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Characterization of YAG:Ce ceramics with graphene oxide

Yttrium-aluminum garnet ($Y_3Al_5O_{12}$) is an optical material that shows great potential due to its favorable light-emitting and mechanical properties, as well as its chemical stability and thermal resistance. It is commonly utilized in laser technology, optical instruments, and solid-state light sources as a result of its activation by transition metal or rare earth element ions. In this work, experiments were carried out on the procedure for compacting samples of luminescent ceramics $Y_3Al_5O_{12}:Ce^{3+}$, by single-base pressing followed by sintering. Comprehensive studies of the influence of graphene oxide of variable concentration (x=0.1;0.5; 1 wt.%) on the radiative characteristics of ceramic samples. It was found that the addition of graphene oxide in an amount from 0.1 to 1 wt. % affects the density and luminescent properties of YAG:Ce ceramics. There is a decrease in the value of the density parameter at concentrations of graphene oxide 0.1-1 wt. % before annealing, after annealing, the relative density value increases to ~99 %. The luminescence spectrum when excited by a blue LED chip appears as a wide band in the spectral range 460-750, the nature of which is associated with radiative transitions in the cerium ion. It has been established that the light-emitting characteristics have a downward trend when activated by graphene oxide. The integral luminescence intensity decreases from 27.1 % to 19 % with an increase in the concentration of graphene oxide.

Keywords: YAG ceramics, sintering process, graphene oxide, mechanic properties.

Introduction

Graphene is a superconducting, two-dimensional material with unique properties which are promising for various technological applications in many fields of science and technology [1-4]. Due to the unique band structure [5] graphene oxide has excellent electrical and thermal conductivity [6] high surface area [7] and good optical properties [8]. Graphene-based materials have good nonlinear optical properties [9]. The mechanical characteristics of graphene are similar to those of carbon nanotubes [10]. But unlike carbon nanotubes, graphene oxide penetrates better into the structure of the matrix material and provides more effective compaction comparing to nanotubes [11]. These properties make graphene a promising material for applying in microelectronics, photonic and optoelectronic devices, and laser technique [12-15].

Yttrium-aluminum garnet ($Y_3Al_5O_{12}$) is a highly promising optical material that possesses excellent optical and mechanical properties, as well as chemical stability and thermal resistance. It is commonly used in laser technology, optical instrumentation, and solid-state light sources. These materials are typically activated by transition metal or rare earth ions [16, 17].

The use of yttrium-aluminum garnet activated by cerium ions (YAG:Ce) has experienced a notable rise in the production of white light-emitting diodes (WLEDs). These materials serve as optical radiation converters, allowing for the production of high-quality white light. Modern commercial WLEDs consist of a blue glow crystal and a Ce:YAG phosphor dispersed in an optically transparent coating consisting of organosilicon, polymer or epoxy resin. The main problem of such light-emitting diodes is the local overheating of the resin (compound), which leads to degradation of the coating and changing of optical characteristics [18, 19]. That is why it is possible to use luminescent ceramics based on YAG:Ce as an optical radiation converter, which is devoid of the above disadvantages. The addition of graphene oxide (GO) to such ceramics in relatively small concentrations can improve the optical and luminescent properties of ceramics.

In this work, the effect of graphene oxide on the density and luminescent characteristics of ceramics based on yttrium-aluminum garnet activated by cerium is investigated

Expiremental

Micro powder of yttrium-aluminum garnet activated by cerium (NIIPP, Russia) and commercial suspension of graphene oxide (Graphenea, Spain) were used as starting materials.

Mixing of the initial components was carried out by the wet method in isopropyl alcohol using a ball mill. The mixing duration was 48 hours. As a result, powder suspensions of the YAG:Ce-xGO (where x = 0.1; 0.5; 1 wt. %) composition were obtained. The resulting suspensions were dried by exposing them to air at a temperature of 120 °C until all the moisture evaporated completely.

To compact the powder, cold static pressing was performed on a test press IP500 AUTO from Zipo in Russia, applying a pressure of 400 MPa.

The sintering process was conducted using a high-temperature furnace (LHT 02/18, Nabertherm, Germany) in an air atmosphere. The temperature range for sintering was set between 1250 °C and 1650 °C. The samples were exposed to each sintering temperature for duration of 2 hours. The heating and cooling rate during the process was maintained at 200 °C per hour. The resulting samples had a thickness of approximately 2 mm and a diameter ranging from 8.5 to 9 mm. The selection of the sintering parameters was based on previous findings [20].

