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Review Article

Upconversion Particles' Role in Composite Materials: From Light Absorption to Energy Conversion

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Abstract

Upconversion materials are capable of emitting high-energy photons in the span of ultraviolet to NIR region of electromagnetic spectrum under excitation by low energy NIR photons through a process termed 'upconversion fluorescence'. The NIR electromagnetic wave has deeper penetration, lower thermal effect, and lower toxicity compared with UV/vis laser, providing UC materials with a wide range of applications. This review focuses on the research progress of the application of UC particles and their role in composite materials, which covers the field of radiation curing technology, anti-counterfeiting technology, photovoltaics, and photocatalysis.

Keywords: Upconversion particles; Light absorption; Energy conversion; Composite materials; Radiation curing; Anti-counterfeiting; Photovoltaics; Photocatalysis

Introduction

UC particles can emit high-energy photons after absorbing low-energy photons. It is a non-linear optical process of absorbing two or more low-energy NIR photons successively, exciting an electron to some intermediate state, and ultimately raising it to a high-energy level [1,2]. This unique characteristic provides a wide variety of applications for these materials in diverse areas, including radiation curing technologies [3-7], security anti-counterfeiting technologies [8-11], photovoltaics [12-14], and photocatalysis [15-17], optical thermometry [18,19], optoelectronic devices [20], drug delivery [21,22], and cellular imaging and biological labeling [23-26].

According to the literature, there are three types of UC materials: (i) Lanthanide-based UC materials, (ii) Transition metal-based UC materials, and (iii) molecular UC materials [27,28]. UCPs are inorganic materials comprised of a transparent host lattice doped with specific trivalent ions. Host material and dopant ions constitute two crucial components of a system exhibiting UC properties. The host material is the parent material, which accepts foreign elements, and that leads to the enhancement in optical properties of a system [29]. Like other classes of luminescence, the UC luminescence efficiency is closely related to host matrices. Upon doping with suitable inorganic elements, such as trivalent rare-earth ions (Y^{3+}, La^{3+}, Gd^{3+}, Sc^{3+}, Yb^{3+}, Tm^{3+}, Er^{3+}, Lu^{3+}), alkaline earth ions (Ca^{2+}, Sr^{2+}, Ba^{2+}), or certain transition metals (Zr^{4+}, Ti^{4+}), host forms a crystalline lattice that maintains the correct arrangement of the hybrid material [30]. Dopant ions play a central role in the UC mechanism by absorbing and emitting photons. Dopants determine the final color of the emitted light.

There are generally five mechanisms involved in the UC process, including Excited-State Absorption (ESA), Energy Transfer Upconversion (ETU), Cooperative Energy Transfer (CET), the Photon Avalanche Effect (PAE), and Energy Migration-Mediated Upconversion (EMU) [1,27]. Regarding the UC with complicated models, energy transfer dominates the UC luminescence processes. UC nanostructures that are the key factors determining luminescence efficiency contain dopants and host matrices. Dopants (sensitizers and activators) provide a luminescence center, while host matrices supply a platform for energy transfer between the dopants and drive them into optimal positions [31]. As the application of UC materials covers a wide range, this review's focus is merely on the recent research progress on the application of UC particles and their role in composite materials, which covers the field of photopolymerization technology, anti-counterfeiting technology, photovoltaics, and photocatalysis.

UCPs in radiation curing technology

Photopolymerization was rapidly developed due to its outstanding advantages such as lower consumption, environment-friendly, and faster reaction rates compared with thermal polymerization [32,33]. UV was the first source that was used for the photopolymerization technology. However, Photo-polymerization of thick materials still remains a significant challenge due to the light-intensity gradient and scattering of high-energy excitation radiation [34,35]. These features have made scientists look for a different light source with minimal quenching of the exciting radiation to enable the uniform fabrication of thick materials.

UCPs in radiation curing of coatings and composites: UC particles are currently being investigated as novel internal radiation sources for the radiation curing of coatings, inks, and composites as they enable deep and uniform polymerization through the composite. Several studies have shown that these internal radiation sources can increase the degree of double bond conversion (DC) that leads to the better curing of the composite [3,4,6,36]. In this sense, Jalili et al. [4] were able to achieve 41mm cure depth at around 80% DC with a uniform DC across the sample when employing $NaLuF_4$: $Yb_{(40\%)}$, $Tm_{(0.5\%)}$ in the presence of Irgacure 784 for the polymerization of Trimethylolpropane triacrylate (TMPTA) monomer. They have shown that by increasing the weight percentage of UC particles, the intensity of the UV-Vis radiation enhances, which results in the activation of more PIs that leads to an initial increase in DC up to 3% UC particles and then becomes constant, whereas the cure depth decreases.

