreement with the above value (taking into account the experimental errors). The dissolution of Eu<sub>2</sub>O<sub>3</sub> (at the addition of  $m_{Eu_2O_3} = 5 \cdot 10^{-3} \text{ mol} \cdot \text{kg}^{-1}$ ) is finished in 40 min and the total deoxidization process is finished in 90 min whereas in the reference experiment the duration of the deoxidization of ‘pure’ melt is 50 min.

**Keywords:** potassium bromide, strontium bromide, europium oxide, melts, solubility, carbohalogenation.

**Особливості допущання розплаву KBr-2SrBr<sub>2</sub> європієм (II) при 973 К.** В.Л. Чергинець, О.Л. Ребров, Т.П. Реброва, Т.В. Пономаренко, Ю.М. Дацько, А.Г. Варич, О.І. Юрченко, В.В. Соловійов

Процеси розчинення Eu<sub>2</sub>O<sub>3</sub> у розтопі KBr-2SrBr<sub>2</sub> при 973 К були досліджені з точки зору введення in situ рідкісноземельних допантів в ростові розтопи для одержання галогенідних сцинтиляторів. Контроль процесу розчинення проводили, виходячи з поточної концентрації of оксид-іонів (.), яку визначали потенціометричним методом. У якості індикаторного використовували мембранний оксигеновий електрод Pt(O<sub>2</sub>)|YSZ. На противагу йодидним розтопам процес прямого розчинення Eu<sub>2</sub>O<sub>3</sub> в розтопі KBr-2SrBr<sub>2</sub> з утворенням Eu<sup>2+</sup> не протікає повністю внаслідок значно нижчих відновних властивостей іонів Br<sup>-</sup> у порівнянні з іонами I<sup>-</sup>. Добуток розчинності EuO у досліджуваному розтопі оцінено, як  $2,8 \cdot 10^{-7} \text{ моль}^2 \cdot \text{кг}^{-2}$ . Процес карбогалогенування (обробка окиснювально-відновною парою ‘C+Br<sub>2</sub>’) забезпечує не

тільки розчинення оксиду, але й повне відновлення Eu<sup>3+</sup> до Eu<sup>2+</sup>. Кінетичне дослідження процесу карбогалогенування розтопу KBr-2SrBr<sub>2</sub>, що містить суспензію оксидів Eu показує, що в цьому випадку на залежностях «рО-час» виникає платоподібна ділянка, а дані, що відносяться до кінцевої частини цієї ділянки, дають іншу можливість оцінити добуток розчинності EuO як 1,6·10<sup>-7</sup> моль<sup>2</sup> кг. Розчинення останнього (при добавці Eu<sub>2</sub>O<sub>3</sub> 5·10<sup>-3</sup> mol·kg<sup>-1</sup>) закінчується через 40 хв, а повний процес розкислення займає 90 хв в той час, як у холостому експерименті (без Eu<sub>2</sub>O<sub>3</sub>) тривалість процесу розкислення складає 50 хв.

## 1. Introduction

The processes of rare-earth (RE) oxide solubilization in ionic melts attract attention of specialists working in different branches of science and industry due to several reasons:

- these processes can be used for the extraction of RE compounds from ores and for the preparation of electrolytes for electrowinning free RE [1];
- it is well-known that several RE are non-radioactive analogs of actinides (La-Ac, Eu-Am), therefore, RE compounds and their solutions in different media are used as safe model objects for studying processes of radioactive pollutants extraction from solutions [2];
- the results on RE oxide dissolution are important for treatment of used nuclear fuel [3, 4];
- these processes can be used for the preparation of RE-halide containing charge for the growth of RE-activated optical materials [5].

First three items can be referred to directions developing for a relatively long time while the last one originates from the beginning of 'new era' of scintillation materials, i.e., the development of new halide materials containing Ce<sup>3+</sup> and Eu<sup>2+</sup> as activators [6–8].

The purpose of the present work is to investigate processes of Eu<sub>2</sub>O<sub>3</sub> dissolution in KBr-2SrBr<sub>2</sub> melt corresponding to the K(Sr<sub>1-x</sub>Eu<sub>x</sub>)<sub>2</sub>Br<sub>5</sub> scintillation material [9] with and without the action of reactive gas medium.

## 2. Experimental

The treatment of the obtained results was performed on the basis of current or equilibrium molalities of oxide ions in the melt which was determined by the potentiometric method with the use of the indicator electrode reversible to oxide ions.