Following the sintering process, the samples underwent grinding and polishing using a Buehler AutoMet 300 Pro machine (Switzerland). Polycrystalline diamond suspensions from Kemix (Russia) were used for this purpose.

The density of the samples was determined by measuring their mass and linear dimensions, according to the formula:

$$\rho = m/V \quad (1)$$

X-ray phase analysis of the ceramics was performed using an XRD-7000 X-ray diffractometer (Shimadzu, Japan). The resulting diffractograms were analyzed using specialized software and the international crystallographic database “PDF-4”.

To study the optical properties of the ceramics, measurements were conducted in the ultraviolet, visible, and near-infrared spectral regions using a two-beam scanning spectrophotometer SF-256 UVI (200-1100) (Lomo-Photonics, Russia).

The integral spectral efficiency was measured using an integrating sphere and a calibrated AvaSpec-3648 spectrophotometer (200-1100 nm, inverse linear dispersion 1.2 nm/mm). A light-emitting diode with a wavelength of 447 nm was used as the excitation source. The excitation of the sample was carried out using a flow integrated by a sphere. The spectrum and radiation flux of the diode falling on the geometric location where the test sample was positioned were measured. Next, the radiation spectrum was measured with the sample under study. By employing this technique, it became possible to precisely subtract the diode spectrum from the luminescence spectra of the ceramics, while maintaining the original form of the spectral lines. As a result, it became feasible to ascertain the reflected flux, absorbed flux, and radiation flux of the ceramic sample. The overall spectral efficiency was then computed as the ratio of the integral flux emitted by the sample to the integral flux absorbed by the sample.

Results and discussion

Figure 1 shows the dependence of the relative density of powder compacts before and after sintering at a temperature of 1650 °C on the concentration of graphene oxide.

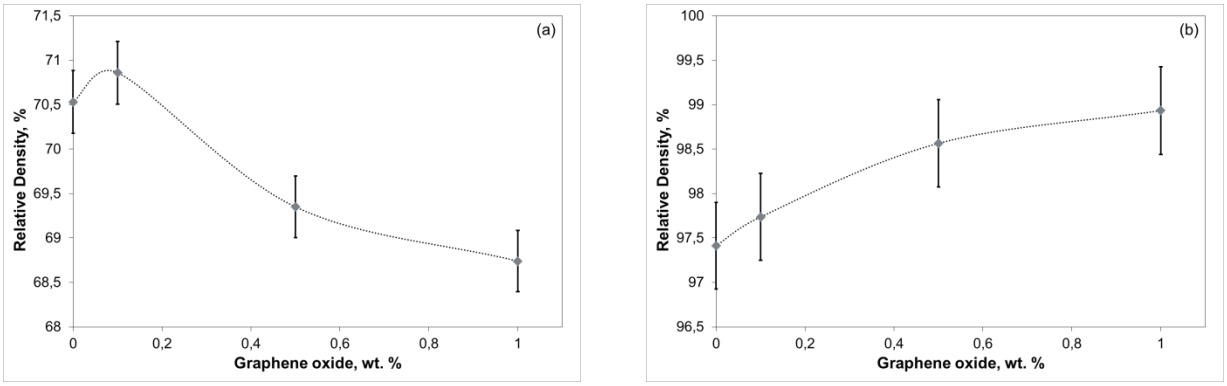

Figure1. Dependence of the relative density of samples on the concentration of graphene oxide (a) before sintering; (b) after sintering at a temperature of 1650 °C.

Relative density of the powder compact from the powder of "pure" yttrium-aluminum garnet activated by cerium is 70.5 ± 0.5 %. The addition of 0.1 wt. % graphene oxide to YAG:Ce leads to a slight increase in the relative density of the compact (up to 70.9 =0.5 %), however, the observed increase in density is within the confidence interval of measuring value. Further increase of graphene oxide concentration from 0.1 wt. % up to 1 wt. % leads to a decrease in the relative density of the compact from 70.9 ± 0.5 % to 68.7 ± 0.5 %.

For samples after sintering, unlike powder compressions, with an increase in graphene oxide concentration from 0 to 1 wt. % there is an increase in the relative density of samples from 97.4 ±0.5 to 98.9±0.5 %. The maximum value of the relative density is observed at a graphene oxide concentration of 1 wt. %.

