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Metal-enhanced fluorescence of mixed coumarin dyes by silver and gold nanoparticles: Towards plasmonic thin-film luminescent solar concentrator

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ABSTRACT

Poly(methyl methacrylate) (PMMA) nanocomposite films doped with mixed coumarin dyestuffs and noble metal nanoparticles (60 nm silver and 100 nm gold) were prepared by spin coating technique. The effect of silver and gold nanoparticles on the film properties was studied by Fourier transform infrared spectroscopy (FT-IR), differential scanning calorimetry (DSC), transmission electron microscopy (TEM), scanning electron microscopy (SEM), UV–vis absorption and fluorescence spectroscopy measurements. DSC measurements indicated the increase of the glass transition temperature of the films by increasing nanogold concentration, recommending their promising thermal stability towards hot climates. It was found that the fluorescence signals of the mixed coumarin dyes were amplified by 5.4 and 7.15 folds as a result of metal enhanced fluorescence (MEF). The research outcomes offered a potential application of these films in solar energy conversion by plasmonic thin film luminescent solar concentrator (PTLSC).

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1. Introduction

1.1. Luminescent solar concentrators (LSCs)

Solar power is considered as a prime candidate for alternative energy sources since the annual average solar radiation in the world has its maximum value of about 7 kW h/m².day; in view of that the potential for solar photovoltaics is enormous [1]. Despite the fact that the photovoltaic (PV) technology is improving rapidly and the methods of manufacturing more efficient PV cells are being developed, but the industry is still being hampered by the cost [2]. The best approach to reduce the cost of PV power is basically to reduce the number of PV cells by concentrating sunlight; therefore, the price of a photovoltaic system can be considerably less important [3]. Common ways of concentrating photovoltaic (CPV) systems involve using lenses and mirrors which have a considerable heating effect on PV cells and consequently cooling systems are needed to avoid the decrease in the conversion efficiency as the temperature is increased [4]. Another disadvantage for traditional concentrators is the tracking system which add a significant cost to PV system [5]. An alternative method of concentrating light is the use of luminescent solar concentrators (LSCs) which made up of luminescent species doped

in a transparent substrate, usually glass or plastic, with photovoltaic cells placed around the edges [6–9]. Light photons enters the face of the substrate where it is absorbed by the luminescent molecule then fluoresces photon at a longer wavelength, which is waveguided via total internal reflection to the edge of the substrate where it can be absorbed by the photovoltaic cell [10–12]. The main advantage of LSC is the idea of covering the area with a luminescent transparent plate is far cheaper than the coverage with solar panels specially in building integrated PV systems [13]. Additionally, LSCs do not require sun tracking because they collect efficiently diffused light as well as the direct light [14]. A more aesthetically beneficial aspect of LSCs is that they can be easily incorporated into windows, thus using the light that is not absorbed by the dye to illuminate rooms indoors [15].

1.2. Thin film LSCs

Fig. 1 shows the construction and the operation principle of Thin film LSCs have several other advantages that they allow the absorption of the full solar spectrum by the stacking several luminescent layers which are containing different types of luminescent species [16]. In addition flexible employments of thin-film LSCs can be made on any commercial host substrate; making them of a great economic value [17]. The main drawback of thin film LSCs is the decrease of the absorption of solar radiation by small film thickness This problem could be solved using the plasmonic properties of noble (Au, Ag) metal nanoparticles (MNPs). MNPs are known to possess interesting properties called surface plasmon

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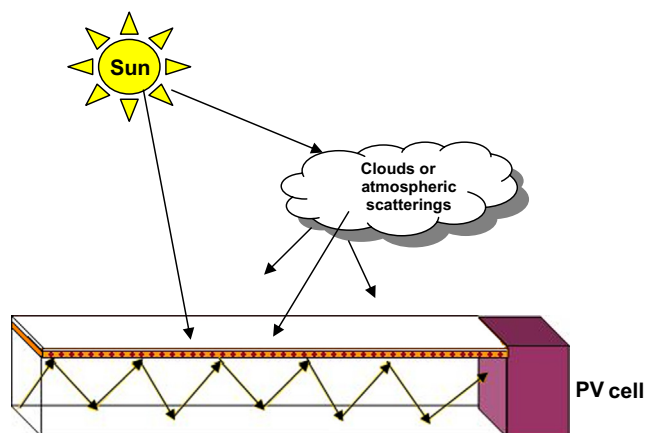


Fig. 1. Thin-film luminescent solar concentrator: Top layer antireflection coating, mid layer fluorescent film and bottom layer transparent plate.

resonance (SPR) bands in the visible region of the electromagnetic spectrum. This property arises due to the collective oscillations of the conduction band electrons induced by incident electromagnetic radiation. The frequency and shape of SPR bands are strongly dependent on the size and shape of metal nanoparticles (MNPs) as well as on the dielectric constant of the surrounding media [18]. The ability of MNPs to amplify fluorescence signals and increase a fluorophore's quantum yield has brought about important changes to fluorescence spectroscopy as well as to its applications [19]. It was found that the maximum amplification occurred when the wavelength of SPR of metal nanoparticle coincides with that of the fluorescence band of the dye molecule. The metal–fluorophore interactions has flourished as it was found that the optical properties of these fluorophores could be modulated by plasmonic interactions [20].

