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Synthesis and Photoluminescence characterization of Sr₃La(AlO)₃(BO₃)₄:Eu³⁺, Sm³⁺ Phosphor for n-UV w-LED

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A red emitting Eu³⁺ doped Sr₃La(AlO)₃(BO₃)₄ and Sm³⁺ doped Sr₃La(AlO)₃(BO₃)₄ phosphors with high efficiency have been synthesized by combustion method and the photoluminescence properties of samples are investigated in detail. Morphology by scanning electron microscopy and chromaticity by CIE were studied. The results demonstrate the excitation ranges from 340 nm to 420 nm, especially, the strongest excitation (394 nm and 406 nm) locates in the UV region for Eu³⁺ and Sm³⁺ doping, suggesting that the phosphor can match well with LED chips. Under n-UV light excitation of 394 nm the Eu³⁺ doped Sr₃La(AlO)₃(BO₃)₄ and 406 nm Sm³⁺ doped Sr₃La(AlO)₃(BO₃)₄ phosphor can emit bright red light with main emission peaks located at 617 nm and 602 nm are observed. The PL properties indicate that the phosphor has excellent stability. These results imply the importance of Sr₃La(AlO)₃(BO₃)₄:Eu³⁺ and Sr₃La(AlO)₃(BO₃)₄:Sm³⁺ red phosphors in white LEDs under n-UV excitation.

Keywords: Rare earth doped, Combustion synthesis, w-LED, CIE-Coordination, Sr₃La(AlO)₃(BO₃)₄:Eu³⁺, Sr₃La(AlO)₃(BO₃)₄:Sm³⁺, red phosphor.

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Introduction

Due to advantages of great brightness, low power consumption, and extended operating life, white light-emitting diodes (w-LEDs) have been regarded as significant solid-state light sources [1, 2]. Commercial w-LED currently employ a blue InGaN LED chip to excite a yellow emitting YAG:Ce phosphor, however this method renders colour poorly in the red region, resulting in a very low colour rendering index (CRI) [3]. Additionally, compared to green and blue phosphors, the luminous efficacy of red phosphor triggered by n-UV is significantly lower. As a result, the creation of novel, highly efficient red phosphors triggered by n-UV chips has received a lot of attention in the process of making w-LEDs [4]. Due to ⁵D₀→⁷F_J (J=0, 1, 2, 3, 4) transitions, the Eu³⁺ ion has been identified as one of the best red activators in the phosphors. Examples of this include Eu³⁺ doped Ca₁₂Al₁₄O₃₂F₂ [5], LaBMoO₆ [6], Sr₂LiScB₄O₁₀ [7], Bi₄O₃(BO₃)(PO₄) [8], LiYGeO₄ [9], Sr₅(PO₄)₃F [10],

ZnAl₁₂O₁₉ [11] and NaGdMgWO₆ [12]. They demonstrate effective red emission when excited by n-UV light.

The desire to use Sm³⁺ ions efficient emission in a variety of materials, particularly for phosphors in red and orange, but with the introduction of InGaN and GaN laser diodes in the excitation peak at 405 nm, where Sm³⁺ ions exhibits a quite substantial absorption, visible laser output has also grown. Numerous studies of Sm³⁺ ions in various materials, including phosphors [13, 14], single crystals [15,16], and glasses [17,18]. The Sm³⁺ in phosphors always exhibits a bright reddish emission, which can improve the color-rendering index (CRI) of phosphor-converted w-LEDs [19]. All lanthanide aluminates hosts are promising since they are used to produce white light by increasing the intensity of a red component [20-23].

A Tb³⁺ doped Sr₃La(AlO)₃(BO₃)₄ phosphor that emits a yellowish-green colour and has exceptional thermal stability and very high quantum efficiency, It has a great potential for combining red and blue Sr₃La(AlO)₃(BO₃)₄:Eu³⁺ to obtain an excellent for n-UV w-LED [24]. In this paper, Sr₃La(AlO)₃(BO₃)₄:Eu³⁺ and

$\text{Sr}_3\text{La}(\text{AlO})_3(\text{BO}_3)_4:\text{Sm}^{3+}$ phosphors were prepared by traditional combustion method and their photoluminescence, morphological and CIE properties were studied. The best proportion of the two rare earth ions was investigated also.