X-ray phase analysis revealed that the YAG:Ce ceramics samples were composed of cubic YAG in a stoichiometric ratio. All observed peaks corresponded to the YAG phase (PDF-Card#010-83-7850). No additional phases were detected in the samples, and there was no broadening of the peaks or presence of any other peaks on the diffractograms. The absence of peaks characteristic of graphene oxide is due to their relatively low content (less than 5 wt. %), which does not exceed the sensitivity limit of the diffractometer.

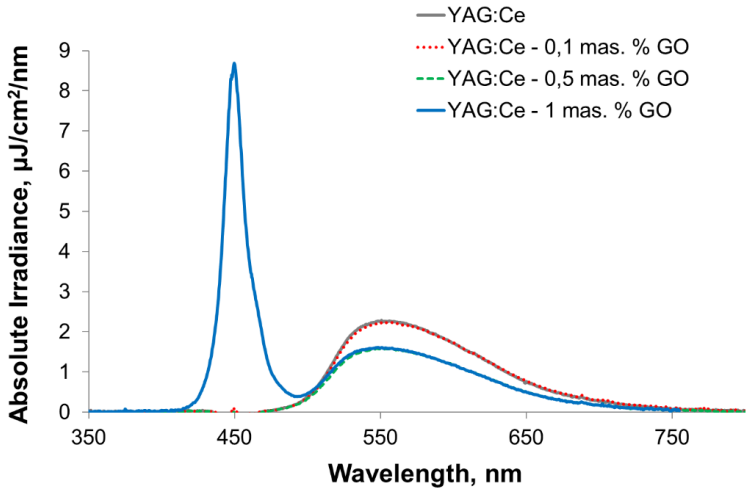

Figure2. Integral photoluminescence spectrum of ceramics based on YAG:Ce

The photoluminescence spectra of the samples excited by an LED with a wavelength of 447 nm are shown in Figure 2. The spectra are typically for the YAG:Ce system [21]. A maximum is observed in the region of 550 nm. Addition of graphene oxide in an amount from 0.5 to 1 wt. % leads to a shift of the maximum position to the region of 545 nm and a decrease in the intensity of radiation.

The results of measuring the efficiency of luminescence and diffuse reflection of the studied samples are shown in Figure 3. It can be seen that the efficiency of luminescence with an increase in graphene oxide concentration from 0 to 1 wt. % varies from 27.1 to 19 %. The lowest luminescence efficiency is observed at a graphene oxide concentration of 0.5 wt. %. Diffuse reflection of samples increases from 13 to 16.8 % with an increase in graphene oxide concentration from 0 to 1 wt. %.

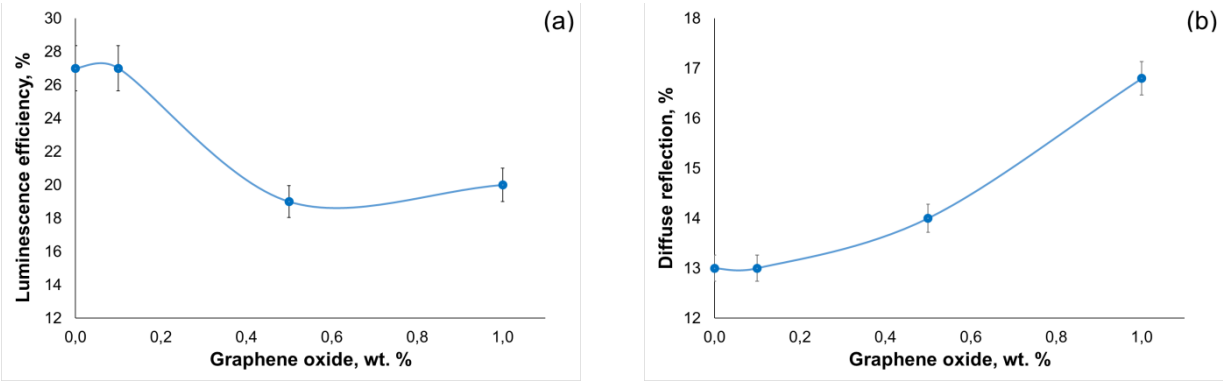

Figure3. Efficiency of luminescence (a) and diffuse reflection (b) of YAG:Ce ceramics with different graphene oxide concentration

Impact of graphene oxide on YAG:Ce ceramics remains ambiguous. Although the inclusion of graphene oxide does result in an increase in ceramic density under identical conditions, it also has a negative effect on the luminescent properties of the ceramics.

