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Simulation-Based Investigation of Tunable Luminescence and Optical Behavior of Polycarbazole-Based Metal Oxide Nanocomposites SnO_2 and TiO_2

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This study explores the optical properties of Polycarbazole (PCz) polymer nanocomposites incorporating tin oxide (SnO_2) and titanium oxide (TiO_2) nanofillers. The influence of nanofiller content on the optical characteristics, including absorption and photoluminescence (PL) behavior, was systematically examined. UV-Vis absorption spectra show that the absorption edge of the nanocomposites shifts to longer wavelengths (red shift) as the concentration of SnO_2 and TiO_2 increases. This shift is attributed to the formation of defect-related energy states and changes in the polymer's conjugation length due to interactions between the polymer chains and nanofillers. Tauc plot analysis was used to estimate the direct and indirect optical band gaps of the nanocomposites. It was observed that both direct and indirect band gaps decrease with increasing nanofiller content. For PCz/SnO_2, the direct band gap reduces from 3.93 eV (pure PCz) to 3.84 eV with 20% SnO_2, while for PCz/TiO_2, the direct band gap decreases from 3.93 eV to 3.81 eV. These decreases are attributed to the modification of the polymer's electronic structure and the creation of defect-related energy levels within the band gap. Photoluminescence spectra reveal a shift in emission peaks to longer wavelengths and a decrease in fluorescence intensity, indicating quenching effects due to the strong interaction between the polymer and the nanofiller. Fluorescence decay measurements show a decrease in carrier lifetime with increasing nanofiller concentration, supporting the hypothesis of enhanced charge transfer and dynamic quenching. These findings highlight the potential of PCz/SnO_2 and PCz/TiO_2 nanocomposites for applications in optoelectronics and photonics.

Keywords: Polycarbazole, SnO_2 nanocomposites, TiO_2 nanocomposites, optical band gap, photoluminescence quenching

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In recent years, fluorescent polymers alongside metal oxide infused polymer nanocomposites have garnered considerable scholarly attention due to their remarkable photoluminescent characteristics and extensive range of applications. The optical characteristics of polymer nanocomposites are influenced by the attributes of nanofillers, including dimensions, form, and structure, as well as the uniform dispersion and molecular interactions between the nanofillers and the polymer chains. A vast array of literature exists regarding the optical characteristics of fluorescent polymers and metal oxide-infused polymer nanocomposites utilising various polymers as host materials. The influence of nanofiller dimensions on the optical characteristics of PANI/SnO2 nanocomposites is examined [1]. Their observations revealed a red shift in the absorption cutoff when contrasted with SnO_2 nanoparticles, attributed to the interaction between the polymer and the nanoparticles. Explored the impact of SnO2 nanofiller levels on the optical and electrical characteristics of (PVA/PEO)/SnO_2 nanocomposites [2]. Researchers have established both the direct and indirect bandgap of polymer nanocomposites, discovering a red shift in the bandgap as the concentration of SnO_2 nanofiller increases, attributed to the creation of defect-related energy states. The influence of nanofiller concentration on the optical characteristics of PVC/SnO2 nanocomposites [3]. They have assessed both the direct and indirect bandgap of nanocomposites using the Tauc relation and noted a reduction in both direct and indirect bandgaps of nanocomposites as the content of SnO2 nanofiller increases.

The influence of varying weight ratios of PANI:TiO2 on the optical absorption characteristics of PANI/TiO_2 nanocomposites [4]. Their observations

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indicated that the absorption peak near 650nm transitioned to a longer wavelength at 690nm as the weight proportion of PANI and TiO_2 nanofiller increased. The optical characteristics, both linear and nonlinear, of PSMA/TiO_2 polymer nanocomposites are examined [5]. Observations revealed a blue shift in the linear absorption edge of PSMA/TiO_2 polymer nanocomposites with the rising concentration of nanofiller. The optical and photoelectric characteristics of PMOT/TiO2 polymer nanocomposites, noting a red shift in the absorption cutoff of these nanocomposites as the TiO2 content increases [6]. The impact of varying nanofiller concentrations (SiO2 and TiO2) on the optical and electrical properties of PPV/SiO2 and PPV/TiO2 nanocomposites. They observed a movement of the absorption peak towards shorter wavelengths as the concentration of the nanofiller rises, attributed to alterations in the conjugation of the PPV chain [7]. The enhancement of optical characteristics in PMMA/TiO2 nanocomposites with varying amounts of TiO2 nanofiller and observed a blue shift in the absorption edge of the nanocomposites as the weight percentage of TiO_2 nanofiller increases [8].

