

Synthesis and characterization of 8-hydroxyquinolinolato-iridium(III) complex grafted on polyhedral oligomeric silsesquioxane core



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ABSTRACT

In this study, a new iridium(III) complex containing coumarin derivative as the cyclometalated ligand (L) and 7-allyl-8-hydroxyquinoline (Hq-allyl) as the ancillary ligand, Ir(III)bis(3-(pyridin-2-yl)coumarinato N,C^{4'})(7-allyl-8-hydroxyquinolinolato) [Ir(L)₂(q-allyl)], was firstly synthesized as the emissive iridium complex. Then a macromolecule consisting of the emissive iridium complex and carbazole moieties covalently attached to a polyhedral oligomeric silsesquioxane (POSS) core, 7Cz-Ir(L)₂(q)-POSS, was successfully synthesized by hydrosilylation reaction in the presence of platinum(0)-1,3-divinyl-1,1,3,3-tetramethyldisiloxane (Pt-dvs) as the catalyst. The macromolecule has good thermal stability due to the significant effect of the POSS core, and its photoluminescence performance is improved because the POSS core reduced interactions among the emissive units and diminished concentration quenching in the solid state.

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1. Introduction

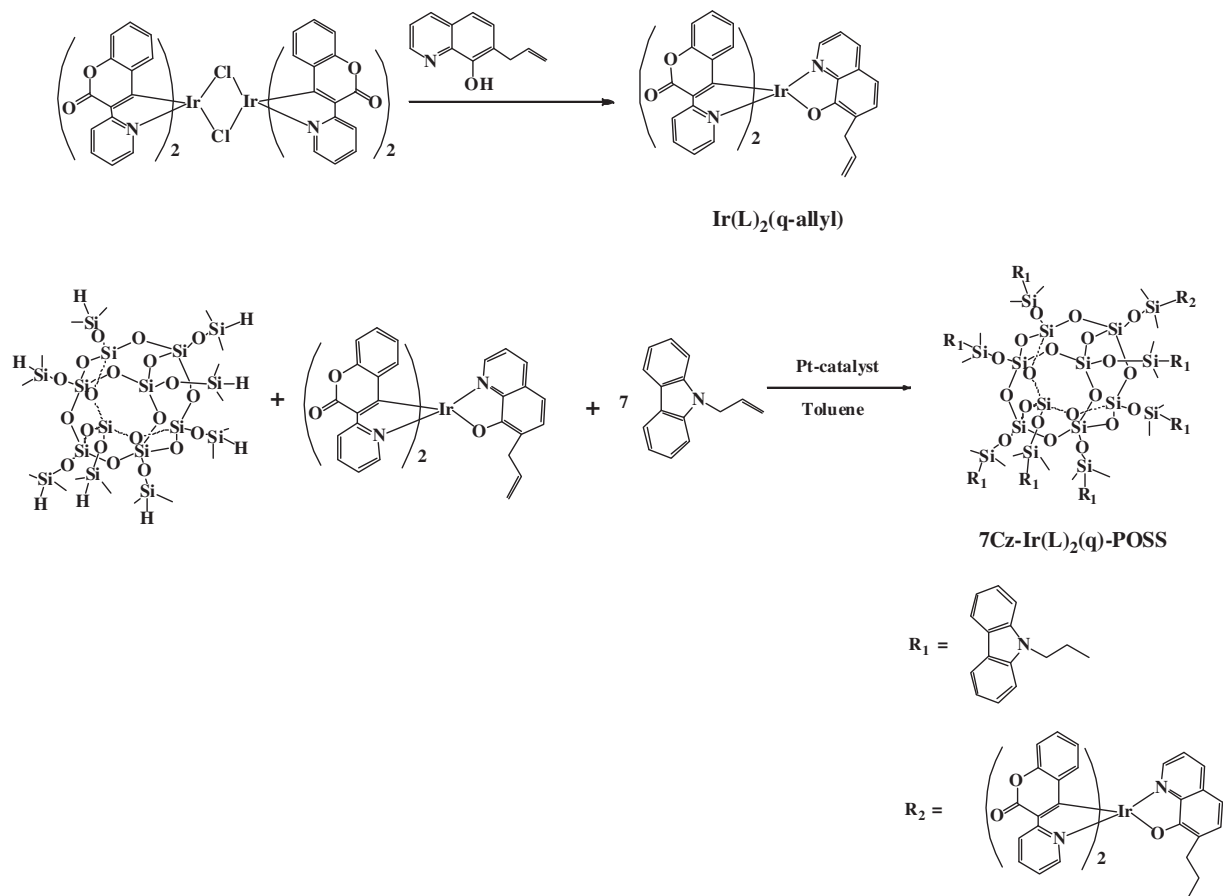
Since tris(8-hydroxyquinolinato)aluminum (Alq₃)-based double layer thin-film device was fabricated [1], a great deal of new materials have been designed and synthesized for organic light-emitting diodes (OLEDs). Metal complexes based on 8-hydroxyquinoline and its derivatives are widely used in organic electroluminescence (EL) due to ease of purification, high luminance and efficiency as well as superior film morphology [2–4]. Among the luminescent metal complexes, aluminum tris(8-quinolinolato) (Alq₃) plays an important role as an electroluminescence and the most stable electron-transporting materials. When the quinoline ring was introduced by different substituents at the different position, the spectral range, quantum efficiency and thermal stability of the materials can be changed [5–8]. For instance, by introduction of electron-donating groups such as methyl substituent on the 2-, 3-, and 4-position of 8-hydroxyquinoline cause a blue shift in the complex emission (495, 520 and 505 nm, respectively) compared to the emission of the parent Alq₃ (525 nm) [9–11], while introduction of methyl substituent on 5-position causes a red shift (545 nm) [10]. The presence of electron-withdrawing groups such as fluoro-, chloro- and cyano groups in 5- or 7-position of 8-hydroxyquinoline results in a blue-shifted emission