Conclusion

As a result of the work samples of luminescent ceramics based on yttrium-aluminum garnet activated with cerium with graphene oxide additives were manufactured. The influence of graphene oxide on the density and luminescent properties of ceramics was investigated.

It was found that the addition of graphene oxide in an amount from 0.1 to 1 wt. % leads to an increase in relative density from 94.7 ± 0.5 to 98.9± 0.5 %, an increase in diffuse reflection from 13 to 16.8 % and a decrease in luminescence efficiency from 27 to 19 %.

It is shown that the addition of up to 1 wt. % graphene oxide to YAG:Ce ceramics provides 1.5 % increase in relative density.

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YAG керамикасының сипаттамасы: графен оксиді бар Ce

Итрий алюминий гранатасы ($Y_3Al_5O_{12}$) жақсы жарық шығаратын және механикалық қасиеттері, химиялық тұрақтылығы және термиялық тұрақтылығы бар перспективалы оптикалық материал. Өтпелі металл немесе сирек жер иондары арқылы белсендірілген иттрий-алюминий гранат негізіндегі материалдар лазерлік технологияда, оптикалық аспаптарда және қатты күйдегі жарық көздерінде кеңінен қолданылады. Бұл жұмыста $Y_3Al_5O_{12}:Ce^{3+}$ люминесцентті керамика үлгілерін бір негізді престеу, содан кейін агломерациялау арқылы нығыздау процедурасы бойынша тәжірибелер жасалды. Керамикалық үлгілердің сәуле шығару сипаттамаларына ауыспалы концентрациядағы графен оксидінің (x=0,1; 0,5;1 мас. %) әсеріне кешенді зерттеулер жүргізілді. Графен оксидінің 0,1-ден 1 мас. % дейінгі мөлшердегі қоспасы YAG:Ce керамикасының тығыздық және люминесценция қасиеттеріне әсер ететіні анықталды. Жұмсарғанға дейін графен оксидінің 0,1-1 мас. % концентрация кезінде тығыздық параметрі мәнінің төмендейтіні, ал жұмсарғаннан кейін салыстырмалы тығыздық мәні ~99 % дейін жоғарылайтыны байқалды. Қозу кезіндегі люминесценция спектрі көк болады, ал жарықдиодты чип 460-750 спектрлік диапазонында кең жолақ түрінде көрінеді, оның табиғаты церий ионындағы сәуле шығаратын ауысумен байланысты. Жарық шығаратын сипаттамалардың графен оксидімен белсендірілген кезде төмендеу динамикасы бар екендігі анықталды. Графен оксидінің концентрациясы жоғарылаған кезде жарықтың интегралды қарқындылығы 27,1-ден 19%-ға дейін төмендейді.

Кілт сөздер: YAG-керамика, агломерация процесі, графен оксиді, механикалық қасиеттері.

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Характеристика керамики YAG:Ce с оксидом графена

Иттрий-алюминиевый гранат ($Y_3Al_5O_{12}$) является перспективным оптическим материалом с хорошими светоизлучающими и механическими свойствами, химической стабильностью и термической стойкостью. Материалы на основе иттрий-алюминиевого граната, активированного ионами переходных металлов или редкоземельных элементов, широко используются в лазерной технике, оптическом приборостроении и твердотельных источниках света. В настоящей работе были проведены эксперименты по процедуре компактирования образцов люминесцентной керамики $Y_3Al_5O_{12}:Ce^{3+}$, путем одноосновного прессования с последующим спеканием. Проведены комплексные исследования влияния оскида графена переменной концентрации (x=0,1; 0,5; 1 вес. %) на излучательные характеристики образцов керамики. Было установлено, что добавка оксида графена в количестве от 0,1 до 1 мас. % влияет на плотность и люминесцентные свойства YAG:Ce керамики. Наблюдается снижение значения параметра плотности при концентрациях оксида графена 0,1–1 вес. % до отжига, после отжига значение относительной плотности повышается до ~99 %. Спектр люминесценции при возбуждении синий, чипом светодиода проявляется в виде широкой полосы в спектральном диапазоне 460–750, природа которой связана с излучательными переходами в ионе церия. Установлено, что светоизлучающие характеристики имеют динамику к снижению при активации оксидом графена. Интегральная интенсивность свечения уменьшается с 27,1 до 19 % при повышении концентрации оксида графена.

Ключевые слова: YAG керамика, процесс спекания, оксид графена, механические свойства.