Polycarbazole (PCz) is a good fluorescent polymer with high refractive index. Moreover, it has relatively high electrical conductivity with good hole mobility and unique photochemical behaviour as compared to another fluorescent polymer. A very less investigations are done on optical properties of polycarbazole [9]. Further, SnO_2 and TiO_2 are most versatile filler materials because of their high optical transparency and good photo stability properties. Hence it is worth to investigate the optical properties of polycarbazole and metal oxide (SnO2 and TiO2) embedded PCz nanocomposites. Therefore, in this work we have studied the effect of metal oxide (SnO2 and TiO2) nanofiller concentration on optical properties of PCz nanocomposites [10]. As mentioned in earlier chapters, for PCz/SnO2 polymer nanocomposites, we denoted pure PCz as PS0 and that incorporated with 5%, 10%, 15% and 20% of SnO2 as PS1, PS2, PS3 and PS4, respectively. For PCz/TiO2 polymer nanocomposites, we refer pure PCz as PT0 and that embedded with different weight percentage 5%, 10%, 15% and 20% of TiO2 as PT1, PT2, PT3 and PT4, respectively [11].

METHODS AND MATERIALS

Polycarbazole (PCz) and metal oxide nanofillers, SnO_2 and TiO_2, were used to prepare nanocomposites. The preparation of these nanocomposites and the characterization of their optical properties were carried out using standard procedures, as detailed below.

Preparation of Nanocomposites

Polycarbazole (PCz) was synthesized by a chemical oxidative polymerization method using carbazole as the monomer and ammonium persulfate (APS) as the oxidizing agent, following the procedure described [12]. The synthesized polycarbazole was purified by washing with methanol and then dried under vacuum. The nanocomposites were prepared by mixing the desired concentrations of SnO_2 and TiO_2 (5%, 10%, 15%, and 20%) with the PCz polymer. SnO_2 nanoparticles and TiO_2 nanoparticles were obtained from commercial sources and were incorporated into the polymer matrix via solution processing [13]. The nanofiller concentrations were chosen based on previous studies [12-13] to explore their effect on the optical properties of the polymer nanocomposites.

Characterization Techniques

To investigate the optical properties of the nanocomposites, UV-Vis absorption spectra were recorded using a UV-Vis spectrophotometer (UV-1800, Shimadzu). The absorption measurements were conducted in the range of 200–800 nm, and the absorption edge was determined to observe the shift in absorption characteristics with increasing nanofiller concentration [14].

Photoluminescence (PL) spectra were obtained using a fluorescence spectrometer (Horiba Scientific, Fluoromax-4). The samples were excited at 300 nm, and the emission spectra were recorded in the wavelength range of 320–550 nm [15]. The PL spectra were used to observe the emission peaks and to analyze the quenching effects due to the interaction between the polymer and nanofillers, as documented [16].

To evaluate the fluorescence lifetime of the samples, time-resolved photoluminescence (TRPL) measurements were performed using a time-correlated single-photon counting (TCSPC) setup. The decay profiles were fitted using a tri-exponential function to estimate the carrier lifetime, following methods used [17].

Tauc Plot Analysis

The optical band gaps of the polymer nanocomposites were estimated using the Tauc relation, which relates the absorption coefficient (α) to photon energy (hv). The Tauc plots for both direct and indirect transitions were constructed from the UV-Vis absorption data. This approach, commonly used for semiconductors was adapted for the polymer matrix to estimate the optical band gaps [18].

Table 1. Calculated values of direct and indirect energy gap of PCz/SnO2 polymer nanocomposites.

