

# Study on the Electroluminescence of $Y_2O_3$ Material Doped with Rare Earth Elements

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## Review Article

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### ABSTRACT

There has been a recent advancement in the field of Electroluminescence. Many Electroluminescent technologies are emerging these days with a variety of EL materials and devices. The  $Y_2O_3$  (Yttrium Oxide material)- a topological luminophore is an insulating, air stable, non-toxic material which exhibits both particle size ranging from micrometer to nanometer. The  $Y_2O_3$  has a good transparency for infrared radiation and has high thermal stability. This material is synthesized by various techniques like sol-gel process etc. and crystal structure is investigated by various techniques like XRD and also morphology by SEM, TEM etc. The development of high-luminance film electroluminophors like  $Y_2O_3$  has increased the electroluminescent performance of various devices. These are used as host in these devices and shows outstanding emission characteristics. Electroluminescence of  $Y_2O_3$ : Eu and  $Y_2O_3$ : Sm films to create full-color electroluminescent indicators, is also a field of interest to researchers for its high efficiency and emission intensity. The  $Y_2O_3$  films are becoming the most stable luminophores and are helping to increase the electroluminescent intensity of various devices based on this material. This report consists of future research and advances in electroluminescence of  $Y_2O_3$  material. The inventions will lead to significant developments in various fields like health care, transportation, electronic communication and other area

## INTRODUCTION

In 1603, an alchemist and cobbler named Vincenzo Cascariolo lived in Bologna, Italy. He heated a mixture of  $BaSO_4$  (Barium Sulphate), a synthetic material ( $BaSO_4$ ) and coal. This was the first investigation about the luminescence. Today, Phosphorus is the name given to the chemical element only and certain microcrystalline luminescent materials were called as Phosphors. The phosphor investigated by Cascariolo was a Barium Sulphide ( $BaS$ ). The first Phosphor which was commercially available was "Balmain's Paint", a Calcium Sulphide ( $CaS$ ) preparation.

In 1672, an English scientist Robert Boyle investigated the phenomenon of luminescence in rotting wood or flesh and by glowworms known since ages. This investigation also aroused the thinking of the Scientists and led in the further development of the topic <sup>[1]</sup>.

A substance which emits light as a result of heating is called Incandescent. The bodies which emit light not resulting from heat is said to be Luminescent. Thus, it is a cold body radiation <sup>[2,3]</sup>. When a substance emits light on account of energy supplied to it from a source of suitable radiation is known as Luminescence <sup>[4]</sup>. The causes of luminescence are chemical reactions, electrical form of energy (electrical energy) or stress applied on a crystal. Materials exhibiting luminescence are broadly called as phosphors <sup>[2,3]</sup>. At atmospheric pressure and atmospheric temperatures, this phenomenon occurs in certain materials exposed to UV or Infrared radiation. Certain luminescent materials exhibit Ferromagnetic behavior (Ferromagnetic Insulators) and are multifunctional materials. A coating of luminescent materials is done on the dials, hands, scales, and signs of aviation and navigational instruments (through Luminescing process) etc. There are various ways of exciting the substances. On the basis of this there is different kind of luminescence <sup>[2]</sup>.

## ELECTROLUMINESCENCE

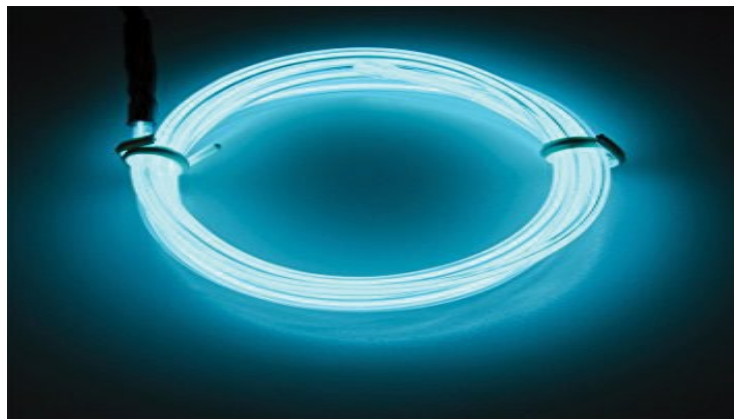
It is the glow of bodies when electric current is passed through substance or the action of electric field is called electroluminescence. For example, the glow of a gas discharge in the tubes of advertisement, EL Wire Belt Tape, EL Panels etc. (**Figure 1 and Figure 2**). Cathodoluminescence comes under this category. These materials emit light when electrons strike on them.