I. Experimental

The combustion process was used to create the $\text{Sr}_3\text{La}(\text{AlO})_3(\text{BO}_3)_4$ phosphors doped with Eu^{3+} and Sm^{3+} ions. Every initial component used in the experiment is of analytical (AR) grade. $\text{Sr}(\text{NO}_3)_2$ (Extra pure 98%), $\text{La}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ (99% AR), $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ (98.5% AR/ACS), $\text{H}_3(\text{BO}_3)$ (Extra pure 98%), Eu_2O_3 (99.9% AR), Sm_2O_3 (99.9% AR) and NH_2CONH_2 (99.5% AR) were employed as starting materials and fuel, respectively. By combining the proper volume of diluted nitric acid, Eu_2O_3 and Sm_2O_3 are transformed into nitrate form. Every compound in the mixture was combined using the stoichiometric ratio. The mixed colourless solution was then heated at a higher temperature while being stirred (100°C), producing an extremely viscous wet gel. The gel that results is put into a furnace that is kept at 550°C and reheated at 600°C for 2 hr and cooled up to room temp., It takes the flame around 40 seconds to die out. The final product obtained is then crushed with a mortar pestle and the fine powder sample is used for further investigations. SEM Images of the prepared sample is taken and Shimadzu RF5301PC Spectrofluorophotometer was used to measure the PL. A spectral slit width of 1.5 nm was used to record the excitation and emission.

II. Results and Discussion

2.1. SEM studies

SEM investigation of the host $\text{Sr}_3\text{La}(\text{AlO})_3(\text{BO}_3)_4$ phosphor was performed to study the morphology and particle size, as shown in Fig.1. The phosphor is prepared using a combustion method approach and has a solid microcrystalline structure with certain uneven shapes and aggregation between crystalline grains. The average particle size of the $\text{Sr}_3\text{La}(\text{AlO})_3(\text{BO}_3)_4$ sample is determined to be between 2 and 15 micrometers. The majority of commercial phosphors now on the market have particle sizes in the range of a few micrometers [25,26].

2.2. Photoluminescence studies

2.2.1. Photoluminescence properties of $\text{Sr}_3\text{La}(\text{AlO})_3(\text{BO}_3)_4:\text{Eu}^{3+}$ phosphor

Figure 2 displays the excitation spectra of $\text{Sr}_3\text{La}(\text{AlO})_3(\text{BO}_3)_4:\text{Eu}^{3+}$ phosphor were studied in the wavelength range of 350 and 400 nm after the emission peak at 617 nm. The excitation peaks at 362, 380, and 394 nm, corresponding to Eu^{3+} ions transitions ${}^7\text{F}_0 \rightarrow {}^5\text{L}_9$, ${}^7\text{F}_0 \rightarrow {}^5\text{G}_3$, and ${}^7\text{F}_0 \rightarrow {}^5\text{L}_6$ [27]. The maximum excitation peak in the $\text{Sr}_3\text{La}(\text{AlO})_3(\text{BO}_3)_4:\text{Eu}^{3+}$ phosphor was determined to be 394 nm.

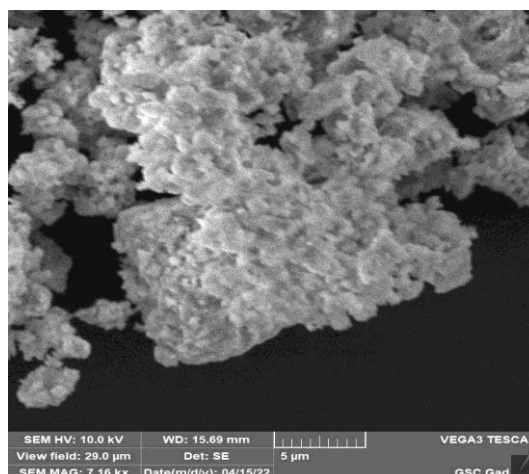


Fig. 1. SEM Photograph of host $\text{Sr}_3\text{La}(\text{AlO})_3(\text{BO}_3)_4$.