Sample ID	Direct band gap (eV)	Indirect band gap (eV)
PS_0	3.93	3.65
PS_1	3.90	3.59
PS_2	3.88	3.55
PS_3	3.86	3.48
PS_4	3.84	3.43

RESULTS AND DISCUSSION

Polycarbazole – Tin Oxide (PCz/SnO_2) Nanocomposites

To know the variation of absorption cut off and formation of defect related energy states within band gap of polymer nanocomposites, we have made UV-Vis absorption measurements of PCz/SnO_2 polymer nanocomposites. Figure 1 illustrates absorption curves of PCz/SnO_2 nanocomposites. Curves a-e respectively, represent absorption intensity for the samples PS_0, PS_1, PS_2, PS_3, and PS_5. The curve (f) represents the absorption intensity of SnO_2 nanoparticles having absorption edge at 298nm. Pure polycarbazole (curve-a) exhibits the absorption cut off wavelength at 293nm due to the bonding – antibonding (π – π*) transition of the benzenoid ring and small hump around at 347nm corresponds to the polaronic energy level and conduction band (n-π*) transition of the quinoid ring. The polaronic energy levels arise due to the misalignment/ branching of polymer chain and creation of defects at the time of polymerization. From absorption curves of samples PS_1, PS_2, PS_3, and PS_5. (Curve: b-e), it is noticed that the absorption peak is red-shifted with an increase in weight percentage of SnO_2 nanofiller. This may be because of the strong interaction between NH – group of polycarbazole and oxygen atom of SnO_2. To have insight into the optical band gap, the energy gap of polycarbazole and its nanocomposites was estimated by using Tauc formula. It may be noted that Tauc relation is used for semiconductors and as a first approximation Tauc relation may be used for some longchained polymers [13]. In figures 2(a-e) and 3(a-e) respectively, we show the direct and indirect band Tauc plots for the samples PS_0, PS_1, PS_2, PS_3, PS_5. The obtained values are tabulated in Table 1. The obtained values of direct and indirect optical band gap of polymer nanocomposites decreases with an increasing content of SnO_2 nanofiller because of homogeneous distribution of nanofiller into polymer matrix and strong inter molecular interaction between nanofiller with polymer host, affecting electronic structure of polymer and also may be due to the formation of defect related states of SnO_2 nanofiller. The plot $(\alpha h\nu)^2$ versus hν is a straight line suggesting the direct band gap re of PCz/SnO_2 polymer nanocomposites [11]. her, the obtained values of energy gap are in good agreement with reported values of direct band gap of PCz. Hence it may be concluded that PCz/SnO2 possesses direct band gap nature.

To gauge the inter and intra gap defect state related emissions of polymer nanocomposites, we carried out the photoluminescence studies of synthesized polymer nanocomposites. The FL spectra of PCz/SnO_2 polymer nanocomposites (PS0, PS1, PS2, PS3, and PS4) recorded in 320 – 550 nm wavelength range keeping an optimized excitation wavelength at 300nm are shown. The sample PS0 shows the emission peak at 410nm and that of samples PS1, PS2, PS3, and PS4 show the peak at 413 nm, 415 nm, 418 nm, and 422 nm, respectively [7]. These emission peaks are due to the S2-S0 transition of carbazole moiety. We also notice that the shift of emission peak of polymer nanocomposites towards longer wavelength as compared to pure PCz with an increasing the content of TiO_2 nanofiller because of the stronger interaction of NH–group of polycarbazole with oxygen atom of SnO_2 and formation of defect related energy levels inside the band gap [17]. Moreover, it is also observed that emission intensity decreases with an increasing weight percentage of SnO_2 nanofiller because of quenching effect. The FL quenching and red shift of emission suggest that PCz has strong π-π* interaction with SnO2 nanofiller. This interaction between polymer and nanofiller and also absorption of defect related energy levels causes the decrease in fluorescence intensity of PCz/SnO_2 nanocomposites. Similar behaviour was noticed in PL studies of PANI/SnO_2 nanocomposites.