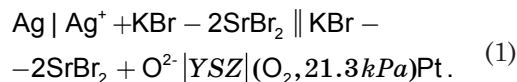
Chemicals and gases for the performing of the experiments were prepared as it was described in [10]. Ar+Br<sub>2</sub> gas mixture was obtained by passing the dry argon through the saturator with bromine thermostated at 0 °C (vapor pressure of Br<sub>2</sub> at this temperature was 8.6 kPa).

The construction of the potentiometric cell was as follows. Its shell was quartz test tube of 65 mm diameter (Ø) and height (h) of 125 mm with chamotte cover. In the cover there were holes for reference and indicator electrodes, thermocouple, alundum tubes for gas and solids (KOH or Eu<sub>2</sub>O<sub>3</sub>) supply. Alundum crucible-container for KBr-2SrBr<sub>2</sub> melt of Ø=25 mm (bottom), Ø=55 mm (top) and h=55 mm was filled with 50 g of the KBr-2SrBr<sub>2</sub> melt charge (9.7 g of KBr and 40.3 g of SrBr<sub>2</sub>) and placed in the shell.

The reference electrode itself was silver wire immersed in the bromide melt containing 0.1 mol·kg<sup>-1</sup> of Ag<sup>+</sup>. Alundum test-tube of Ø=8 mm (outer), Ø=5 mm (inner) and h=160 mm was used as a container for the reference electrode.

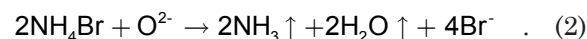
The indicator oxygen electrode Pt(O<sub>2</sub>)|YSZ was platinum foil attached to platinum wire placed inside the YSZ test-tube of Ø=8mm (outer) and h=160 mm. YSZ was sintered solid electrolyte ceramics ZrO<sub>2</sub>(Y<sub>2</sub>O<sub>3</sub>) with the yttria content of 10 mol. %. The partial pressure of oxygen in the electrode was 21.3 kPa (air).

The electrochemical scheme of the cell can be presented by such a manner:

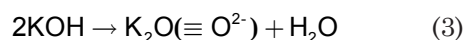


After assembling the shell the supply of argon was switched on and it was placed in tube furnace (Ø=75 mm) and heated to provide the necessary temperature. The temperature control was performed with the use of Pt-Pt(Rh) thermocouple.

When the necessary temperature was achieved the melt was deoxidized using ammonium bromide:



Before the carbohalogenation investigations the potentiometric cell (1) was calibrated with KOH of reagent grade. Its dissociation in melts occurs according to the equation:



Then series of KOH weights were recalculated in oxide ion molalities ( $m_{O^{2-}}$ ,  $2\text{KOH} \equiv \text{O}^{2-}$ ) and in  $pO$  values (where  $pO \equiv -\log m_{O^{2-}}$ ). After addition of each weight the equilibrium  $emf$  value ( $E$ ,  $V$ ) was measured. The measurements were performed by the Pogendorff method with the use of P-309 potentiometer. The  $emf$  value was considered as equilibrium one if its oscillations within 10 min did not exceed 0.001 V and the directed  $emf$  drift was absent.

The dissolution of  $\text{Eu}_2\text{O}_3$  in  $\text{KBr-2SrBr}_2$  melt was studied by sequential addition method (SAM) similar to the calibration one. Before the beginning of this experiment oxide ions entered in the melt by additions of KOH were removed by  $\text{NH}_4\text{Br}$  according to Eq. (2). After the saturation the weight of  $\text{Eu}_2\text{O}_3$  corresponding to  $\text{Eu}_2\text{O}_3$  total concentration of  $5 \cdot 10^{-3} \text{ mol kg}^{-1}$  was added to the melt together with graphite powder.

The addition of  $\text{Eu}_2\text{O}_3$  powder to the melt resulted in the  $emf$  shift. The bubbling of argon saturated with bromine vapor was started after this shift ended. The rate of the gas mixture passing through the melt was  $150 \text{ cm}^3 \cdot \text{min}^{-1}$  that provided the constant concentration of bromine in the atmosphere over the studied melt. The  $emf$  measurements were performed each 30 sec for first 10 min, each minute in 10 to 20 min range and each 2 min after 20 minutes of the carbobromination process.