It is an optical and electrical phenomenon by nature in which light emission takes place by certain materials due to the action of a strong electric field or passage of an electric current through them is called electroluminescence <sup>[3]</sup>. The phenomenon can be considered as both electrical and optical in nature. It is distinct from Black Body Radiation, Incandense, Chemiluminescence, Sonoluminescence and Mechanoluminescenceetc <sup>[2]</sup>. It involves conversion of electrical energy into non-thermal emitted light. Electroluminescence is sometimes observed at the electrodes- cathode and anode during the process of electrolysis. Under the impact of accelerated electrons a great number of materials show luminescence. For example- diamond, ruby, crystal phosphors etc <sup>[4]</sup>. The mechanism behind electroluminescence is the radiative recombination, or spontaneous emission. Electroluminescence is the principle behind the construction of various LEDs, automobile displays, and night lamps. Semiconductors are the most commonly available electroluminescent materials. Electroluminescent technologies have low power consumption and are valuable to the advertising industry <sup>[2]</sup>. This phenomenon is also studied in multilayer diode structures which are based on SiC (Substituted Indolo Carbazoles) <sup>[5]</sup>. An Organic Light Emitting Diode (OLED) has self-luminous property and acts as a simple LED <sup>[6]</sup>. Electroluminescence phenomenon is also in rectification of graphene <sup>[7]</sup>. Nano-particle luminescence technology is also developing day by day <sup>[8]</sup>. Electroluminescence and its application will lead to significant developments in the field of health, transport services, electronic communication and other areas <sup>[2]</sup>.

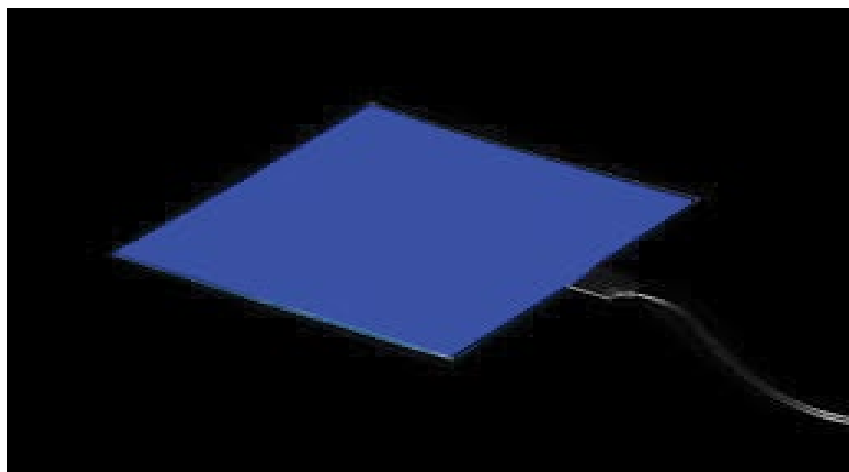
### YTTRIUM- A SILVERY METALLIC TRANSITION METAL

Europium belongs to the group of metals known as rare earth elements. Yttrium shares some common properties with these elements and much of its chemistry is similar to these elements in their +3 oxidation states. Yttrium <sup>[2]</sup>- a silvery metallic transition metal has the symbol Y, **Figure 3** shows the symbol and atomic number 39. It occurs commonly with rare-earth minerals. **Figure 4** shows the Yttrium metal.

Yttrium (named for Ytterby, a Swedish village near Vaxholm) was discovered in 1794 by a Finnish chemist, physicist and



**Figure 1:** Electroluminescent wire belt tape.



**Figure 2:** Electroluminescent panel.



**Figure 3:** Symbol of Yttrium.



**Figure 4:** Yttrium.

mineralogist named Johan Gadolin from Ytterby. In 1828, Friedrich Wohler isolated it as an impure extract of yttria through the reduction of  $YCl_3$ . Ytteria was the name used for most basic ones and other were re-named as erbia and terbia.

Yttrium is found in rare earth minerals and also in Uranium ores but not as a free element. By reduction of yttrium fluoride with calcium metal and by some other techniques, it can be commercially produced. Y-89 is the only one isotope of natural Yttrium. It is lustrous, relatively stable in air and resembles like Lanthanides chemically. When the temperature exceeds 4000C, metal shavings or turnings can be ignited. It has low neutron cross-section for nuclear capture. It has a common oxidation state of +3. The most important Yttrium Compound-Yttrium (III) Oxide is widely used to make red emitting  $YVO_4:Eu^{3+}$  and  $Y_2O_3:Eu^{3+}$ . In modern technology, Yttrium oxide is used as in Yttrium iron garnets which are very effective microwave filters. Yttrium iron garnets have interesting magnetic properties and are used as very efficient acoustic energy transmitter and transducer.

Yttrium has no biological role and is highly toxic for human beings and animals. It is also used as a material enhancer, a superconductor, in the medical field as a drug (Y-90) for treatment of various cancers etc. <sup>[9,10]</sup>.