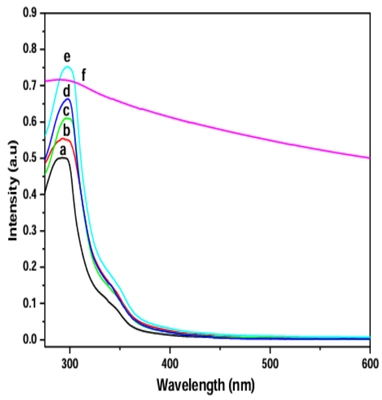

Figure 1. UV-Vis absorption spectra of PCz/SnO_2 nanocomposites. Curves (a-e) represent, absorption intensity for the sample PS0, PS1, PS2, PS3, and PS4, respectively. The curve (f) represents absorption spectrum of SnO_2 nanoparticles.

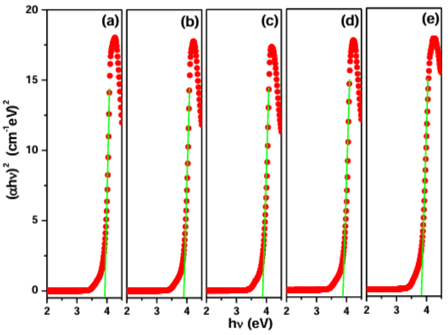

Figure 2. Tauc plots of PCz/SnO2 nanocomposites for direct band transition. Graphs (a-e) respectively, represent Tauc plot for samples PS0, PS1, PS2, PS3, and PS5.

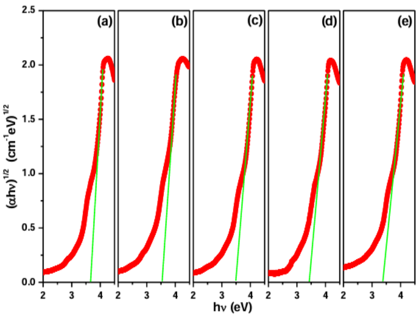

Figure 3. Tauc plots of PCz/SnO_2 polymer nanocomposites for indirect band transition. Graphs (a-e) respectively, represent Tauc plot for samples PS0, PS1, PS2, PS3, and PS5.

Table 2. Value of fitted constants and average life time of PCz/SnO2 samples.

Sample ID	τ_1 (ns)	τ_2 (ns)	τ_3 (ns)	A_1	A2	A_3	τ_{ave} (ns)	χ^2
PS$_0$	5.47	0.610	0.185	0.6676	0.2669	0.0655	5.17	1.03
PS$_1$	5.06	0.615	0.144	0.6531	0.2879	0.059	3.83	1.08
PS$_2$	3.76	0.437	0.179	0.6710	0.3804	0.0514	3.54	1.07
PS$_3$	3.40	0.346	0.145	0.6090	0.3910	0.0231	3.20	1.15
PS$_4$	3.38	0.494	0.158	0.5466	0.5104	0.0560	2.87	1.07

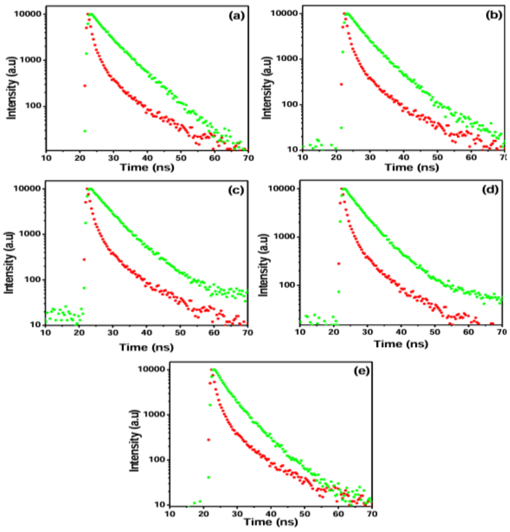

Figure 4. Fluorescence decay profile for the samples PS$_0$, PS$_1$, PS$_2$, PS$_3$, and PS$_4$ respectively.