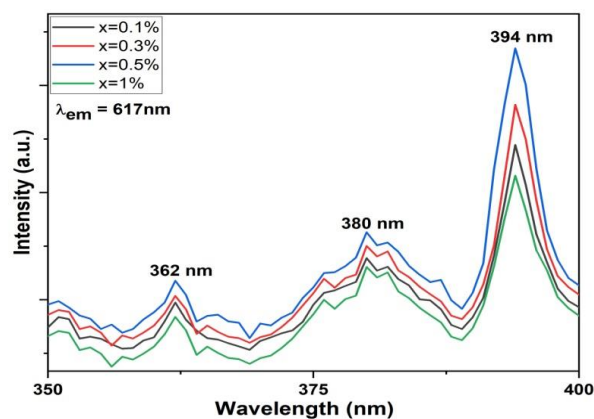


Fig. 2. The excitation spectrum of $\text{Sr}_3\text{La}(\text{AlO})_3(\text{BO}_3)_4:\text{Eu}^{3+}$ (monitored at 617 nm).

Figure 3 displays the emission spectra of $\text{Sr}_3\text{La}(\text{AlO})_3(\text{BO}_3)_4$ phosphors doped Eu^{3+} ions at an excitation wavelength of 394 nm. Two well-resolved emission bands 590 nm (${}^5\text{D}_0 \rightarrow {}^7\text{F}_1$) and 617 nm (${}^5\text{D}_0 \rightarrow {}^7\text{F}_2$) might potentially be seen based on the luminescence spectra, in line with the well-documented Eu^{3+} ions doped phosphor literature of published data. It is widely accepted that the electron dipole and magnetic dipole transitions are responsible for the emission peaks at 617 nm and 590 nm, respectively [28].

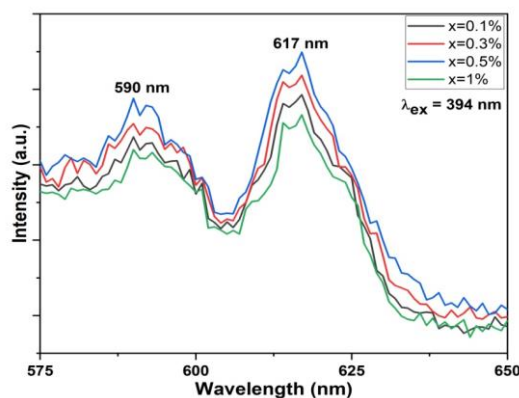


Fig. 3. The emission spectra of $\text{Sr}_3\text{La}(\text{AlO})_3(\text{BO}_3)_4:\text{Eu}^{3+}$ phosphors under 394 nm excitation.

2.2.2. Photoluminescence properties of $\text{Sr}_3\text{La}(\text{AlO})_3(\text{BO}_3)_4:\text{Sm}^{3+}$ phosphor

The photoluminescence excitation spectra of $\text{Sr}_3\text{La}(\text{AlO})_3(\text{BO}_3)_4:\text{Sm}^{3+}$ phosphor are shown in Fig. 4. These materials' PLE spectra were obtained at room temperature by measuring emission wavelength at 602 nm. These spectra exhibit four narrow and strong excitation peaks at 347, 364, 377 and 406 nm ascribed to transitions from ground state $^6\text{H}_{5/2}$ to excited states $^4\text{H}_{9/2}$, $^6\text{D}_{3/2}$, $^4\text{D}_{1/2}$ and $^4\text{F}_{7/2}$ arising from f-f transitions of Sm^{3+} ions [29-33]. The highest excitation peak formed at 406 nm as a result of the $^6\text{H}_{5/2} \rightarrow ^4\text{F}_{7/2}$ transition.

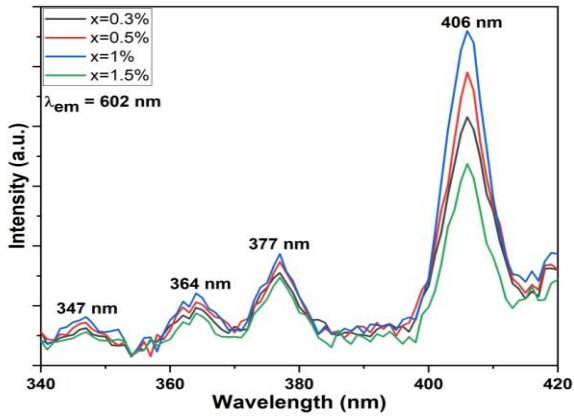


Fig. 4. The excitation spectrum of $\text{Sr}_3\text{La}(\text{AlO})_3(\text{BO}_3)_4:\text{Sm}^{3+}$ (monitored at 602 nm).