## EUROPIUM- MOST REACTIVE RARE EARTH ELEMENT

Atomic number of Europium is 63, **Figure 5** shows the symbol of Europium and was named after Europe (a Continent). It is the one of the most reactive of the rare earth elements and instantly oxidizes in air. When reacted with water, it resembles like Calcium. It ignites in air around 150°C to 180°C. It is also ductile and its hardness is approximately equal to the hardness of Lead. In 1890, existence of Eu was first found by Paul Emile Lecoq Boisbaudran but however, its discovery is generally credited to French Chemist Eugene-Antole Demarcay. **Figure 6** shows the Europium metal.

<sup>151</sup>-Eu and <sup>153</sup>-Eu are the two stable isotopes of naturally occurring Europium in which <sup>153</sup>-Eu is the most abundant one and its natural abundance is 52%. In nature Europium is never found in Free State, however many minerals are there which contains Europium in which the most important sources being bastnasite and monazite. It is also identified in the spectra of certain stars and the sun. It is isolated commercially by ion exchange and solvent extraction.

Europium is mildly toxic. Europium is used to dope some types of glass to make lasers and has a few commercial uses. It is used for screening for Down's syndrome and some other genetic diseases. It is also used in nuclear reactors due to its ability to absorb neutrons.  $Y_2O_3$  doped with  $Eu^{3+}$  is widely used as a red phosphor in television sets and in fluorescent lamps.  $Eu^{3+}$  is also used as an activator for many other yttrium-based phosphors. In interrogation of bimolecular interactions in drug discovery screens, Europium fluorescence is used <sup>[9,11]</sup>.

## $Y_2O_3$ MATERIAL- A TOPOLOGICAL LUMINOPHOR

Yttrium Oxide is also known as Yttria. It is a white solid substance which is stable in air <sup>[12]</sup>. **Figure 7** shows the Yttrium Oxide powder. Yttrium Oxide is a refractory material and has less thermal expansion than zirconium, alumina etc. Its surface morphology



Figure 5: Symbol of Europium.



Europium Metal

Figure 6: Europium metal.

depends on the condition of its preparation. Yttrium Oxide is soluble in acids and is also slightly soluble in water and is available in the market in various purities between 99.9% and 99.99%.

$Y_2O_3$  is mainly extracted from the mineral  $YPO_4$  (Xenotime).  $Y_2O_3$  has a good transparency for infrared radiation and has high thermal stability. It has affinity for Sulphur and Oxygen. It is used as an additive to stabilize Zirconia and as a sintering aid in Silicon Nitride. It has a good transmission range from 1 to 8 micrometer wavelength in the infrared region. It is a good resistant to erosion and thermal shocks and becomes ideal for protection domes for infrared sensors.

The crystal structure of Yttrium Oxide is BCC (Body Centered Cube) for use in phosphors application. In this arrangement, each  $Y^{3+}$  cation is surrounded by six oxygen atoms located at the corners of cube. In the lattice there are two different  $Y^{3+}$  cation sites. Two corners are vacant and they can be along a body or face diagonal of the cube resulting in two  $Y^{3+}$  site symmetries known as  $S_6$  and  $C_2$  respectively. The ratio of  $C_2$  to  $S_6$  sites is 3:1. **Figure 8** shows crystal structure of  $Y_2O_3$ .

There are various methods to prepare Yttrium Oxide and it has different aspects with rare earth and non-rare earth doped different shapes (wires, rods, cones, sphere and flower like) surface morphology and different crystal structures. Selection of an appropriate organometallic precursor and also controlling its combustion helps in the synthesis of monoclinic as well as cubic  $Y_2O_3$  nanorods,  $Y_2O_3$  changes morphology from nanosphere. The luminescence properties of rare earth doped  $Y_2O_3$  depends on surface morphology of phosphor particles. It also depends on annealing time and this annealing time depends on the size of Yttrium Oxide and on Photoluminescence intensity <sup>[10]</sup>.

It is used in making material for solid-state laser. For efficient operation in continuous wave mode and in pulsed mode in lasers, Ytterbium is used as a dopant or doping agent. It is used in gas lighting which is now obsolete these days. It is also used in making dental ceramics like stabilization of Zirconia. It is used in making Yttrium iron garnets for microwave filters and is also used to make superconductors like  $YBa_2Cu_3O_7$  and in synthesis of inorganic compounds. To sum up they find applications in sensors, laser, Fluorescence Bio-Imaging (FBI), photoluminescence, cathodoluminescence, electroluminescence and additive coatings in MOSFET devices or optoelectronic devices <sup>[12]</sup>.