The effect of nanofiller concentration on carrier life time and trapping of charge carriers in intermediate defect states of polymer nanocomposites is investigated through time resolved spectroscopy. Fluorescence decay profiles for PS0, PS1, PS2, PS3, and PS4 samples are shown in figure 4. The decay profile of all samples is well fitted by tri-exponential function with goodness of $\chi2$ very close to unity. The presence of two or more life times can be described by absorption and misalignment of polymer on the surface of nanofiller [8]. The fast decay is associated with content of nanofiller, which results of charge transfer mechanisms and dynamic quenching. The slow decay component is corresponding to the branching/misalignment of polymer chain during polymerization. Using fitted constants, we have estimated the average carrier life time of samples using equation 5.5. The value of fitted parameters and average carrier life time are tabulated in Table 1 [5]. We find that the average carrier lifetime of PCz/SnO2 nanocomposites decreases with an increasing weight percentage of SnO2 nanofiller. This may be because of charge transfer mechanisms and strong interaction between polymer matrix and nanofiller and absorption of defect related energy levels causing the decrease in carrier life time of polymer nanocomposites.

Polycarbazole - Titanium oxide (PCz/TiO2) Polymer Nanocomposites

TiO2 is another interesting filler material for PCz because of its unique properties. The change in absorption edge and conjugation length of PCz with TiO2 nanofiller content is studied through UV absorption characterization. The blue/red shift in absorption edge of polymer nanocomposites is dependent on unique properties of filler materials. In figure 5, curves (a-e) represent the absorption intensity for PT0, PT1, PT2, PT3, and PT4 samples, respectively [2]. Curve (f) shows the absorption spectrum of TiO2 nanoparticles with maximum absorption peak at 300 nm. From absorption curve of pure PCz sample, PT0 (curve-a), the absorption edge at 293 nm is due to bonding – antibonding ($\pi - \pi^*$) transition of the benzenoid ring and weak shoulder at 350 nm due to the polaronic energy level (n-π^*) transition of the quinoid ring. The creation of polaronic energy levels is due to the misalignment/branching of polymer chain at interface of polymerization medium. From the absorption curves of the samples PT1, PT2, PT3, and PT4(curve b-e), we notice the shift in absorption edge towards the longer wavelength as compared to the pristine polycarbazole [11]. This suggest that the variation of conjugation length of polymer and strong intermolecular interaction between polycarbazole and TiO2 nanofiller. However, there is no clear peak shift for peak around at 350nm because of broad nature of peak. Using the Tauc relation we have estimated the optical energy gap of PCz and its nanocomposites. In figures 6(a-e) and 7(a-e) we show the direct and indirect transition Tauc plots for the samples PT0, PT1, PT2, PT3, and PT4, respectively [6]. The obtained values are tabulated in Table 3. The evaluated values of direct band gap of PCz/ TiO2 decreases from 3.93eV to 3.81eV and that of indirect band varies from 3.65eV to 3.43 eV with an increase in weight percentage of TiO2 nanofiller. This decrease in the value of band gap is because of change in the effective conjugation length of polymer, strong interaction between polycarbazole chain and oxygen atom of TiO2 nanoparticles and formation of defect related energy levels. Further, the calculated values of Eg are similar to those of previously reported values of other TiO2 polymer nanocomposites [18].

Table 3. Estimated values of direct and indirect band gap of PCz/TiO_2 polymer nanocomposites.

Samples	Direct band gap (eV)	Indirect band gap (eV)
PT_0	3.93	3.65
PT_1	3.89	3.59
PT_2	3.87	3.55
PT_3	3.84	3.48
PT_4	3.81	3.43

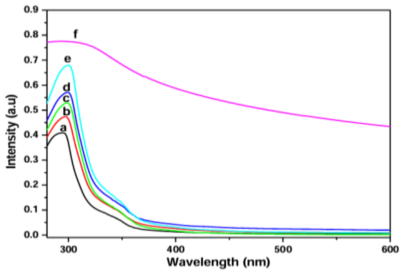

Figure 5. UV-Vis spectra of PCz/TiO_2 polymer nanocomposites. Curve (a-e) represent the absorption spectrum for samples PT_0, PT_1, PT_2, PT_3, and PT_4 respectively. The curve (f) indicates the absorption spectrum of TiO_2 nanoparticles.