Before the carbobromination of  $\text{Eu}_2\text{O}_3$  the process of the carbobromination of  $\text{KBr-SrBr}_2$  containing ca.  $10^{-2} \text{ mol kg}^{-1}$  of  $\text{O}^{2-}$  was studied. Its conditions were similar to the carbobromination of  $\text{Eu}_2\text{O}_3$ .

### 3. Results and discussion

#### 3.1. The calibration plot and processes accompanying $\text{Eu}_2\text{O}_3$ dissolution in $\text{KBr-2SrBr}_2$ melt

The calibration ' $emf(E)$ - $pO$ ' plot is presented in Fig.1, plot 1, 1'. It can be conditionally divided in two sections. The point 1' corresponds to added oxide ion molality ( $m_{O^{2-}}^0$ ) comparable with residual molality of  $\text{O}^{2-}$  after the deoxidization with  $\text{NH}_4\text{Br}$  ( $m_{O^{2-}}'$ ), i.e.,  $m_{O^{2-}}^0 \sim m_{O^{2-}}'$  whereas the points belonging to plot 1 correspond to oxide ion concentrations considerably exceeding the residual  $\text{O}^{2-}$  molality,  $m_{O^{2-}}^0 \equiv m_{O^{2-}}'$ .

The plot 1 was treated by least squares method to obtain the ' $E$ - $pO$ ' dependence in the form:

$$E = E_0 + \theta \cdot pO, \quad (4)$$

(where  $E$  and  $E_0$  the measured and the standard  $emf$  values of cell (1),  $V$ ,  $\theta$  the slope,  $V$ ), namely:

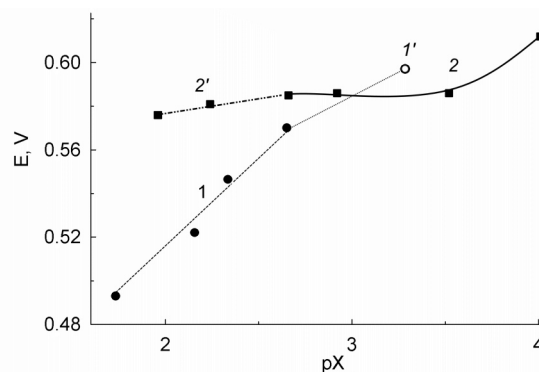


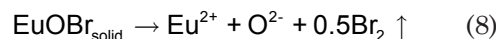
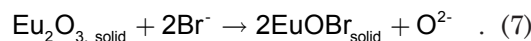
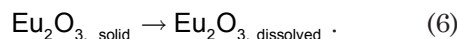
Fig. 1. The dependences of  $emf$  of cell (1) vs.  $pO$  (1, 1',  $pX \equiv pO$ ) and vs. negative logarithm of  $\text{Eu}_2\text{O}_3$  molality ( $pX \equiv -\log m_{\text{Eu}_2\text{O}_3}^0$ ) in  $\text{KBr-2SrBr}_2$  melt at 973 K.

$$E = 0.345(\pm 0.018) + 0.085(\pm 0.008) \cdot pO, \text{ V}, \quad (5)$$

here and after the standard deviations are used for the statistical estimations of the results.

Since  $emf$  value of cell (1) for the  $\text{KBr-2SrBr}_2$  melt purified with  $\text{NH}_4\text{Br}$  is equal to 0.610 V the  $pO$  value obtained using dependence (5) is equal to 3,19 and  $m_{O^{2-}}' = 6.5 \cdot 10^{-4} \text{ mol kg}^{-1}$ . For point 1'  $m_{O^{2-}}^0 = 5.2 \cdot 10^{-4} \text{ mol kg}^{-1}$  and for the first point of plot 1  $m_{O^{2-}}^0 = 2.2 \cdot 10^{-3} \text{ mol kg}^{-1}$  that principally agrees with the said above.

Now let us consider the process of  $\text{Eu}_2\text{O}_3$  dissolution in the studied melt. The dissolution process can be described by the following sequence of equations:



Processes similar to Eq. 6 are proper for all oxides dissolved in molten salts, however, usually they cannot be detected due to slight solubility and difficulties with the detection of non-dissociated oxide.

