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Synthesis, structure, and luminescence properties of a novel double-perovskite Sr₂LaNbO₆:Mn⁴⁺ phosphor



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ABSTRACT

A novel phosphor, $\rm Sr_2LaNbO_6:Mn^{4+}$, is synthesized by conventional solid-state reaction. The phosphor can be excited at 300–500 nm and emit a deep-red light. An emission band peak of 694 nm is observed from the spin-forbidden transition $^2E \rightarrow ^4A_2$ of $\rm Mn^{4+}$ ions. Energy transfer between the $\rm Mn^{4+} - Mn^{4+}$ pairs decreases fluorescence decay time from 0.815 ms to 0.512 ms. The resistance of luminescence against thermal impact is also investigated by emission spectroscopy at heating temperatures from 300 K to 500 K. The calculated activation energy ΔE (\sim 0.345 eV) demonstrate good thermal stability of the phosphors. All these results indicate that the phosphor can be applied in white-light-emitting diodes.

1. Introduction

White-light-emitting diodes (W-LEDs) are extensively used as light sources because they are highly efficient and environmentally friendly and have long service lives [1,2]. Many W-LEDs are generally produced by combining yellow phosphors (YAG:Ce³⁺) with blue indium gallium nitride (InGaN) chip [3-6]. However, the color-rendering index of W-LEDs is not sufficient because of the absence of red components. Therefore, exploring red phosphors that can be used in W-LEDs is an important concern [7]. In recent years, Eu²⁺-, Eu³⁺-, Ce³⁺-, and Sm³⁺-doped phosphors have been developed to compensate for the red component of W-LEDs [1.8.9]. However, these phosphors have some disadvantages, such as the high cost of the rare earth and presence of toxic properties [1]. The excitation spectra of the widely used Eu²⁺-doped phosphor range from the green to the red range because of the $4f^7 \rightarrow 4f^65d$ transitions of Eu²⁺. Thus, these phosphors are not generally preferred.

 $\rm Mn^{4^+}$ -doped phosphors attract considerable attention because of their broad excitation wavelength and deep-red photoluminescence quality, and thus they are extensively used to improve the color rendering property and increase the efficiency of W-LEDs [10–12]. The $\rm Mn^{4+}$ ion has a $\rm 3d^3$ electron configuration, and the 3d electrons can be distributed in the 5d orbitals through the 10

allowed states [13]. Attributed to the crystal field splitting of the Mn 3d states into degenerate $T_{\rm 2g}$ and $E_{\rm g}$ states, the large gap can stabilize Mn $^{4+}$ in an octahedral environment [13]. The red emission and near-infrared emission of the Mn $^{4+}$ -doped phosphors result from the typical spin-forbidden transition $^2E \!\rightarrow \,^4\!A_{2g}$ of Mn $^{4+}$ [1].

The double perovskite compound A₂BB'O₆ has been widely used in the luminescence field [14,15]. Compared with traditional noble metal catalysts, perovskite-mixed oxides are demonstrated to be valuable alternatives because they are cheap and have thermally stable properties. A₂BB'O₆ is constructed through a three-dimensional network, in which the alternate corners are shared by the BO₆ and BO'₆ octahedras [16]. The A²⁺ cation is at the interstitial space and forms the cubo-octahedral geometry. Sr₂LaNbO₆ belongs to double perovskite crystals and has a cubic crystal system. In the present work, it is used as the base material for doping Mn⁴⁺ and synthesized by conventional high-temperature-solid-state reactions. Furthermore, the luminescence properties, fluorescence lifetimes, thermal quenching of luminescence, and International Commission on Illumination (CIE) chromaticity coordinates are also investigated.

2. Experimental procedure

2.1. Materials and synthesis

The Mn⁴⁺-doped Sr₂LaNbO₆ was prepared through the standard solid-state reaction, using high purity starting powders of SrCO₃,

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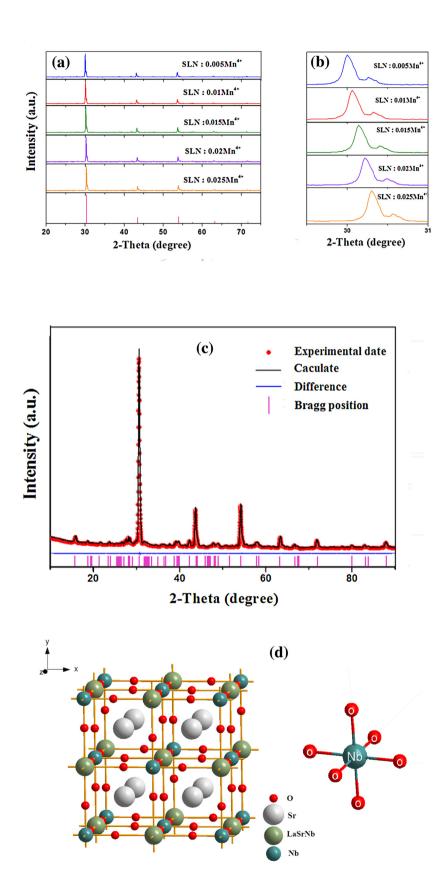


Fig. 1. (a) X-ray powder diffraction patterns of Sr_2LaNbO_6 : xMn^{4+} (x = 0.005, 0.01, 0.015, 0.02, 0.025) phosphors and the Sr_2LaNbO_6 JCPDS standard pattern. (b) magnified XRD curves in the range of 29–31° (c) Rietveld refinement result of Sr_2LaNbO_6 : xMn^{4+} (d) Projection view of crystal structure of Sr_2LaNbO_6 unit cell and the coordination environment of Nb^{5+} .

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