## TOPOLOGICAL LUMINOPHOR MODEL OF $Y_2O_3:Eu^{3++}Ag$ WITH HIGH ELECTROLUMINES- CENCE PERFORMANCE

For promising applications in optoelectronic devices, biomedical analysis, quantum information technology and so on, luminescent materials are used. Enhancement of luminescent intensity is the hot spot for research and in this research, rare earth (f-orbital configuration) luminescent materials have received much interest among other luminescent materials because of their stability and their luminescence dating. Some precious metal nano-materials are also used to improve the performance of rare earth luminescent materials.



Figure 7:  $Y_2O_3$  Powder (Topological Luminophore).

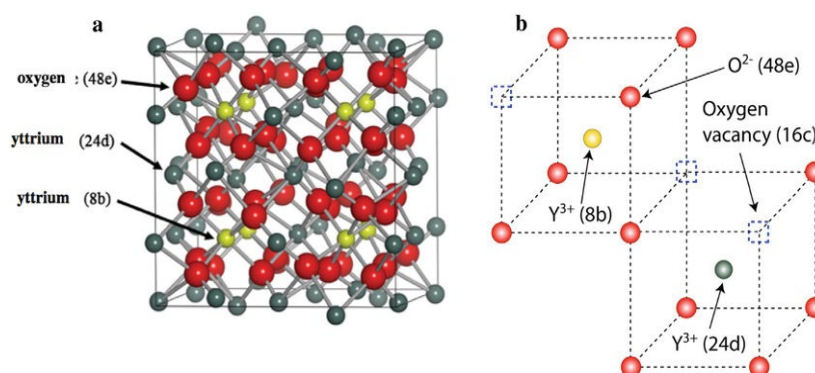


Figure 8: Crystal Structure of  $Y_2O_3$  (Cubic-C type).

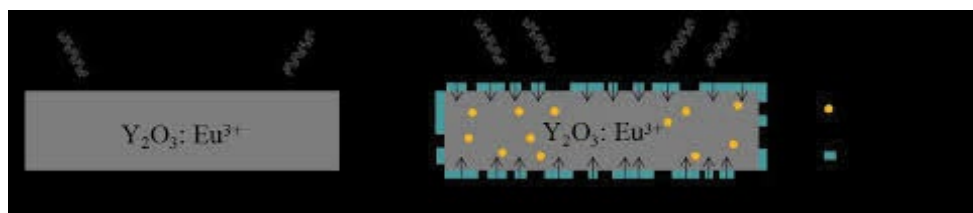


Figure 9: Luminophor Model of  $Y_2O_3:Eu^{3+}$ .

### Topological Luminophor Model

It is based on the hydrothermal preparation  $Y_2O_3:Eu^{3+}$  micro sheet phosphors and its fundamental model were designed by giving references of concepts based on topological materials which were prepared just via illumination. In this model,  $Eu^{3+}$  acts as luminescence center and  $Y_2O_3:Eu^{3+}$  phosphor was excited to produce electroluminescence under the application of electric field. Ag nano-crystal structure enhances the photoluminescence of the samples to comply with surface plasma resonance. Hence the performance of  $Y_2O_3:Eu^{3+}Ag$  is greatly improved. shows the luminophore Model of  $Y_2O_3:Eu^{3+}$ .

### Reagents Used

Rare earth oxides like  $RE_2O_3$  ( $RE=Y$  and  $Eu$ , AR) and  $NaOH$ ,  $HCl$  and dehydrated ethanol (AR),  $AO$  (AR,99%) and silver nitrate and no further purification was done.

### Preparation

$Y_2O_3:Eu^{3+}Ag$  phosphor, synthesis was done through illumination after using methods based on hydrothermal conditions. A typical procedure- 0.153 g of  $Y_2O_3$  powders and  $Eu_2O_3$  powders (molar ratio=0.95:0.05) were taken and stirred and heated with  $HCl$ . After sometime, a milky white solution was turned into a viscous transparent solution. Excess  $HCl$  was removed by heating and white crystals were separated.  $Y(Eu)Cl_3$  was prepared and was dissolved into Ammonium Oxalate solution under constant magnetic stirring. Into the mixed solution  $AgNO_3$  was added into mixed solution and pH was maintained at 9. Then the mixture was transferred to a Teflon-lined autoclave being treated solvothermal and autoclave was cooled to room temperature, centrifuged and washed several times. Calcination of  $Y_2O_3:Eu^{3+}AgCl$  was done at  $800^\circ C$  and the sample was kept in sunlight for 72 hours till nits colour changes to gray due to decomposition of  $AgCl$  into  $Ag$ . **Figure 10** shows its preparation.

### Characterizations

XRD (X-Ray Diffraction) technique was performed with High Resolution Transmission Electron Microscopy (HRTEM) with an



Energy Dispersive Spectrometer (EDS). Sample's surface size was recorded by Bruker dimension fast scan and dimension icon AFM (Atomic Force Microscope). **Figure 11** shows the EL Spectra. The EL structure is a typical sandwich structure and EL emission spectrum was measured by fiber optic spectrometer.