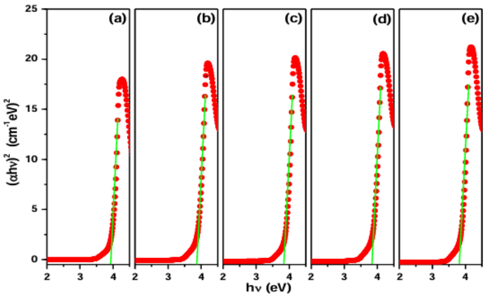

Figure 6. Tauc plots of PCz/TiO_2 polymer nanocomposites for direct band transition. Plots (a-e) respectively, represent Tauc plot for the samples PT_0, PT_1, PT_2, PT_3, and PT_5..

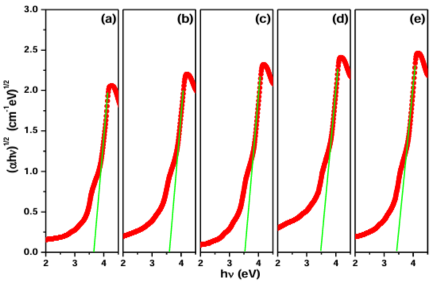

Figure 7. Tauc plots of PCz/TiO_2 polymer nanocomposites for indirect band transition. Graphs (a-e) respectively, represent Tauc plot for the samples PT_0, PT_1, PT_2, PT_3, and PT_4 .

The formation of non-fluorescent complex and misalignment of polymer chain on surface of nanofiller will affect the FL intensity of samples. To know about the defect related emissions and variation of emission peak and intensity of polymer nanocomposites with filler material, we have performed photoluminescence studies of PCz/TiO_2 polymer nanocomposites with varying content of TiO_2 nanofiller [4]. In figure 8, curves (a-e) respectively, depict the fluorescence emission spectra for the samples PT$_0$, PT$_1$, PT$_2$, PT$_3$, and PT$_4$ recorded in 350nm – 500nm wavelength range and the excitation wavelength is kept at 300nm.The emission peak for the sample PT$_0$ is found to be at 410 nm and that of samples PT$_1$, PT$_2$, PT$_3$, and PT$_4$ are 414nm, 417nm, 422nm, and 425nm, respectively due to the S$_2$-S$_0$ transition of carbazole moiety [16]. Furthermore, it is noticed the shift of emission peak of polymer nanocomposites towards higher wavelength and the decrease in emission intensity with an increasing the TiO_2 content is because of the strong molecular interaction between the nanofiller and polymer matrix and absorption by intermediate energy levels related to defects due to the misalignment/branching of polymer chain at time of polymerization [12]. The small hump observed around at 470nm for samples PT$_2$, PT$_3$, and PT$_4$ is due to defects such as oxygen vacancies of nanoparticles at higher concentration of nanofiller. The major surface defects such as oxygen vacancies and impurities (Fe and Cl) are noticed from the EDX spectra. Similar behavior is observed by some other researchers for different polymer nanocomposites [14].

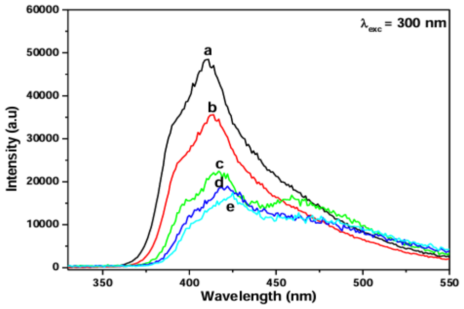

Figure 8. Fluorescence (FL) emission spectra of PCz/TiO_2 polymer nanocomposites. Curves (a-e) respectively, represent the FL intensity of the samples PT$_0$, PT$_1$, PT$_2$, PT$_3$, and PT$_5$.