Rare-earth oxides are quantitatively or partially transformed into oxy-halides in halide melts [11]. The data of [11] were obtained for  $\text{KCl-LiCl}$  eutectic at 723 K, therefore, the transformation of  $\text{Eu}_2\text{O}_3$  into  $\text{EuOBr}$  in  $\text{KBr-2SrBr}_2$  seems very probable since the oxoacidic properties of both melts are close and the temperature is 250 K higher. The data of plot 2 (Fig. 1) give us the possibility to calculate the degree of this transformation. Indeed, the addition of  $3.02 \cdot 10^{-4} \text{ mol kg}^{-1}$  of  $\text{Eu}_2\text{O}_3$  to the studied melt results in the increase of  $\text{O}^{2-}$  molality by  $7.08 \cdot 10^{-4} \text{ mol kg}^{-1}$  (the second from the right point in plot 2, Fig. 1). The latter value means that the transformation of  $\text{Eu}_2\text{O}_3$

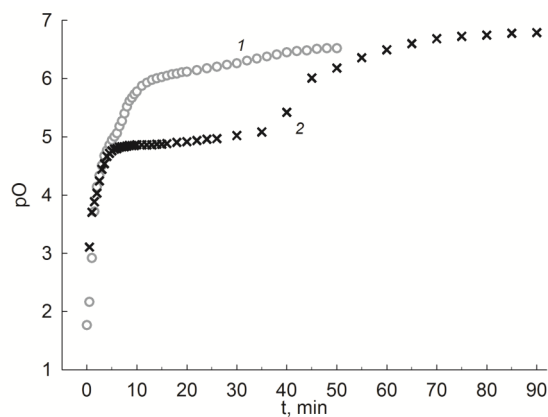
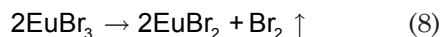


Fig. 2. The dependences of  $pO$  vs. time for processes of carbobromination of KBr-2SrBr<sub>2</sub> melt containing  $ca. 10^{-2} \text{ mol kg}^{-1}$  of  $O^{2-}$  (1) and carbobromination of KBr-2SrBr<sub>2</sub> melt with addition of  $5 \cdot 10^{-3} \text{ mol kg}^{-1}$  of  $Eu_2O_3$  (2) at 973 K.

into  $EuOBr$  is complete and  $6.04 \cdot 10^{-4} \text{ mol kg}^{-1}$  of the latter is formed. This explains the formation of  $3.02 \cdot 10^{-4} \text{ mol kg}^{-1}$  oxide ions in the melt. Remaining  $4.06 \cdot 10^{-4} \text{ mol kg}^{-1}$  should be ascribed to process (8), i.e. from  $6.04 \cdot 10^{-4} \text{ mol kg}^{-1}$  of  $EuOBr$   $4.06 \cdot 10^{-4} \text{ mol kg}^{-1}$  are dissolved with dissociation and  $ca. 2 \cdot 10^{-4} \text{ mol kg}^{-1}$  of  $EuOBr$  remains undissolved. The dissociation process is accompanied with  $Eu^{3+}$  reduction to  $Eu^{2+}$  since the existence of unbound  $Eu^{3+}$  ion is impossible under the experimental conditions, according to [12] the process:



occurs at 300 °C (573 K).

This means that the solubility product of  $EuO$  can be estimated as  $2.8 \cdot 10^{-7}$  (molality scale). The said fact is confirmed by the stability of  $emf$  at the following  $Eu_2O_3$  additions which do not results in the  $emf$  changes since the achieved molality of  $O^{2-}$  in the melt completely suppresses process (8). Plot 2' in Fig.2 corresponds to the case when amount of  $Eu_2O_3$  in the melt increases considerably so that the equilibrium of reaction (7) shifts to the right according to Le Chatelier-Shreder rule. However, this process occurs too slow and the observed changes of  $emf$  corresponds to time interval of 2 h.

### 3.2. The carbobromination of $Eu_2O_3$ in KBr-2SrBr<sub>2</sub> melt

The results of the carbobromination of the reference KBr-2SrBr<sub>2</sub> melt and of the melt containing suspension of  $5 \cdot 10^{-3} \text{ mol kg}^{-1}$   $Eu_2O_3$  at 973 K are presented in Fig. 2.

