



Synthesis of BaF₂:Ce nanophosphor and epoxy encapsulated transparent nanocomposite

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ABSTRACT

BaF₂:Ce nanophosphors with a peak emission at 355 nm were synthesized using a chemical precipitation method with oleic acid as the capping ligands. The cerium doping concentration was optimized and it was found that the photoluminescence intensity reached a maximum at about 15 mol% of Ce doping. Two distinct photoluminescence excitation peaks were observed from all samples at about 255 and 290 nm. The nanophosphors were incorporated into an epoxy matrix to form bulk nanocomposite samples for potential scintillation applications. Index matching between the nanophosphor and the epoxy matrix is critical to obtain high transparency.

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1. Introduction

Nanoparticles and nanomaterials have been intensively studied for the past decade due to their unique size-dependent properties such as optical, electrical, magnetic and mechanical properties, which are largely different than those of their equivalent bulk materials [1–3]. As one of the most attractive nanomaterials, polymer nanocomposites show great potential for applications in science and technology because they combine the advantages of polymeric materials, such as low cost and easy fabrication, with the functional properties of nanoparticles [4]. For optical nanocomposites with luminescent nanophosphors blended in a clear organic matrix, high transparency is desired to maintain the function of the nano light emitters for potential applications such as optical amplifiers, X-ray imaging and radiation scintillation.

Scintillation detectors made from nanocomposites embedded with nanophosphors may be of advantage for gamma ray detection [5,6]. Use of these nanomaterials may bypass the difficulties of preparing large pure crystals to assure effective transparency for the generated light. Preparation of suitable particle size and doping will also permit selection of a photon wavelength that optimally matches the photodetector response curve to increase the number of photons collected per pulse and improve resolution. The challenge is to fabricate a highly transparent nanocomposite. Although the small optical scattering from nanoparticles

with diameters less than 50 nm made it possible to synthesize transparent nanocomposites [6], a highly homogeneously dispersed and/or refractive index matched nanocomposite system is required, especially for nanocomposite scintillators with extremely high nanoparticle loading necessary for gamma ray absorption efficiency and stopping power.

Research reports have focused on the non-agglomeration and homogeneous distribution of nanoparticles in the polymer matrix through surface modification of the nanocrystals by a number of strategies ranging from multivalent surface passivation to functionalized chain-end attachment [4,7]. However, little has been reported on fabrication of transparent light emitting nanocomposite by matching the indices of nanoparticles and the matrix. In this paper, we report the synthesis of cerium doped BaF₂ nanophosphors and transparent nanocomposites by index matching. Bulk single crystalline BaF₂ and BaF₂:Ce have been used as traditional scintillators for gamma ray detection [8–10]. BaF₂ has a low refractive index of about 1.478 at a wavelength of 500 nm and can be relatively easily matched with a proper polymeric matrix.

Synthesis of BaF₂ nanocrystal and Ce doped BaF₂ nanoparticles has been reported by various techniques such as hydrothermal reaction [11,12], liquid–solid–solution method [13], chemical precipitation [14,15], microemulsion [16,17] and thermal decomposition of barium trifluoroacetate [18]. For example, monodisperse oleic acid capped BaF₂ nanocrystals were synthesized by a liquid–solid–solution approach. Their size, shape and phase could be well controlled by the Ostwald ripening process in a heated autoclave at 140–250 °C [13]. Hua et al. reported Ce doping in BaF₂ nanocrystals prepared by microemulsion, yielding a strong emission peak located at 365 nm [16].

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2. Experimental

BaF₂:Ce nanophosphors were synthesized by an adaptation of a chemical precipitation synthesis for LaF₃:Ce [19]. BaF₂ with cerium doping levels from 1 to 30 mol% was prepared. For example, in a typical synthesis of BaF₂: 1% Ce, 3.2 g of NaF was dissolved in 200 ml of de-ionized water and mixed with 7.2 g of oleic acid dispersed in 200 ml of ethanol. The resulting mixture was stirred and heated to 78 °C. Then a solution of 9.86 g of Ba(NO₃)₂ and 0.174 g of Ce(NO₃)₃·6H₂O in 160 ml of water was added drop-wise into the NaF solution to form the white BaF₂:Ce precipitate. After stirring at 78 °C for another hour, the mixture was cooled to room temperature. To remove the residual oleic acid and inorganic salt, the white precipitate was washed three times with ethanol and de-ionized water in turn by ultrasonication, followed by centrifugation at a speed of 3000 rpm for 15 min. About 8 g of nanocrystals were obtained and dried at ambient temperature.