**Results**

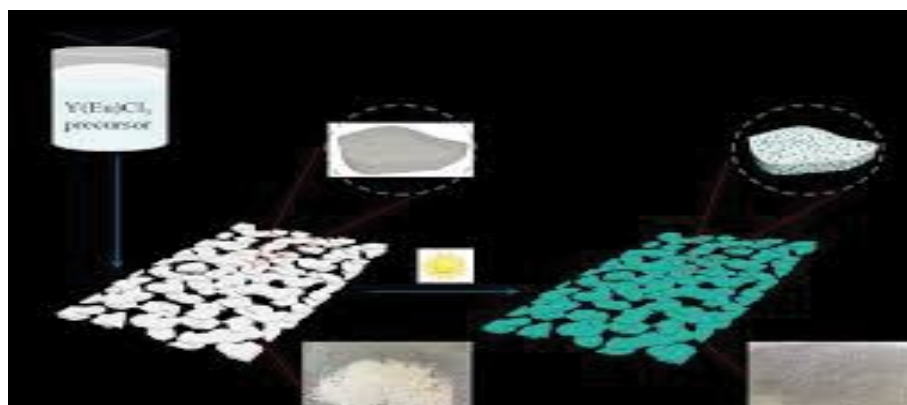
The luminescence performance of  $Y_2O_3:Eu^{3+}Ag$  was increased by about 300% by composite luminescence consisting of Electroluminescence and Photoluminescence. Thus, this proposed design of luminophor gives a new approach to further improve electroluminescence intensity of phosphors [13].

**ELECTROLUMINESCENCE OF  $Y_2O_3: Tb$  NANOPHOSPHOR**

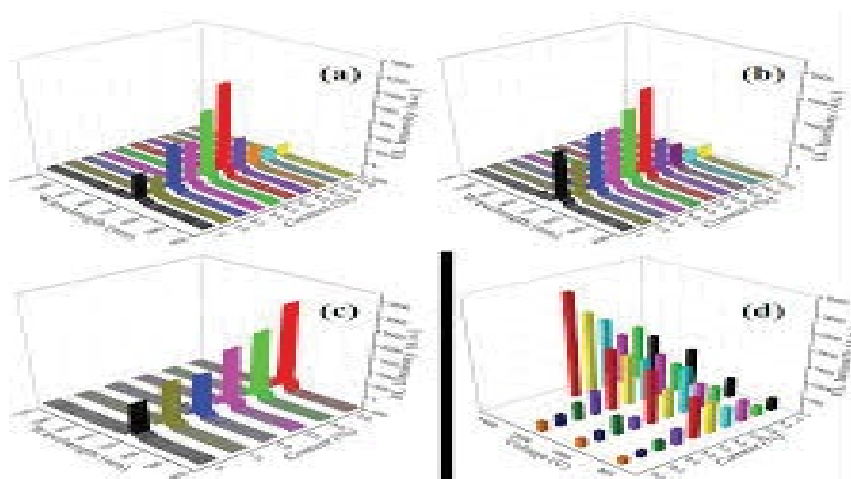
The development of nanocrystalline phosphors has become a trend due the ever-shrinking size of the recent technology. Particularly this oxide is chosen as the host material because of its low phonon frequency which makes non-radiative relaxations in excited states inefficient. Rare earths as doping ions are considered as elements which are activators of phosphors as they have some particular optical properties and because of their unique electronic configuration. If we consider  $Tb^{3+}$  ions, it has 4f electrons and the 4f shell readily releases an electron and an intra-ionic transition takes place at low energy from  $4f^8$  to  $4f^75d$ . The 5d orbit is strongly influenced by the electric field of surrounding ions as it is the outermost orbit which creates efficient absorption bands. Several techniques like aerosol pyrolysis, sol-gel method, liquid phase reaction etc. are incorporated in nanocrystalline  $Y_2O_3$  matrix with rare earths. **Figure 12** shows the mechanism of emission of white light.