The PL spectra of $\text{Sr}_3\text{La}(\text{AlO})_3(\text{BO}_3)_4:\text{Sm}^{3+}$ phosphor stimulated at 406 nm n-UV light are shown in Fig. 5. There are two primary emission peaks in the 525-625 nm range, which are located at 566 and 602 nm, as a result of intra 4f-4f transitions from excited states ($^4\text{G}_{5/2}$) to ground states ($^6\text{H}_{5/2}$ and $^6\text{H}_{7/2}$) respectively. The magnetic dipole (MD) permitted transitions ($^4\text{G}_{5/2} \rightarrow ^6\text{H}_{5/2}$ and $^4\text{G}_{5/2} \rightarrow ^6\text{H}_{7/2}$) detected at 566 and 602 nm dominate the emission spectra and are responsible for the orange-red emission of activator ions, according to the selection rule (change in $j = 0, \pm 1$). [34-36].

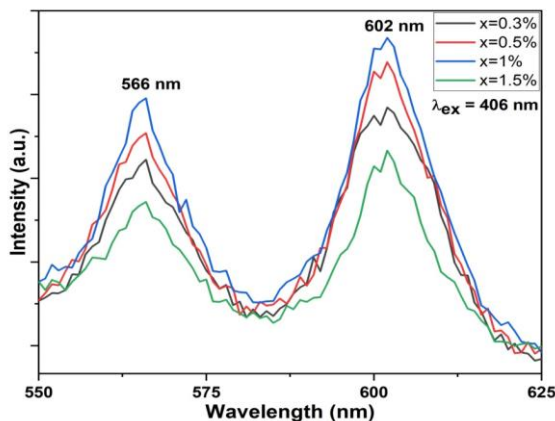


Fig. 5 The emission spectra of $\text{Sr}_3\text{La}(\text{AlO})_3(\text{BO}_3)_4:\text{Sm}^{3+}$ phosphors under 406 nm excitation.

Concentration Quenching

It should be noted that the position of the emission peaks and the form of the emission spectra are similar in each doping concentration; nevertheless, the intensity

factor varied gradually with varying Eu^{3+} and Sm^{3+} ions concentrations in each phosphor. When compared to other concentrations of synthesized phosphors, $\text{Sr}_3\text{La}(\text{AlO})_3(\text{BO}_3)_4$ phosphor (Fig. 6 & 7) has a maximum emission intensity of 1.0 mole % for Eu^{3+} ions and 1.5 mole % for Sm^{3+} doping. The drop in emission intensities was correlated with a rise in Eu^{3+} and Sm^{3+} ions concentrations as a result of the concentration quenching process. As the overall average between the Eu^{3+} and Sm^{3+} ions reduces with increasing ion concentrations in $\text{Sr}_3\text{La}(\text{AlO})_3(\text{BO}_3)_4:\text{Eu}^{3+}$ and $\text{Sr}_3\text{La}(\text{AlO})_3(\text{BO}_3)_4:\text{Sm}^{3+}$ phosphor respectively, a non-radiative energy transfer mechanism occurs via cross-relaxation, enhancing concentration quenching (Fig. 6 & 7) [37, 38, 39].