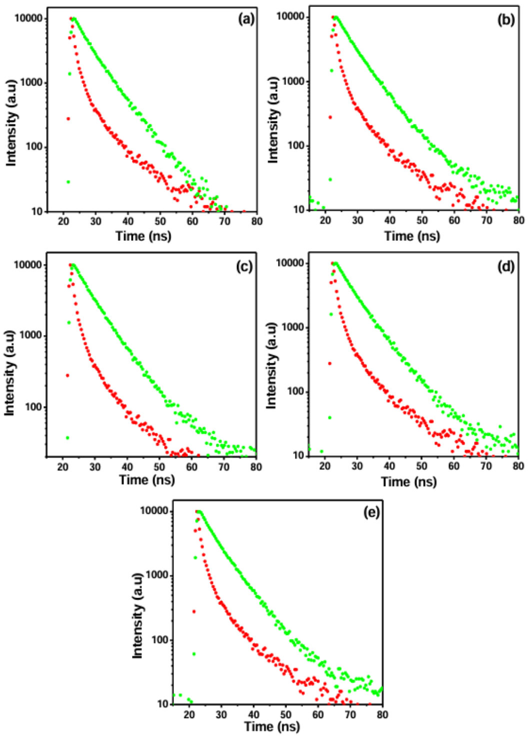

Figure 9(a-e). Fluorescence decay curves for the samples PT$_0$, PT$_1$, PT$_2$, PT$_3$, and PT$_5$.

Table 4. Value of fitted constants and average life time of PCz/TiO_2 samples.

Sample ID	τ_1 (ns)	τ_2 (ns)	τ_3 (ns)	A_1	A2	A_3	τ_{ave} (ns)	χ^2
PT_0	5.47	0.610	0.185	0.6676	0.2669	0.0655	5.17	1.03
PT_1	5.35	0.669	0.119	0.5779	0.3366	0.0885	5.03	1.09
PT_2	5.27	0.626	0.136	0.5544	0.3662	0.0794	3.93	1.12
PT_3	5.11	0.573	0.149	0.5493	0.3648	0.0859	3.80	1.15
PT_4	3.99	0.668	0.128	0.5416	0.3607	0.0977	3.63	1.07

To study the carrier dynamics and the variation of carrier life time with nanofiller content, we have made the life time measurements of synthesized PCz/TiO2 polymer nanocomposites. Fluorescence decay profiles of PCz/TiO2 polymer nanocomposites (PT0, PT1, PT2, PT3, and PT4) are depicted in figure 9. The emission decay curves of samples were fitted by tri-exponential function and confirmed with the goodness of $\chi 2$ very close to unity. The various decay components of polymer nanocomposites can be described by absorption of energy states related to the defects and misalignment/branching of polymer chain. The higher value of $\tau 1$ in comparison with $\tau 2$ is a result of misalignment of polymer chain on surface of nanofiller and formation of polaronic energy states [5]. By making use of fitted constant and equation 5.5, we have calculated the average carrier life time of polymer nanocomposites. The value of fitted components and average carrier life time are listed in Table 4. It is clearly noticed that the average carrier life time of polymer nanocomposites decreases from 5.17 ns to 3.63 ns with an increasing content of TiO2 nanofiller due to the charge transfer mechanisms, strong interaction between nanofiller and polymer chain and change in conjugation length of polymer chain [1].

CONCLUSION

From both polymer nanocomposites (PCz/SnO_2 and PCz/TiO_2) we notice that the variation of absorption and emission wavelength and intensity with an increase in nanofiller concentration. This may due to the formation of new energy levels within band gap and the change in conjugation length of polymer upon addition of nanofiller. The FL intensity significantly decreases with nanofiller content, which can be attributed due to the misalignment of polymer chain at interface and the formation non-fluorescent complex between nanofiller and polymer matrix and is also in well agreement with FL carrier lifetime measurements. The formation of defect related new energy states inside the optical band gap causes red shift in absorption onset of polymer nanocomposites. This systematic change in optical behavior of synthesized polymer nanocomposites with nanofiller concentration, makes both PCz/SnO_2 and PCz/TiO_2 a suitable material for application in optoelectronic and photonic systems. In the next chapter we give Forster resonance energy transfer mechanism between TiO2 nanoparticles and coumarin dye.

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