This paper aims to develop metal-enhanced fluorescence (MEF) by SPR between stabilized Ag-NPs, Au-NPs and mixed coumarin dye molecules doped in PMMA films. There are many potential advantages of using Ag-NPs and Au-NPs for MEF because of their chemical and thermal stability. All these information will finally be used to discuss and demonstrate the aspects of a promising implication, regarding the application of the prepared films in the manufacturing of PTLSCs.

2. Experimental techniques

2.1. Materials

PMMA was obtained from (Aldrich, USA). Coumarin dyestuffs MACROLEX Fluorescent Red G and MACROLEX Fluorescent Yellow 10GN were obtained from (Bayer, Germany). Spherical gold and silver nanoparticles Ag-NPs and Au-NPs with average particle diameters 60 nm and 100 nm were obtained from Aldrich (USA). HPLC grade Dichloromethane (CH_2Cl_2) was obtained from Aldrich (USA) and used as high purity solvent for PMMA grains.

2.2. Fluorescent PMMA films

PMMA grains and coumarin dyestuffs were dissolved in dichloromethane (CH_2Cl_2) and mixed by a magnetic stirrer at 40 °C for 6 h. The polymer solution was doped by different concentrations (ppm) of Coumarin dyestuffs MACROLEX Fluorescent Red G and MACROLEX Fluorescent Yellow 10GN. All the dye doped polymer solutions were sonicated for 6 h before pouring on glass substrates and spin coated in a centrifuge at 2000 rpm for 1 min to obtain uniform film coverage [18], then they are left to

dry in an electric oven at 40 °C for 6 h. The film thicknesses were measured using a profilometer (Talystep, Taylor Hobson, UK) on a scratch made immediately after deposition of five independent measurements on each sample, and found to be in the range of $50 \pm 10 \mu\text{m}$ [21].

2.3. Metal enhanced fluorescent PMMA films

The optimized fluorescent PMMA film was determined by fluorescence measurements and found to have dye concentrations of 70 ppm MACROLEX Fluorescent Yellow 10GN and 30 ppm MACROLEX Fluorescent Red G, respectively. The fluorescence spectra of optimized fluorescent PMMA film was enhanced by spherical Ag-NPs and Au-NPs doped as received with different concentrations (ppm) using the same procedure described before.

2.4. Characterization and measurements

FT-IR spectra for all samples were recorded in the wave number range ($4000\text{--}400 \text{ cm}^{-1}$) with an FT-IR spectrophotometer (Genesis Series, USA). The nanoparticle distribution was examined by the transmission electron microscope (TEM), (JEOL JEM-1400, Japan) and scanning Electron Microscope (SEM) (JEOL, JSM-5400, Japan). The absorption spectra were recorded in the wavelength range (190–900 nm) using a UV–vis spectrophotometer (UNICAM, Helios Co., Germany). The steady-state fluorescence spectra were recorded in the wavelength range (400–800 nm) using a spectrofluorimeter (Perkin Elmer LS 50 B, UK).

3. Results and discussion

3.1. Material characterization

Fourier transform infrared spectroscopy (FT-IR) was used in order to know some information about the interactions between the vibrational energy states of Ag-NPs, Au-NPs, coumarin and PMMA molecules. Fig. 2 shows the FT-IR transmission spectra in the wavenumber range ($4000\text{--}400 \text{ cm}^{-1}$), before and after doping with 20 ppm Ag-NPs and Au-NPs. It is noted that all the spectra neat and doped PMMA films are almost identical without noticeable shifts in the positions of the vibrational peaks of the characteristic groups [22]. The behavior observed in the spectra can be attributed to the fact that the nanoparticle concentrations are too small to make any observable effects on the chemical bonds of PMMA, reflecting the chemical stability of the PTLSC films since all the physical properties of PMMA films can be retained after doping with noble metal nanoparticles.

Fig. 3 shows TEM images for Ag-NPs and Au-NPs used as received, it is clear that all the nanoparticles are spherical and close to each other with the affinity to form aggregates. Therefore the nanoparticles were sonicated for enough time in the polymer solution in order to obtain a homogeneous coating to prepare PTLSC waveguides. The optimal sonication time for Ag-NPs and Au-NPs suspended in the dye doped PMMA/ CH_2Cl_2 solution was found to be 6 h determined by SEM photographs shown in Fig. 4. It is clearly noticed that all Ag-NPs and Au-NPs exhibit regular smooth spherical shape with a slight change in the mean diameter of the nanospheres, this convinced that the sonication process is important to reduce the probability of cluster formation [10].

3.2. Spectroscopic properties of PMMA films doped with mixed coumarin dyestuffs

Fig. 5 shows the absorption spectra of MACROLEX dyes before and after being mixed in PMMA film with concentration 50 ppm of

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