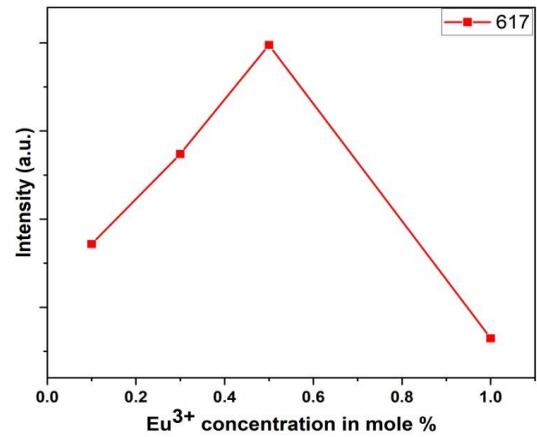


Fig. 6. Variation in the emission intensity 617 nm as function of the Eu^{3+} ion concentration in $\text{Sr}_3\text{La}(\text{AlO})_3(\text{BO}_3)_4:\text{Eu}^{3+}$ phosphor.

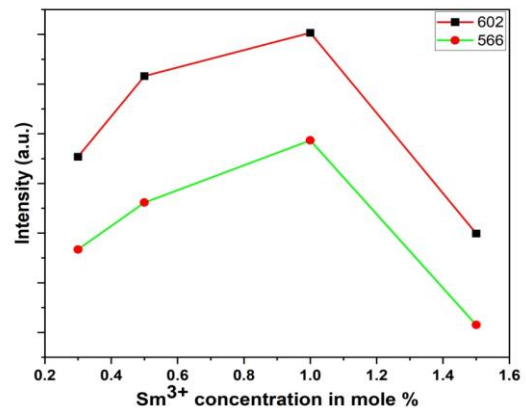


Fig. 7. Variation in the emission intensity 566 nm and 602 nm as function of the Sm^{3+} ion concentration in $\text{Sr}_3\text{La}(\text{AlO})_3(\text{BO}_3)_4:\text{Sm}^{3+}$ phosphor.

2.3. Chromaticity Analysis

Colour coordinates are the essential parameters for examining the phosphor's CIE chromaticity performance. The CIE chromaticity coordinates of $\text{Sr}_3\text{La}(\text{AlO})_3(\text{BO}_3)_4:\text{Eu}^{3+}$ and $\text{Sr}_3\text{La}(\text{AlO})_3(\text{BO}_3)_4:\text{Sm}^{3+}$ Phosphors excited at 394 nm and 406 nm respectively are presented in Fig.7. The corresponding x and y coordinates are (0.684, 0.314) for 617 nm and (0.635, 0.363) for 602 nm. To our observation for all the studied $\text{Sr}_3\text{La}(\text{AlO})_3(\text{BO}_3)_4:\text{Eu}^{3+}$ and $\text{Sr}_3\text{La}(\text{AlO})_3(\text{BO}_3)_4:\text{Sm}^{3+}$

phosphors emission points are found to be located in the red region. Hence, the present red-emitting phosphors find remarkable applications in near ultraviolet excited for w-LEDs [40, 41].

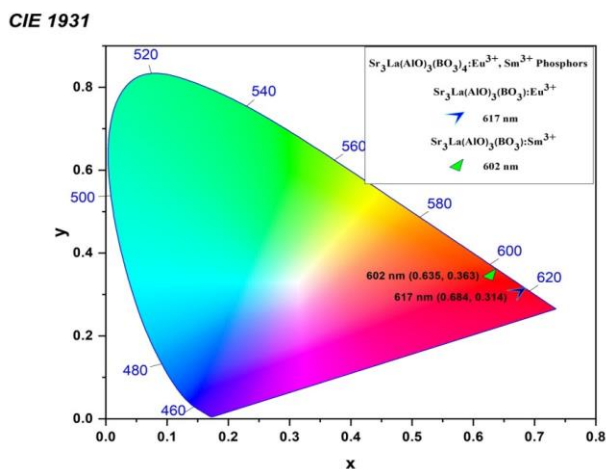


Fig.7. CIE chromaticity diagram of the $\text{Sr}_3\text{La}(\text{AlO})_3(\text{BO}_3)_4:\text{Eu}^{3+}$ and $\text{Sr}_3\text{La}(\text{AlO})_3(\text{BO}_3)_4:\text{Sm}^{3+}$ phosphors.