The refractive index of BaF₂:Ce nanophosphors was experimentally determined with index-matching fluids of known refractive indices obtained from Cargille Labs Inc. (Cedar Grove, NJ). Index matching of the nanoparticle agglomerates was performed under a microscope using 400 nm illumination from an LED. After identifying the refractive index, commercial epoxy products were selected to be used as a matrix to encapsulate the nanophosphors. For this investigation, two different commercially available epoxies were used: Epo-TEK 301-2FL and Epo-TEK 305 (Epoxy Technology, Inc., Billerica, MA) with indices of 1.476 and 1.512 at 589 nm, respectively. Both polymers are two part epoxy mixtures and both have room or high temperature cure capabilities. BaF₂:Ce-epoxy nanocomposites with loading levels of 10–200 mg nanophosphor per ml epoxy were prepared by directly mixing the

dry powders with one of the epoxy components. The dimensions of these samples are 32 mm in diameter and 3–15 mm thick. After curing at room temperature, the disks were polished prior to minimize surface scattering.

X-Ray Diffraction (XRD) of packed nanophosphor powders was conducted with an X'Pert PRO α -1 diffractometer equipped with a Cu K α X-ray tube emitting at 1.54 Å. Particle imaging was accomplished using a JEOL 100CX II TEM operating at 100 kV. Photoluminescence (PL) and photoluminescence excitation (PLE) measurements were conducted with a Cary Eclipse Fluorescence Spectrometer and absorption and transmission measurements were performed using a Cary 5000 UV–vis–NIR Spectrophotometer.

3. Results and discussion

BaF₂ nanophosphors with various Ce doping levels were synthesized and characterized. Fig. 1(a) shows the XRD pattern of a 1% Ce doped BaF₂ nanophosphor sample. The peak positions and intensities agreed well with the data reported for pure cubic BaF₂ crystals [17]. Broadening of the diffraction peaks was observed due to the nanoparticle size effect. A mean particle size of 42 nm was calculated based on the Debye–Scherrer formula assuming that the particles are spherical in shape. Via TEM observation as shown in Fig. 1(b), it was concluded that the synthesized powder was composed of individual nanoparticles although they were agglomerated in an ethanol solution. The average particle size was about 40–50 nm, with the smallest particle size less than 20 nm as indicated in the image.

Fig. 2(a) shows the PL spectrum of a BaF₂ nanophosphor sample with 15% Ce doping under excitation at 288 nm. An emission band centered at around 355 nm with a width of

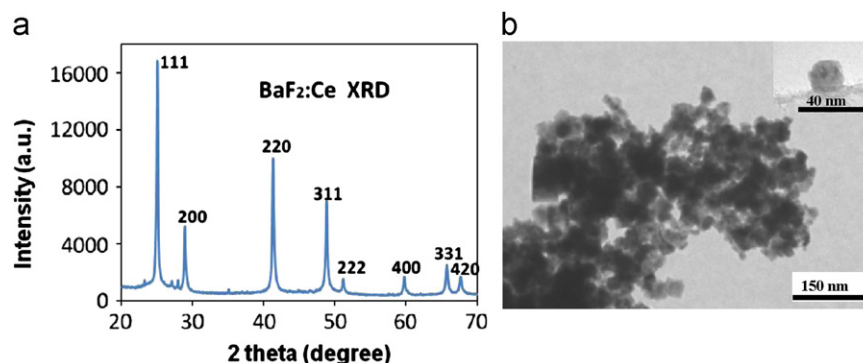


Fig. 1. (a) XRD pattern of BaF₂:1% Ce nanophosphors; the diffraction peaks of cubic BaF₂ crystal are indicated and (b) TEM images of the BaF₂:Ce nanophosphors. Inset shows an individual nanoparticle with particle size about 20 nm.

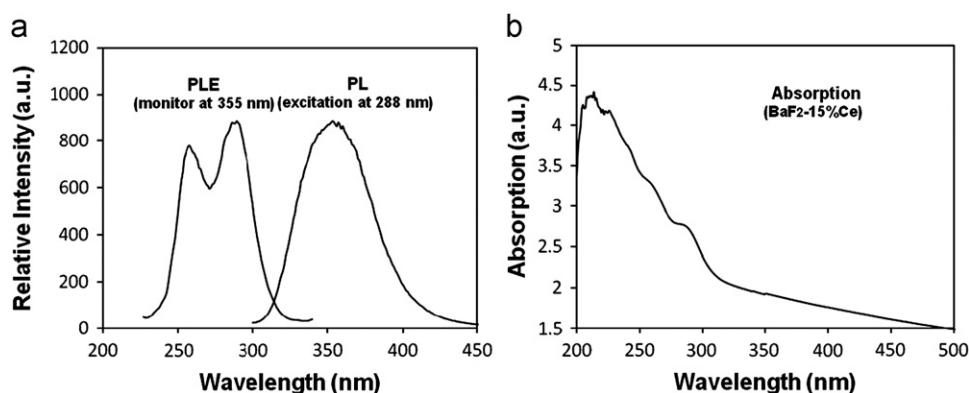


Fig. 2. (a) PL and PLE spectra of BaF₂:15% Ce nanophosphor and (b) absorption spectrum of the BaF₂:15% Ce nanophosphor dispersed in ethanol.

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