(520–530 nm) [9,12,13]. So far many reports on luminescent metal complexes containing different derivatives of 8-hydroxyquinoline have been synthesized for tuning of emission wavelength and improving luminescence performance by changing the central metal ion [14–18], while investigations on iridium(III) complexes containing 8-hydroxyquinolinolato are still rare [19,20]. S. Kappaun and co-workers introduced the electron-withdrawing groups (–CHO and –NO₂) in 5-position of 8-hydroxyquinoline and synthesized the iridium(III) complexes which give orange to red luminescence. However, due to significant concentration quenching in this class materials, the maximum luminances of OLEDs fabricated from these materials were 940 and 490 cd/m², respectively, in nonoptimized device structures.

Recently, polyhedral oligomeric silsesquioxanes (POSS) have attracted a great deal of attention in the materials field because of their unique nanoscale cage-shaped structures and good solubility in organic solvents. POSS molecules are regarded as organic–inorganic hybrid materials at a molecular level. They have been successfully applied in biomedical materials [21], aerospace [22], and semiconducting materials for optoelectronic devices [23]. The incorporation of POSS derivatives into polymeric materials produces nanocomposites with improved properties such as increases in glass transition temperature, thermal stability, mechanical strength, oxidation resistance and ease of processing. In addition, POSS has also been successfully used in the enhancement of luminescence efficiencies and color purities of the

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Scheme 1. The synthetic route of 7Cz-Ir(L)₂(q)-POSS.

conjugated polymers by suppressing undesired excimer formation and intermolecular aggregation.

In this work, we first used 7-allyl-8-hydroxyquinoline (Hq-allyl) as the ancillary ligand to synthesize a new iridium(III) coumarin complex, Ir(L)₂(q-allyl), which was as an emissive iridium complex. Then a macromolecule consisting of the emissive iridium complex and carbazole moieties covalently attached to a polyhedral oligomeric silsesquioxane (POSS) core, 7Cz-Ir(L)₂(q)-POSS, was successfully synthesized by hydrosilylation reaction in the presence of platinum(0)-1,3-divinyl-1,1,3,3-tetramethyldisiloxane (Pt-dvs) as the catalyst. The photophysical properties and thermal stability of the macromolecule were investigated.

The synthetic route of 7Cz-Ir(L)₂(q)-POSS was shown in Scheme 1.

2. Experimental

2.1. Materials and methods

Carbazole and allyl bromide were bought from Alfa Aesar. Platinum complex (platinum-1,3-divinyl-1,1,3,3-tetramethyldisiloxane, Pt-dvs, 2 wt% Pt in xylene) was purchased from Aldrich, USA. Octakis(dimethylsiloxy)silsesquioxane (Q₈M₈^H) containing eight hydro-silane groups was purchased from the Hybrid Plastics Co., USA. 8-Hydroxyquinoline was obtained from Tianjin Chemical Reagent Research Institute (China). Toluene was dried by distillation before use in the hydrosilylation reaction. All other chemicals were analytical grade reagent.

The cyclometalated coumarin ligand (3-(pyridine-2-yl)coumarin, L) and the cyclometalated Ir(III) μ -chlorobridged dimer ((L)₂Ir(μ -Cl)₂Ir(L)₂) were prepared as previously described [24].

7-Allyl-8-hydroxyquinoline (Hq-allyl) was synthesized according to the procedure described in literature [25].

¹H NMR spectra were obtained on Unity Varian-500 MHz. ¹³C NMR spectra were recorded at 100 MHz on a Varian Mercury plus-400 instrument using CDCl₃ as solvent and TMS as internal standard. IR spectra (400–4000 cm⁻¹) were measured on a Shimadzu IRPrestige-21 FT-IR spectrophotometer. C, H, and N analyses were obtained using an Elemental Vario-EL automatic elemental analysis instrument. Mass spectrum was obtained from a Thermo Scientific Orbitrap Elite mass spectrometer (Electrospray ionization, ESI). Thermogravimetric analysis (TGA) was performed on a Perkin-Elmer Pyris system. UV-Vis absorption and photoluminescent spectra were recorded on a Shimadzu UV-2550 spectrometer and on a Perkin-Elmer LS-55 spectrometer, respectively. Melting points were measured by using an X-4 microscopic melting point apparatus made in Beijing Taiké Instrument Limited Company, and the thermometer was uncorrected.

2.2. Synthesis and characterization of Ir(L)₂(q-allyl) and 7Cz-Ir(L)₂(q)-POSS

2.2.1. 9-allyl-9H-carbazole (Cz-allyl)

A mixture of carbazole (10.0 g, 0.06 mol), potassium hydroxide (8.98 g, 0.16 mol) and acetone (100 mL) was stirred vigorously for 2 h at room temperature. Allyl bromide (6.6 mL, 0.072 mol) was

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