Conclusion

A Series of $\text{Sr}_3\text{La}(\text{AlO})_3(\text{BO}_3)_4:\text{xEu}^{3+}$ ($x = 0.2, 0.5, 1, 1.5$ mole %) phosphor and $\text{Sr}_3\text{La}(\text{AlO})_3(\text{BO}_3)_4:\text{xSm}^{3+}$ ($x = 0.5, 1.0, 1.5, 2.0$ mole %) phosphor powders were successfully synthesized by the combustion method. The morphological and photoluminescence properties were investigated. The SEM image identifies the irregular morphology of the $\text{Sr}_3\text{La}(\text{AlO})_3(\text{BO}_3)_4$ phosphor samples with micron ranged particles. The photoluminescence properties investigated under photo-excitation showed that the Eu^{3+} doped $\text{Sr}_3\text{La}(\text{AlO})_3(\text{BO}_3)_4$ phosphors exhibited a main red emission peak at 617 nm corresponding to ${}^5\text{D}_0 \rightarrow {}^7\text{F}_2$ transition, also the Sm^{3+} doped

$\text{Sr}_3\text{La}(\text{AlO})_3(\text{BO}_3)_4$ phosphors exhibited main red emission peak at 602 nm corresponding to ${}^4\text{G}_{5/2} \rightarrow {}^6\text{H}_{7/2}$ transition. The optimized concentration of Eu^{3+} and Sm^{3+} ions in the present host is 1% and 1.5% mole respectively. Beyond that concentration, the quenching effect dominates. The CIE of present phosphor doped with Eu^{3+} and Sm^{3+} ions was calculated to be (0.684, 0.314) and (0.635, 0.363) respectively. All results indicate that the as-prepared $\text{Sr}_3\text{La}(\text{AlO})_3(\text{BO}_3)_4:\text{Eu}^{3+}$ and $\text{Sr}_3\text{La}(\text{AlO})_3(\text{BO}_3)_4:\text{Sm}^{3+}$ phosphor have excellent red luminescence properties and is suitable as red-emitting phosphor for w-LEDs application.

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Declaration of Competing Interest

The authors declare that they do not have any known competing financial interests or personal ties that may seem to have influenced the work reported in this paper.

Data availability

No data was used for the research described in the article.

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Синтез і фотолюмінесцентні характеристики люмінофору $\text{Sr}_3\text{La}(\text{AlO})_3(\text{BO}_3)_4:\text{Eu}^{3+}, \text{Sm}^{3+}$ для n-УФ w-LED

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Методом спалювання синтезовано високоефективні люмінофори $\text{Sr}_3\text{La}(\text{AlO})_3(\text{BO}_3)_4$, леговані Eu^{3+} і Sm^{3+} , збуджені червоним світлом, а також детально досліджено фотолюмінесцентні властивості зразків. Вивчено морфологію за допомогою скануючої електронної мікроскопії та хроматичність за допомогою СІЕ. Результати демонструють діапазони збудження від 340 нм до 420 нм, особливо, найсильніше збудження (394 нм і 406 нм) знаходиться в УФ-області при допуванні Eu^{3+} і Sm^{3+} , що свідчить про те, що люмінофор може добре поєднуватися зі світлодіодними чіпами. Під дією n-УФ-світла 394 нм люмінофор $\text{Sr}_3\text{La}(\text{AlO})_3(\text{BO}_3)_4$, легований Eu^{3+} , і 406 нм Sm^{3+} , легований $\text{Sr}_3\text{La}(\text{AlO})_3(\text{BO}_3)_4$, може випромінювати яскраво-червоне світло з основними піками випромінювання, розташованими на 617 нм і 602 нм. Властивості ФЛ вказують на чудову стабільність люмінофора. Ці результати вказують на важливість червоних люмінофорів $\text{Sr}_3\text{La}(\text{AlO})_3(\text{BO}_3)_4: \text{Eu}^{3+}$ і $\text{Sr}_3\text{La}(\text{AlO})_3(\text{BO}_3)_4: \text{Sm}^{3+}$ у білих світлодіодах при n-УФ-збудженні.

Ключові слова: легування рідкісноземельними елементами, горіння, w-LED, СІЕ-координація, $\text{Sr}_3\text{La}(\text{AlO})_3(\text{BO}_3)_4:\text{Eu}^{3+}$, $\text{Sr}_3\text{La}(\text{AlO})_3(\text{BO}_3)_4:\text{Sm}^{3+}$, червоний люмінофор.