An alternative approach for increasing curing depth in composite systems is using a combination of photoinitiators activated by radiations at various wavelengths. Darani et al. [3] demonstrated the ability of UCPs to be used in combination with UV/blue light to induce the dual cross-linking of coatings. The feasibility of dual curing process utilizing $NaYF_4$: $Yb_{(20\%)}$, $Tm_{(1.2\%)}$ (maximum absorption @ 345, 365, 450, 475nm) as a radiation source was carried out in presence of a combination of photo-initiators (benzophenone (maximum absorption @ 340nm) + camphorquinone (maximum absorption @ 470nm)). The results indicated the increment of DC from 24% to 30% and 27% to 37% for 3W and 6W laser intensity, respectively. These results indicate the great potential of UCPs as miniaturized UV/visible sources for the fabrication of thick 3D composites.

Three dimensional (3D) rapid prototyping: Radiation-based photopolymerization in 3D printing allows the fabrication of materials with high spatial and temporal resolution in a non-contact manner. The single-photon polymerization is usually activated by irradiation with ultraviolet (UV) or blue/green visible light, although without the repetition steps of adding of the subsequent polymer layers, it is mainly limited to the fabrication of two-dimensional (2D) structures [37]. This shortage comes from the fact that the intense light absorption by the photo initiator results in stable radical formation and immediate triggering of the polymerization process [38]. In addition, linear light absorption does not allow direct production of bulk 3D structures of consistent density due to the light intensity gradient of UV and visible light. Penetration depth is limited to a near-surface layer due to the exponential light attenuation of the depth of photopolymerization and 3D structure formation [39]. One of the strategies that can be used to overcome these challenges is shifting to a near-infrared (NIR) source as a photo-trigger to increase light penetration depth and achieve volumetric 3D printing through a two-photon polymerization process [40,41]. However, the expensive instruments (e.g., femtosecond lasers), high laser intensities, and time-consuming spot-by-spot curing process hinder the broad applications of this technology [35]. To overcome the problems associated with NIR light, recently, 3D rapid prototyping technology based on NIR light-induced polymerization of photo-curable compositions containing upconversion nanoparticles has been explored. Rocheva et al. [37] were able to design the core/shell upconversion nanoparticles $NaYF_4$:Yb^{3+}, Tm^{3+}@$NaYF_4$ with high NIR-UV conversion efficiency (η=2%). They were able to produce 3D structures inside the volume of a photo-curable composite with resolution at millimeter and sub-micron scales.

Recently, Yan et al. [42] synthesized $NaYF_4$:Yb^{3+}, Tm^{3+} upconversion microparticles with different fluorescence properties by varying temperature and the Tm^{3+} concentration (0.2-1.7%) for Stereolithography printing (SLA). They showed that the curing depth could reach 41mm, and the curing cross-section diameter could be as small as 0.22mm by incorporating UCPs in the composite system. They controled the curing depth and cross-section diameter by changing the Tm^{3+} concentration, hydrothermal temperature, and UCPs' concentration. Therefore, UCPs and their concentration can be reasonably selected according to the different requirements of the final structure.

In tissue engineering applications, NIR radiation with deep tissue penetration, low tissue absorption, weak thermal effect, and lower toxicity compared to UV light can be used in combination with UCPs for energy-converter-assisted photopolymerization [2,43,44]. These features, along with previous findings, suggest

that UCPs could be efficiently employed for the fabrication of thick 3D scaffolds and real-time 3D printing in vivo. However, further research on this field is still required.