**Preparation**

In this Europium oxide (99.99%), Yttrium nitrate (99.99%), nitric acid, urea (90%) was used as raw material.  $RE(NO_3)_3$  stock solutions were prepared by dissolving  $Tb_2O_3$  in  $HNO_3$ . Then in a beaker, the solutions of  $Y(NO_3)_3$  and  $Tb(NO_3)_3$  were mixed according to the formula  $(Y_{0.95}Tb_{0.05})_2O_3$  and suitable amount of urea is added. The sample finally is transferred to crucible and heated in a furnace at  $600^\circ C$ . The reaction for synthesis is given as-  $(2-2x)(NO_3)_3 + 2xTb(NO_3)_3 + 5NH_2CONH_2 \rightarrow (Y_{1-x}Tb_x)_2O_3 + 5CO_2 + 8N_2 + 10H_2O$



**Figure 10:** Schematic Representation for preparation of  $Y_2O_3:Eu^{3+}Ag$  phosphor.



**Figure 11:** EL Spectra of topological luminophor  $Y_2O_3:Eu^{3+}Ag$ .

**Characterization of sample**

The samples were characterized by TEM (Transmission Electron Microscopy), XRD and SEM (Scanning Electron Microscopy). The X-Ray diffraction pattern of the prepared samples are recorded in a wide range of Bragg angle by the use of Bruker D8 advanced XRD measuring instrument with copper as a target radiation. The FTIR (Fourier Transform Infrared) was recorded in the range of  $4000\text{ cm}^{-1}$  to  $400\text{ cm}^{-1}$  (wavenumber range) by the use of Shimadzu FTIR spectrometer. TEM was Zeiss EVO40 microscope and EL intensity was measured with the help of photomultiplier tube. **Figure 13** shows the XRD pattern of  $\text{Y}_2\text{O}_3:\text{Tb}$  Nanophosphor.

**Result**

It is observed that the light emission starts as a threshold voltage and increases with the increasing value of voltage applied. The EL efficiency depends on the carrier life time of the charge carriers injected. Input voltage also increases with the continuous increase in current. This also shows ohmic nature. Also, on increasing frequency of the input signal, EL brightness increases and attains a saturation value and after this value, dissipation of heat takes place which leads to decrease in brightness and damage of EL device. Thus, this helps in the increase in EL efficiency of devices. **Figure 14** shows electroluminescence of  $\text{Y}_2\text{O}_3:\text{Tb}$  Nanophosphor [14].

**ELECTROLUMINESCENCE OF  $\text{Y}_2\text{O}_3:\text{Eu}$  AND  $\text{Y}_2\text{O}_3:\text{Sm}$  FILMS**

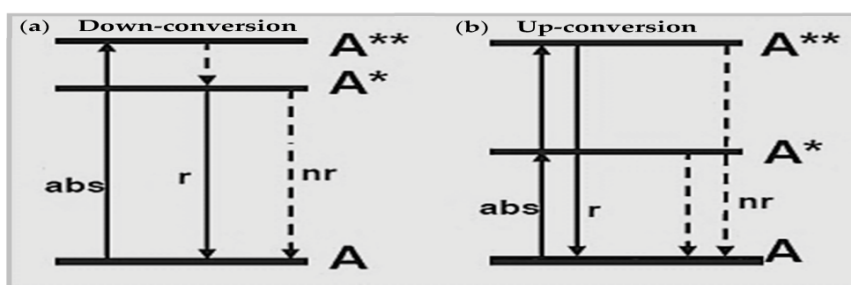
To create full-color electroluminescent indicators, there is a development of high-luminance film electroluminophors (compared with ZnS films doped by Mn) for high efficiency and emission intensity. Highly stable oxide and oxy-sulfide have attracted the attention of many researchers and scientists.  $\text{Y}_2\text{O}_3$  films are the most stable luminophores and we are using various methods to increase the luminance and for creation of these luminophores. The probability of transition for  $\text{Eu}^{3+}$  and  $\text{Sm}^{3+}$  ions are calculated by various methods. Practically at some particular value of electric field strength, the emission intensity cannot exceed the luminescence intensity of all excited ions. **Figure 15** shows a thin film EL indicator.

**Preparation**

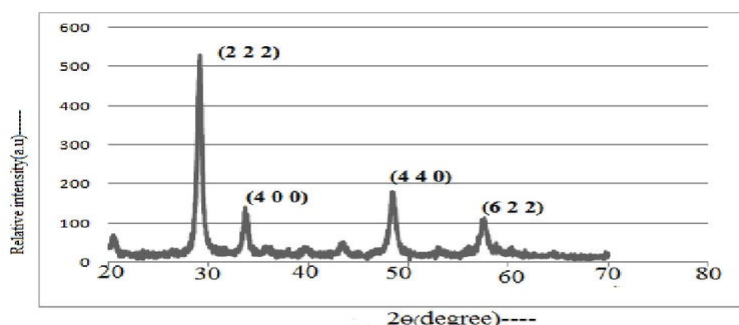
By applying pressure on the powder which consists of  $\text{Y}_2\text{O}_3$  and  $\text{EuF}_3$  with different concentrations of  $\text{EuF}_3$ ,  $\text{Y}_2\text{O}_3$  films were obtained by electron beam evaporation of pellets. Powder of  $\text{SmF}_3$  is also added as a co-doping impurity to the input material. The substrate's temperature was maintained between  $120^\circ\text{C}$  to  $200^\circ\text{C}$ . The specimens are annealed out in air and in oxygen-argon atmosphere for an hour at different temperatures like  $600^\circ\text{C}$ ,  $750^\circ\text{C}$ ,  $900^\circ\text{C}$  and  $1050^\circ\text{C}$ . The luminescent structure includes ceramic substrate with metal electrodes built-in them and with 40 micrometer thick working layer of ceramics based on  $\text{BaTiO}_3$ . The luminescent layer is positioned between thin film dielectric layers which are  $0.05\text{ }\mu\text{m}$  to  $0.1\text{ }\mu\text{m}$  thick and on the top, a layer of transparent ITO electrode was placed.

