

EVA thin film with thermo- and moisture-stable luminescent copolymer beads composed of Eu(III) complexes for improvement of energy conversion efficiency on silicon solar cell



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ABSTRACT

Luminescent beads composed of $\text{Eu}(\text{hfa})_3(\text{TPPO})_2$ (hfa: hexafluoroacetylacetonate, TPPO: triphenylphosphine oxide) in PMMA copolymer (polymethylmethacrylate-styrene and polymethylmethacrylate-trifluoromethylmethacrylate copolymers), PMMA-St-Eu and PMMA-TF-Eu have been reported for improvement of energy conversion efficiency on silicon solar cell. The PMMA-St-Eu and PMMA-TF-Eu beads are prepared using radical initiator AIBN (2,2-azobisisobutyronitrile) without BPO (Benzoyl peroxide) which promotes decomposition of $\text{Eu}(\text{hfa})_3(\text{TPPO})_2$. The emission properties of EVA (ethylene vinyl acetate) film with PMMA-St-Eu or PMMA-TF-Eu beads are characterized by the emission spectra and lifetimes. Thermo- and moisture-stabilities of the EVA films are performed under high temperature and high moisture condition (85°C/85%RH). Increase percentage the solar cell short circuit current efficiency in the solar cell modulation using with EVA film containing PMMA-St-Eu beads with size in 70 μm was estimated to 1.2%. Thermo- and moisture-stable PMMA-St-Eu and PMMA-TF-Eu beads for solar sealing film are demonstrated for the first time.

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1. Introduction

Lanthanide complexes with characteristic emission caused by 4f orbital have been regarded as attractive luminescent materials for use in electroluminescent (EL) devices, laser, and luminescent bio-sensing applications [1–7]. Bünzli and Parker have reviewed various types of luminescent lanthanide complexes [8]. Kido and Koike have described EL devices and plastic optical fibers containing luminescent lanthanide complexes [9–11]. We have also reported strong-luminescent Eu(III), Tb(III), Sm(III), Yb(III) and Nd(III) complexes with large emission quantum yield and radiative rate constants, recently [12]. A large number of scientific studies on luminescence lanthanide complexes have been reported [13].

We here focus on photovoltaic application for enhancement of solar cell efficiency, and highly durability using strong-luminescent Eu(III) complexes. Recently, we have reported photo- and thermo-stable luminescent PMMA beads composed of Eu(III) complexes, which shows photo-durability for ten years [14]. Zhang, Liu and Moudam have also described luminescent PMMA matrix composed of Eu(III) complexes with relatively high emission quantum yields [15–18]. Eu(III) complexes in PMMA matrix provide

luminescence properties with photo-durability. Attachment of polymer films with PMMA beads including Eu(III) complexes on the solar cells directly increases the solar energy conversion efficiency [14].

PMMA matrix is prepared by the polymerization of methylmethacrylate (MMA) with polymerization initiators. Generally, benzoyl peroxide (BPO) and 2,2'-azobisisobutyronitrile (AIBN) are used as a suspension polymerization initiator for lower and higher temperature, respectively. The BPO molecule plays roles of not only as a source of radical initiator but also as an acid-generator under water (Fig. 1a) [19]. The presence of H^+ source in PMMA might lead to decomposition of Eu(III) complex under moisture. With this chemical finding, improvement of PMMA matrix would be required for construction of moisture-stable PMMA beads with Eu(III) complexes.

We here designed novel acid- and water-resistant polymer matrices with bright luminescent Eu(III) complexes, $\text{Eu}(\text{hfa})_3(\text{TPPO})_2$ (hfa: hexafluoroacetylacetonate, TPPO: triphenylphosphineoxide). Luminescent polymer matrices are composed of PMMA (Polymethylmethacrylate) and Styrene (St) or Trifluoromethylmethacrylate (TF) copolymer to control their hydrophobicity. Increase of hydrophobicity in PMMA copolymer leads to increase of blocking rate of water molecules. Luminescent $\text{Eu}(\text{hfa})_3(\text{TPPO})_2$ shows high emission quantum yield in organic

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media ($\Phi = 65\%$). We also consider that the removal of BPO and water in polymer film with Eu(III) complexes are key factor for construction of thermo- and moisture-stable structure. Luminescent PMMA copolymer beads including Eu(III) complexes, PMMA-St-Eu beads and PMMA-TF-Eu beads were prepared by the polymerization of MMA and St or TF with Eu(III) complexes and AIBN under water at 95 °C without BPO molecules (Fig. 1b). Stability of luminescent beads was evaluated under 85 °C and 85% humidity (85 °C/85%RH). Remarkable moisture- and thermo-durability of PMMA copolymer beads with Eu(III) complexes are demonstrated for the solar energy conversion systems.

2. Experimental

2.1. Apparatus

FT-IR measurements were performed at room temperature by Perkin-Elmer system 2000 FT-IR spectrometer. ^1H NMR measurements were measured using JEOL AL-300 spectrometer (400 MHz). Chemical shifts are reported in ppm and are referenced to an internal tetramethylsilane (TMS) standard for ^1H NMR spectroscopy. Short-circuit currents of solar cell module under irradiation were obtained with SAN-EI ELECTRIC XES-220CS1 solar simulator. Thermo- and moisture-stability of solar sealing film sell module was measured by using ESPEC corporation PH-4KT (85 °C/85%RH).