UCPs in security anti-counterfeiting technology

Recent advances in chemical synthesis have enabled the precise incorporation of multiple dopant ions into a single substrate of well-controlled size and morphology, which render emissions with different spectroscopic fingerprints as well as spatially and temporally resolved features [45]. The remarkable tunability of upconversion luminescence holds great promise in anti-counterfeiting applications by designing high-capacity optical codes with high-level security [46]. Multilevel encoding is possible through multicolor, orthogonal, and temporal upconversion [47,48]. Roy et al. [49] employed a Ho^{3+}/Yb^{3+} co-doped $YTaO_4$ phosphor sample for security ink applications in the green region via 980nm excitations and in the blue region in the presence of Bi^{3+} via UV (365nm) excitation. Thus, the dual-mode emission of this phosphor is applicable in two different regions. This material also permits one to get intense tunable radiation from 350 to 650nm on excitation with 342nm. There are very few dye lasers, which emit intense tunable radiation in this wavelength region in such a wide wavelength range indicating the potential applicability of this material as a security ink. Recently, Li et al. [50] suggested that the Li^+ doped $NaGdF_4$: Yb^{3+}, Tm^{3+} can be used for screen printing material for anti-counterfeiting recognition. Ni et al. [51] employed $NaYF_4$: RE nanoparticles with red ($NaYb_{(99\%)}Er_{(1\%)}F_4$), green ($NaY_{(80\%)}Yb_{(19\%)}Er_{(1\%)}F_4$), and blue ($NaY_{(80\%)}Yb_{(15.5\%)}Tm_{(4.5\%)}F_4$) UC emission to produce 3D printed objects with optical anti-counterfeiting based on high concealment in daylight and multicolor UC emission under exciatation by a NIR laser at 980nm. They also showed that the 3D part with UC emission can be used for ratiometric temperature sensing from 303.15K to 463.15K, making it possible to map the temperature distribution for studying the thermal diffusion process in complex objects.

UCPs in photovoltaics and photocatalysis

Pollutant degradation: Lack of photocatalysts with strong sunlight-harvesting capacity is one of the challenges in the realization of pollutant degradation and water splitting [52]. The UCPs can be used to settle this issue due to their ability to absorb low-energy NIR radiation and emit higher energy irradiations. Wang et al. [53] coupled the Gd_2MoO_6: Er^{3+}, Yb^{3+} (GMEY) upconverting particle with the BiOI microplate to construct the BiOI@GMEY-x composites. GMEY particle converted near-infrared (NIR) light to green and red emissions, which was reabsorbed by the BiOI microplate, employing the complete region of visible-NIR light for photocatalysis. Compared with that of the BiOI microplate, enhanced photocatalytic activity was achieved in the resultant composites due to the effect of a built-in electric field and enhanced sunlight-harvesting ability. Wu et al. [16] proposed an efficient and eco-friendly method employing NaYF4:Yb, $Tm@TiO_2$ for the degradation of Deoxynivalenol (DON), one of the most common contaminants of cereal grains, for food safety and environmental protection. The results showed the DON degradation rate at 10μg/mL under simulated sunlight using NaYF4:Yb, $Tm@TiO_2$ (6mg/mL) approached 100% within 60 minutes.

Solar cells: The development of an efficient broadband photocatalyst to improve the use of solar energy has gained much attention in recent years [54]. In a recent study by Li et al. [54], a composite consisting of Sr_2LaF_7: Yb^{3+}, Er^{3+} UC nanocrystals and Bi nanoparticles loaded BiOBr nanosheets with oxygen vacancies (OVs, SLFBB) was fabricated. The Bi metal and OVs enhanced the sunlight-harvesting capacity in the Vis-NIR region and promoted the separation of electron-hole (e^-/h^+) pairs. Moreover, the surface plasmon resonance (SPR) effect of Bi metal enhanced the energy transfer from Sr_2LaF_7:Yb^{3+}, Er^{3+} to BiOBr via non-radiative energy transfer process, resulting in improving the light utilization from upconverting NIR into Visible light. Due to the synergistic effects of UC, SPR and OVs, the SFLBB exhibited enhanced photocatalytic ability for the degradation of BPA with a rate of $8.9\times10^{-3}min^{-1}$, 2.78 times higher than $3.2\times10^{-3}min^{-1}$ of BiOBr (BOB) under UV-Vis-NIR light irradiation. Table summarizes some of the research progress and their relative findings in the application of UC particles and their role in the above-mentioned areas.

Conclusions and Future Directions

The methods for synthesis and surface modification for UCNPs have been widely investigated. The application of UCNP-assisted photopolymerization is relatively mature in the area of coatings and composites. However, it is still in the early stage of research in the biological field. More attention must be paid to tissue damage and material toxicity in the field of tissue engineering applications. The UCNP-assisted photopolymerization in combination with safe NIR radiation has the potential to be applied to realize in vivo real-time 3D printing.

Anti-counterfeiting inks that are authenticated by general single security alarms, such as stimuli-chromism, fluorescence, or phosphorescence properties suffer from low security. The design of multilevel authentication systems by employing a combination of UCPs with multiple dopant ions in a single ink formulation could provide a practical solution for this issue.

The use of UC materials has improved the performance of solar cells. Despite recent progress, the increase of efficiency remains less than 2%. This discrepancy is ascribed to several problems of current photon UC materials. Design of hierarchical nanostructures to incorporate a range of rare-earth ions, without introducing deleterious cross relaxations, collectively can produce intense broadband UC.

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