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Surface modification and characterization of 8-hydroxyquinoline aluminum/nano-TiO₂



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ABSTRACT

8-hydroxyquinoline aluminum/nano-TiO₂ composites were prepared via in situ. In order to make a better combination of the two component, Octavinyl-T8-silsesquioxane (8-vinyl POSS) was introduced into the composite system. The structure and properties of the samples were characterized by XRD, SEM, XPS, UV-visible absorption, Fluorescence spectra and Time-resolved emission-decay. These results show both β -phase and γ -phase of Alq₃ exist in the composite products. Meanwhile, the addition of nano-TiO₂ and 8-vinyl POSS does not change the crystalline and structure of as-prepared Alq₃. Interestingly, however, XPS spectra shows that two new kinds of interaction forces emerge after the addition of 8-vinyl POSS, owing to the formation of Al–O–Si and Si–O–Ti bond, presumably. Besides, the emission intensity of Alq₃/nano-TiO₂ and Alq₃/8-vinyl POSS/nano-TiO₂ increases by 22% and 33%, respectively. The addition of TiO₂ and 8-vinyl POSS will not affect the position of emission peak, but will increase the transmission efficiency in electron–hole pairs. In the meantime, surprisingly, the modified Alq₃ has a longer lifetime than as-prepared Alq₃.

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1. Introduction

Over the past few decades, a growing number of organic materials have been utilized in the preparation of electronic devices, such as organic light-emitting device [1,2], organic solar cells [3] and organic thin-film transistors [4–6]. 8-hydroxyquinoline (Alq₃), as one kind of critical material in Organic Light-Emitting Diodes (OLEDs), its preparation process is simple and low-cost. What's more, it has exceptional electron conductivity and thermal stability, and can be deposited as a dense film [7,8]. In the previous study, an efficient organic electroluminescent (EL) device was firstly reported by Tang and Van Slyke [9], in which the Alq₃ was applied. There is no doubt that some factors do affect the life span and stability of OLEDs [10,11], such as humidity and oxygen atmosphere, etc. Therefore, Alq₃ must be deposited in the vacuum to avert the fluorescence quenching effect, due to the factors mentioned above [12,13]. In another research, Li Meng ting [14] stated that they had successfully improved the antioxidant activity and anti-stripping performance of OLEDs in a great extent. Unfortunately, they merely did little or nothing with their luminescent properties, which have a vital impact on extending the life span of OLEDs and lowering costs. Accordingly, how to solve this problem is becoming an urgent issue.

In recent years, there are more and more researches on organic/inorganic composite materials and blended with wide band gap semiconductor compounds such as GaN [15], ZnO [16]. As a wide band gap (3.3 eV) semiconductor, nano-TiO₂, with the excite binding energy of 40 meV, is comprehensively used in the field of both photo electricity and catalysis. In addition, it has shown that nano-materials, such as TiO₂ and Poly [2-methoxy-5-(3', 7'-dimethyloctyloxy)-1, 4-phenylenevinylene] (MDMO-PPV) [17,18], can not only promote the electrical conductivity and expand luminous bound, but also widen the absorption range in NIR and UV jointly. Recently, Cuba [19] fabricated a type of Alq₃/ZnO composites and coated it onto a glass substrate using the dip coating method, via which gained a thin film containing 30 wt% of ZnO, exhibiting the maximum luminescence yield. In the early days, Xu [11] has researched the electroluminescent properties of three different device structures based on tris-(8-hydroxyquinoline) aluminum, and further investigated the blue and green light-emitting mechanisms. What is more, Chen [20] reported a facile solution-based route for the synthesis of bis-(8-hydroxyquinoline) cadmium (CdQ₂) complex nanorods and nanoflowers in an oleic acid-sodium oleate-ethanol-hexane (or not)-H₂O system. They investigated that the existence of hexane could sabotage the anisotropic crystal growth of CdQ₂/bis-(8-hydroxyquinoline), leading to shorter nanorods, and improved photoluminescence properties.

In this work, as a surface-modify method, nano-TiO₂ with excellent chemical stability, high optical absorption and corresponding lower price, was added into Alq₃, exhibiting perfectly

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safety and photoluminescence properties. Subsequently, the issue that whether this method could have an influence on the luminescent properties of Alq_3 was investigated. As far as I know there were some lacks in information about using nano- TiO_2 to decorate Alq_3 . Whereas, as the icing on the cake, 8-vinyl poss (Octavinyl-T8-silsesquioxane), which possesses a cage-like structure, was introduced into this reaction system, for the purpose of improving the surface activity of composite system. Essentially, the fluorescence emission spectra shows that the modified Alq_3 composites perform better than that of as-prepared Alq_3 . The theory of possible PL mechanism indicates the presumable explanation, which is that the interaction between Alq_3 and nano- TiO_2 has improved the luminous properties. Moreover, the addition of the 8-vinyl poss makes this performance more excellent, which is possibly attributed to the formation of the Ti–O–Si.

2. Experimental

2.1. Materials

Nano- TiO_2 and 8-vinyl poss were purchased from Aladdin and used without further purification. $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$ and anhydrous ethanol were supplied by Tianjin chemical Reagent Red Rock (Tianjin, China). As one of the principal components, 8-hydroxyquinoline was manufactured by Tianjin Guangfu Fine Chemical Research Institute (Tianjin, China). In addition, to adjust the pH value, the ammonium acetate was purchased from Tianjin Hengxing Chemical Reagent Co. Ltd. (Tianjin, China).