It is seen that at the beginning of carbobromination (0–3 min range) the dependences 1 and 2 (Fig.2) are practically coincident. This means that in this section the rate of  $O^{2-}$  sup-

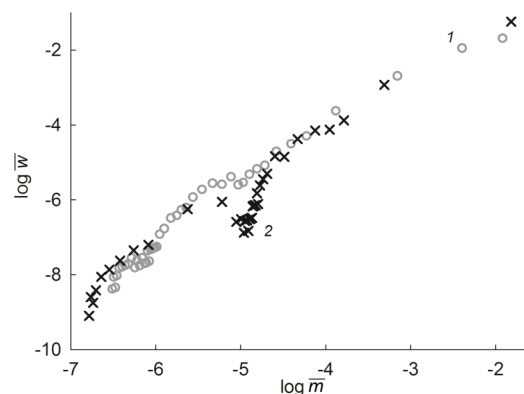


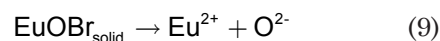
Fig. 3. The dependences of reaction rate ( $-\log \bar{w}$ ) vs.  $O^{2-}$  molality ( $-\log \bar{m}$ ) for the carbobromination process of KBr-2SrBr<sub>2</sub> melt containing  $ca. 10^{-2} \text{ mol kg}^{-1}$  of  $O^{2-}$  (1) and carbobromination of KBr-2SrBr<sub>2</sub> melt with addition of  $5 \cdot 10^{-3} \text{ mol kg}^{-1}$  of  $Eu_2O_3$  (2) at 973 K.

ply into solution from the suspension is not the limiting process. After 3 min a plateau-like section arises within the curve 2 (Fig.2) since the process of  $Eu_2O_3$  (or  $EuOBr$ ) dissolution according to reactions (7) and (8) become rate-defining. This means that there are two solid participants in this process (a solid particle to supply  $O^{2-}$  into the melt and solid C particle to remove it into the atmosphere) that makes its rate especially slow.

The said is confirmed by the data from the Van't Hoff diagram (Fig. 3) where the dependences of the rate of the carbobromination process vs. oxide ion concentration are presented. In dependence 2 at  $-\log \bar{m} \sim -5$  we observe a 'peak' arising due to a sharp decrease of the carbobromination process rate. Several points are not presented since for them neighboring  $emf$  values (and, hence  $m_{O^{2-}}^0$  values) are the same and, hence, for these points  $\log \bar{w} = -\infty$ .

From Fig. 2 it also can be seen that the running of the process of solid  $Eu$  oxocompound dissolution slows down the total carbobromination process by  $ca. 40$  min. Due to this reason the carbobromination of KBr-2SrBr<sub>2</sub> melt with  $Eu_2O_3$  addition is finished after 90 min whereas the melt containing only own oxide ions is purified within 50 min.

Fig. 2 also gives the possibility to estimate the solubility products of  $EuO$ :



Indeed, at the end of the plateau section  $pO$  value is 4.8 ( $m_{O^{2-}}^0 = 1.6 \cdot 10^{-5}$ ) and  $Eu^{3+}$  is completely reduced to  $Eu^{2+}$  ( $10^{-2} \text{ mol kg}^{-1}$ ). This gives  $K_{s, EuO} = 1.6 \cdot 10^{-7} \text{ mol}^2 \cdot \text{kg}^{-2}$  and  $pK_{s, EuO} = 6.8$ , whereas  $pK_{s, EuO}$  estimated on the basis of SAM method data (see description to Fig. 1) is equal to 6.56. Taking into account that usual accuracy

of  $pO$  determination is *ca.* 0.2 the data should be considered as being in a good agreement.

Performing the carbobromination of KBr-2SrBr<sub>2</sub> melt with addition of Eu<sub>2</sub>O<sub>3</sub> in the transparent quartz ampule gave us the possibility to obtain visually homogeneous solution containing Eu<sup>2+</sup> (as EuBr<sub>2</sub>). Such routine can be recommended for the direct doping of bromide melts used for the growth of Eu<sup>2+</sup>-activated bromide scintillators.

#### 4. Conclusions

The processes of Eu<sub>2</sub>O<sub>3</sub> solubilization in KBr-2SrBr<sub>2</sub> melt and the carbobromination of KBr-2SrBr<sub>2</sub> melt containing suspension of Eu oxocompounds were studied.

The solubility product of EuO was estimated as  $1.6 \cdot 10^{-7} \text{ mol}^2 \cdot \text{kg}^{-2}$  ( $pK_{s, \text{EuO}}=6.8$ ) using the data on Eu<sub>2</sub>O<sub>3</sub> solubilization and the transformation of small additions of Eu<sub>2</sub>O<sub>3</sub> in EuOBr is complete.