**Characterization**

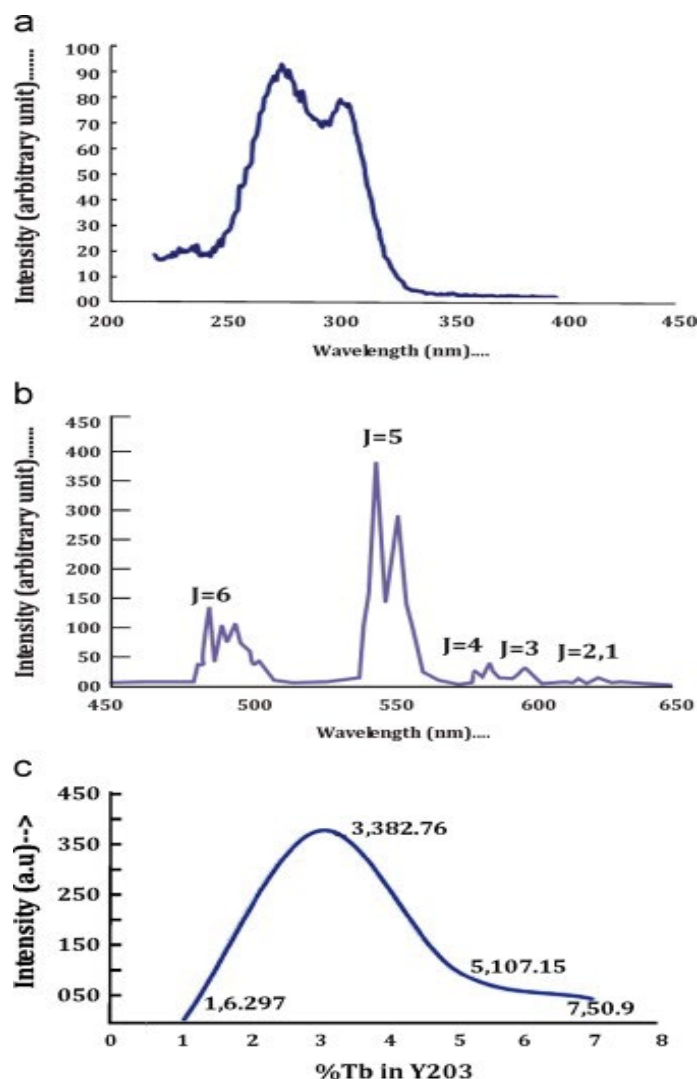
The emission spectra and luminance were studied by a diffraction luminescence spectrometer SDL-2 and FPI and FPCh-



**Figure 12:** Nanophosphor based white light.



**Figure 13:** XRD pattern of  $\text{Y}_2\text{O}_3:\text{Tb}$  Nanophosphor.



**Figure 14:** Electroluminescence of  $Y_2O_3:Tb$  Nanophosphor.

BPU photometers. With the help of a pulse generator with adjustable frequency, the excitation of electroluminescence was recorded. The film's crystal structure was studied by X-ray diffraction with a spectrometer DRON-3M. With the help of atomic force microscope, surface morphology of the films was investigated.

**Result**

**Figure 16** shows spectral characteristics of electroluminescence of  $Y_2O_3:Sm$  Films. Spectral compositions of the films have not changed. The addition of Gallium and high temperature annealing caused increase in the electroluminescent emission intensity. The electroluminescent emission intensity was increased up to 4000  $cd/m^2$  with increase in the temperature of films up to 1000°C. To increase the electroluminescent emission intensity up to 8000  $cd/m^2$   $SmF_3$  impurities are doped. This does not change the emission spectrum. Thus, helps in increasing the electroluminescent intensity.

**APPLICATIONS OF ELECTROLUMINESCENT  $Y_2O_3$  MATERIAL**

**Yttrium Oxide Based Lasers**

$Y_2O_3$  material is used in solid state laser. They allow operation in both continuous wave mode and in pulsed mode. During high concentrations of excitations, electroluminescent emission quenches at frequency of laser and avalanche broadband emission takes place.