2.2. Experimental methods

1.11 g $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$ was dissolved into 17 mL demonized water with ultrasonic dispersion to accelerate the dissolution and obtain solution A. At the same time, 1.45 g 8-hydroxyquinoline was dissolved in 50 mL absolute ethanol, using ultrasonic dispersion to get solution B, which was transparent but slightly yellow.

According to the requirements of different amounts of nano- TiO_2 in each formulation, the weighed nano- TiO_2 of different components were added to the solution B, and then the ultrasonic dispersion solution quickly became turbid yellow. Besides, with the increasing of the amount of nano- TiO_2 added, the color deepened steeply. After that, 0.005 g 8-vinyl poss was put into the reaction system, when A and B (with nano- TiO_2 added) dispersion system were transferred to a three neck flasks, which was under reflux condensation, with magnetic stirring for 30 min, during which the temperature was controlled at 60–70 °C. As the reaction proceeded, ammonium acetate was added to adjust the pH value of the solution for about 6–7. Soon afterwards, chartreuse pulp was obtained. After the pulp filtered by vacuum suction filter and washed by deionized water for two times, once by acetone, then dried in the vacuum for 1 h at 120 °C. Finally, yellow–green powders were gained successfully. The reaction procedure above was presented in a flow chart shown in Fig. 1.

2.3. Characterization

The X-ray diffraction (XRD) measurement was carried out using $\text{Cu K}\alpha$ ($\lambda=0.154056$ nm) radiation with the scan velocity of 1.2°/min and the scan range of $5^\circ < 2\theta < 35^\circ$ in a RigakuD/max. The Search Engine Marketing (SEM) photographs were obtained on the FEI F20 transmission electron microscope (Philips Company). And X-ray photoelectron spectroscopy (XPS) was performed on an AXIS ULTRADLD (Kratos Shimadzu Company), which was operated at about 5×10^{-8} Pa using a 1486.6 eV $\text{K}\alpha$ aluminum X-ray sources. In order to analyze the XPS data, we applied mixed Gaussian–Lorentz

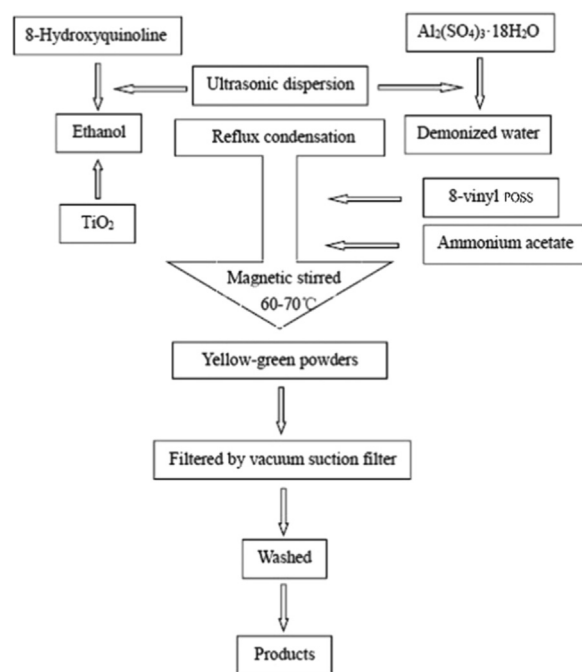


Fig. 1. The preparation process of $\text{Alq}_3/8\text{-vinyl poss/nano-TiO}_2$.

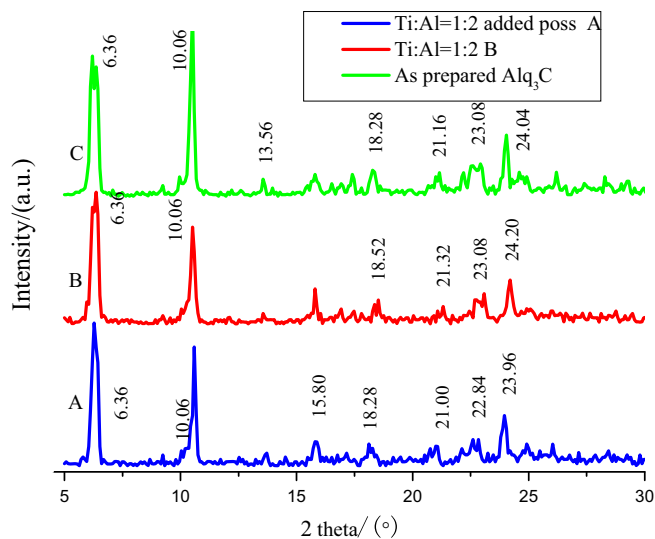


Fig. 2. The XRD pattern of three samples (A) $\text{Alq}_3/8\text{-vinyl poss/nano-TiO}_2$ (B) $\text{Alq}_3/\text{nano-TiO}_2$ and (C) as-prepared Alq_3 , Ti and Al molar ratio of 1:2.

curves to fit the spectra. The fluorescence and emission spectra of the samples were measured by a GANGDONG F-280 fluorophotometer (input slit widths=5 nm, output slit widths=5 nm); the UV absorption spectra was measured by Shimadzu UV-2550. And Fluorescence decay analysis was carried out by Edinburgh Analytical Instruments F900.

3. Results and discussion

3.1. XRD analysis

XRD analysis is used to characterize the crystal properties of Alq_3 . XRD patterns of as-prepared Alq_3 and incorporated Alq_3 ($\text{Alq}_3/\text{nano-TiO}_2$, $\text{Alq}_3/8\text{-vinyl poss/nano-TiO}_2$) are shown in Fig. 2. XRD pattern of samples mentioned above reveals the presence of characteristic diffraction peaks of Alq_3 at $2\theta=6.36^\circ, 10.06^\circ, 24.20^\circ$,

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