The features of the carbobromination process in KBr-2SrBr<sub>2</sub> melt with addition of Eu<sub>2</sub>O<sub>3</sub> are found, the main of them is the existence of a plateau-like section in 'pO-time' dependences due to participation in the reaction of two solid substances: carbon and one of the Eu oxo-compounds. The carbobromination data gave the possibility to estimate the solubility product of EuO as  $1.6 \cdot 10^{-7} \text{ mol}^2 \cdot \text{kg}^{-2}$  ( $pK_{s, \text{EuO}}=6.56$ ) that is in a good agreement with the data obtained at the Eu<sub>2</sub>O<sub>3</sub> solubilization (experimental accuracy of  $pO$  determination is  $\pm 0.2$ ).

The dissolution of Eu<sub>2</sub>O<sub>3</sub> ( $5 \cdot 10^{-3} \text{ mol kg}^{-1}$ ) in 50 g of the studied melt is finished in 40 min and the total deoxidization process is finished in 90 min whereas in the reference experiment (without Eu<sub>2</sub>O<sub>3</sub>) the duration of the deoxidization is 50 min.

The studied carbobromination routine can be recommended for direct dissolution of Eu<sub>2</sub>O<sub>3</sub> in bromide melts with the formation of soluble Eu<sup>2+</sup> species.

#### References

1. Y. Castrillejo, M.R. Bermejo, R. Pardo, A.M. Martinez, *J. Electroanal. Chem.*, **522**, 124 (2002).
2. M.S. Lukashova, K.N. Belikov, K.Yu. Bryleva, S.G. Kharchenko, S.G. Vishnevsky, V.I. Kalchenko. *Funct. Mater.*, **23**, 111 (2016).
3. Y. Castrillejo, M.R. Bermejo, E. Barrado, A.M. Martinez, P. Diaz Arocas., *J. Electroanal. Chem.*, **545**, 141 (2003).
4. K. Sridharan. Thermal Properties of LiCl-KCl Molten Salt for Nuclear Waste Separation, Project No. 09-780, University of Wisconsin, Madison, NEUP Final Report, 115 p. (2012). (<https://www.osti.gov/servlets/purl/1058922>).
5. A.L. Rebrov, Ya.A. Boyarintseva, V.L. Cherginets, T.E. Gorbacheva, A.Yu. Grippa, T.P. Rebrova, T.V. Ponomarenko, O.I. Yurchenko, N.V. Rebrova, V.A. Tarasov, P.N. Zhmurin. *Funct. Mater.*, **28**, 633 (2021). (<https://doi.org/10.15407/fm28.04.1>).
6. K. W. Krämer, P. Dorenbos, H. U. Güdel, C. W. E. van Eijk., *J. Mater. Chem.*, **16**, 2773 (2006). (<https://doi.org/10.1039/B602762H>).
7. N.J. Cherepy, G. Hull, A.D. Drobshoff, S.A. Payne, E. van Loef, C.M. Wilson, K.S. Shah, U.N. Roy, A. Burger, L.A. Boatner, W.-S. Choong, W.W. Moses., *Appl. Phys. Lett.*, **92** Art No 083508 (2008). (<https://doi.org/10.1063/1.2885728>).
8. E.D. Bourret-Courchesne, G. Bizarri, S.M. Hanrahan et al, *Nucl. Instrum. Methods Phys. Res. A*, **613**, 95 (2010). (<https://doi.org/10.1016/j.nima.2009.11.036>).
9. L. Stand, M. Zhuravleva, H. Wei, C.L. Melcher, *Opt. Mater.*, **46**, 59 (2015). (<https://doi.org/10.1016/j.optmat.2015.04.002>).
10. A.L. Rebrov, V.L. Cherginets, T.P. Rebrova, T.V. Ponomarenko, A.G. Varich, O.I. Yurchenko, V.V. Soloviev, *Funct. Mater.*, **30**, ... (2023).
11. H. Hayashi, K. Minato, *J. Phys. Chem. Solids*, **66**, 422 (2005). (<https://doi.org/10.1016/j.jpcs.2004.06.054>).
12. M.C. Petri, A.E. Klickman, M. Hori, *Appl. Nucl. Power*, Oarai, Japan, 2007. <https://www-pub.iaea.org/MTCD/Meetings/PDFplus/2007/cn152/cn152p/Petri%20LWR%20H2%20IAEA%20conf%20040407.pdf>.