**As Gas Lighting**

It produces artificial gases like  $H_2$  (hydrogen), coal gas, paraffin and other products and converts into visible light which can be seen through the naked eyes.



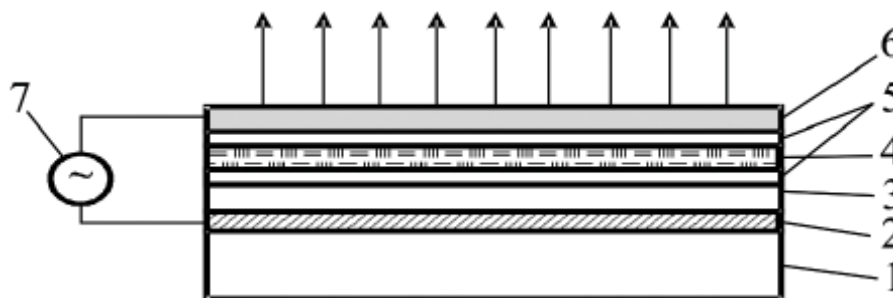


Figure 15: Structure of thin film EL indicator.

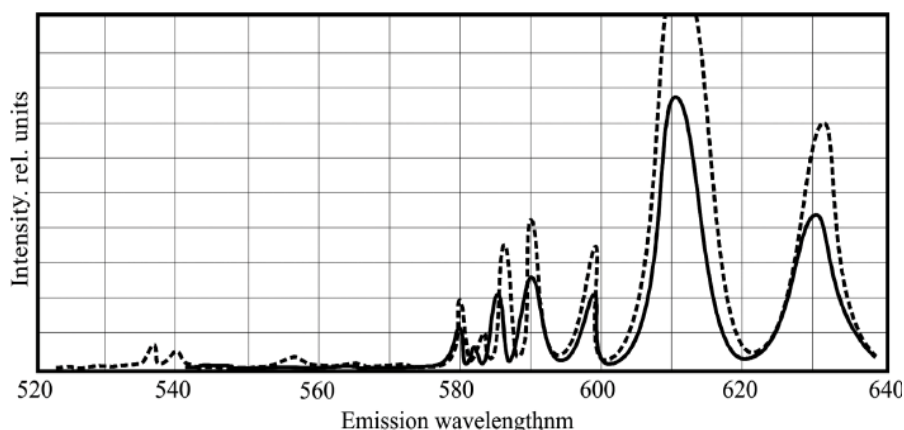


Figure 16: Spectral characteristics of electroluminescence of  $Y_2O_3:Sm$  Films.

**In Dental Ceramics**

$Y_2O_3$  is used for stabilizing Zirconia which is a very hard ceramic used in dentistry.

**In making Microwave Filters**

$Y_2O_3$  is used for making Yttrium Iron Garnets (Garnets are used in data storage, as solid-state lasers in Faraday rotators and in various non-linear optics applications) which are very efficient for making microwave filters [12,15].

**$Y_2O_3:Eu^{3+}Ag$  with high Electroluminescence Performance**

The luminescence performance of  $Y_2O_3:Eu^{3+}Ag$  was increased by about 300% by composite luminescence which consists of electroluminescence. This proposed design of luminophor gives a new approach to further improve electroluminescence intensity of phosphors and new designs of phosphors [13].

**Electroluminescence of  $Y_2O_3:Tb$  Nanophosphor**

The development of nanocrystalline phosphors has become a trend due the ever-shrinking size of the recent technology. Rare earths as doping ions are considered as elements which are activators of phosphors as they have some particular optical properties and because of their unique electronic configuration. Thus, this helps in the increase in electroluminescent efficiency of devices [14].

**Electroluminescence of  $Y_2O_3:Eu$  and  $Y_2O_3:Sm$  films**

To create full-color electroluminescent indicators, there is a development of high-luminance film electroluminophors (compared with ZnS films doped by Mn) for high efficiency and emission intensity. Highly stable oxide and oxy-sulfide have attracted the attention of many researchers and scientists.  $Y_2O_3$  films are the most stable luminophores and we are using various methods to increase the luminance and for creation of these luminophores. This does not change the emission spectrum. Thus, helps in increasing the electroluminescent intensity [16].

**CONCLUSIONS**

The old picture of EL devices has totally changed due to the introduction of electroluminescent  $Y_2O_3$  material and new devices are emerging with better performance than the old ones. Scientists are looking forward to develop such devices incorporating this electroluminescent luminophor which are highly efficient, have low-cost and are reliable. Research is still going on to develop new materials and devices incorporating electroluminescent  $Y_2O_3$  material and to also improve the old ones to meet the future demand and to make the technology reach to every home in this world for everyday use.

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