2.2. Preparation of $\text{Eu}(\text{hfa})_3(\text{TPPO})_2$

Methanol (100 mL) containing $\text{Eu}(\text{hfa}-\text{H})_3(\text{H}_2\text{O})_2$ (4.28 g, 6 mmol) and triphenylphosphine oxide (TPPO) (2.78 g, 10 mmol)

was refluxed under stirring for 12 h. The reaction mixture was concentrated using a rotary evaporator. Recrystallization by addition of excess hexane solution produced crude crystals, which were washed in toluene several times. Recrystallization from hot toluene/cyclohexane gave white needle crystals ($\text{Eu}(\text{hfa})_3(\text{TPPO})_2$). Yield: 74%. ^1H NMR (400 MHz, CD_3COCD_3 , TMS): δ 5.41 (s, 3H : $-\text{CH}$), δ = 7.62–7.66 (m, 12H : Ar), 7.79–7.83 (m, 6H : Ar), 8.65 (s, 12H : Ar) ppm. Selected IR (KBr): ν = 1125 (P=O), 1150–1250 ($-\text{CF}$), 1650 ($-\text{C}=\text{O}$) cm^{-1} . Elemental analysis calculated (%) for $\text{C}_{51}\text{H}_{36}\text{EuF}_{18}\text{O}_8\text{P}_2$: C, 46.07, H, 2.50; found: C, 46.03, H, 2.51.

2.3. Preparation of PMMA copolymer beads with $\text{Eu}(\text{hfa})_3(\text{TPPO})_2$

$\text{Eu}(\text{hfa})_3(\text{TPPO})_2$ (0.3 g) was dissolved in methyl Methacrylate (MMA 100 g), styrene or trifluoromethylmethacrylate, 2,2'-azobisisobutyronitrile (AIBN, 0.3 g), and ethylene oxide dimethacrylate (10 g), at room temperature. The liquid solution was added to deionized water, stirring and reacting at 55 °C for 1 h and 95 °C for 3 h by suspension polymerization method. IR (KBr): ν = 1123 ($-\text{CF}$), 1130 (P=O), 1700 ($-\text{C}=\text{O}$) cm^{-1} .

2.4. Preparation of EVA films containing $\text{Eu}(\text{hfa})_3(\text{TPPO})_2$ -copolymer beads

Triarylisocyanate (0.5 g), t-butyl peroxy-2-ethylhexyl monocarbonate (0.5 g), silane coupling agent (0.3 g), and PMMA copolymer beads including Eu(III) complex (0.3 g) were dispersed with EVA(Ethylene Vinyl Acetate) resin (copolymer:ethylene:vinyl acetate contents = 74:26, 100 g), before kneading process. The dispersion containing with them was kneaded at temperature 75 °C, and

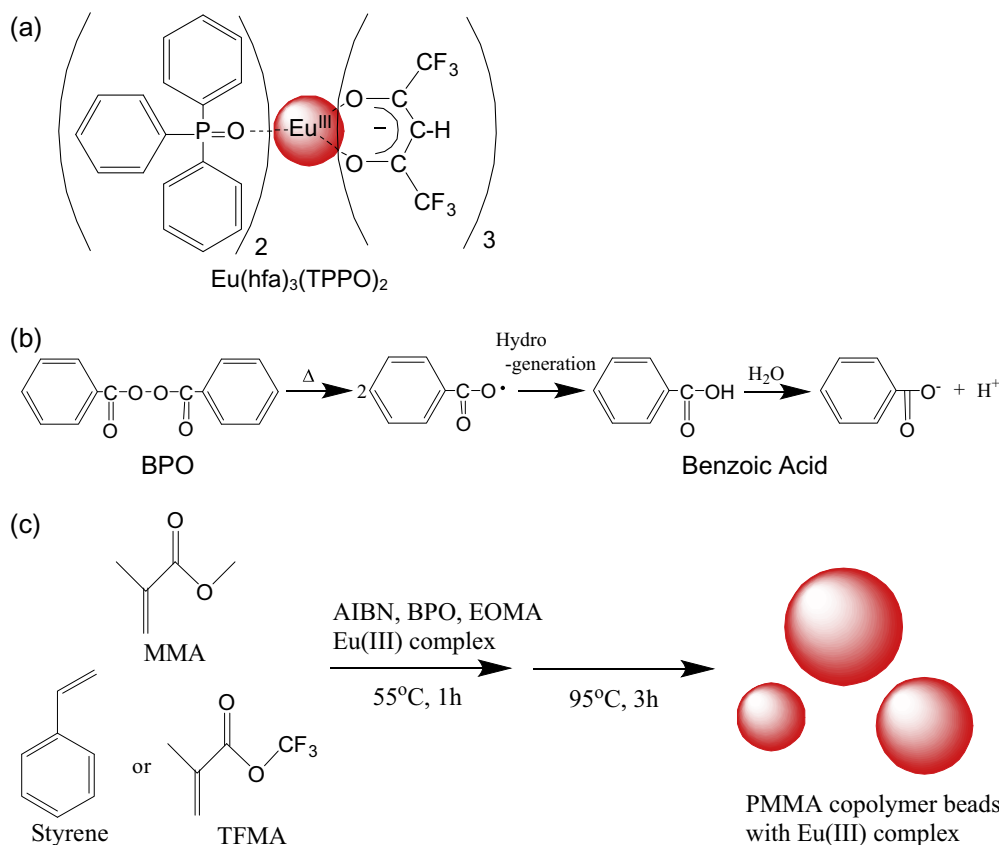


Fig. 1. (a) Chemical structure of $\text{Eu}(\text{hfa})_3(\text{TPPO})_2$. (b) Preparation scheme of PMMA copolymer beads containing Eu-complexes. (c) Chemical structure transition of BPO during PMMA copolymerization and after production.

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