

Influence of donor-donor transport on excitation energy transfer in binary system C343-Rh6G doped in Poly(methylmethacrylate)

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Abstract: Energy transfer and migration from Coumarin343 to Rhodamine6G in Poly(methylmethacrylate) have been investigated using steady-state and time domain fluorescence spectroscopy. It is found that the influence of energy migration on energy transfer can be described quite convincingly on the basis of theories of Loring, Anderson and Fayer(LAF) and Huber. At higher acceptor concentrations direct donor-acceptor transfer occurs through Förster mechanism. It is suggested to use a mixture of these two dyes in the LSCs.

Keywords: Energy transfer, Energy migration, luminescent solar concentrators

I. Introduction

Long range Förster energy transfer is a well known process responsible for the transfer of excitation energy from donor to acceptor molecules[1]. It is a useful means for enhancing the performance of light emitting diodes/OLEDs[2], solar cells[3] and polymer lasers[4]. However, the short range energy migration between the donor molecules themselves may modulate the former and the resultant rate of energy transfer is due to the contribution of both the processes active simultaneously. Time dependent studies of donor emission in the presence of different concentrations of the acceptor enable to provide the insight into the Förster energy transfer and migration rates. With these studies it is possible to explain the energy transfer characteristics between several donor- acceptor pairs for a clear understanding of the energy transfer and migration processes [5-21]. The donor –acceptor interaction strength (α) and donor –donor interaction(β) can be determined separately. Depending upon the values of α and β two models viz. (i) the Yakota Tanimoto diffusion model [6] for $\alpha \gg \beta$ and Burshtein model for $\alpha \ll \beta$ [7,8] are applied to explain the overall energy transfer mechanisms. The former is applicable in the case of slow diffusion. In this case the Förster long range dipole-dipole energy transfer takes place explicitly and the decay follows a non exponential behavior. In the Burshtein hopping model short range energy transfer occurs from one donor to another neighboring donor and this continues till the energy is finally transferred to the acceptor molecules. In this case the decay is single exponential. Gochanour, Anderson and Fayer (GAF) [14-15] developed a model for excitation transport in a one component system. Later on Loring, Anderson and Fayer (LAF) [16] extended the GAF technique to the general problem of electronic excitation in a two component disorder system and applied it to the case of dipole -dipole interaction. Huber[9] proposed an approximate theory based on the coherent potential approximation for a special case of comparable donor –donor and donor – acceptor interactions ($\alpha \approx \beta$) for low as well as high concentrations[10-11]. Jang et al[21] proposed that by considering the constant distance between donor and acceptor, modification in a fast excitation migration limit could be introduced.

Energy transfer studies are important because of their applications in luminescent solar collectors (LSC). These collectors have been found to be cost effective and possess high efficiency [22-24]. The aim of these studies is to find suitable donor acceptor pairs to utilize the solar radiations in the UV- visible to augment the intensity in the infrared region by means of energy transfer. This is done by doping high quantum yield luminescent dyes absorbing in the UV- visible range of the solar spectrum and emitting in the infrared. This is achieved by using two or multiple dyes doped in thin polymer films used in solar concentrators. In the present work we have studied excitation energy transfer and migration in a new donor- acceptor pair of Coumarin343 and Rhodamine6G dyes doped in transparent thin films of Poly methyl methacrylate (PMMA). The aim of the study is to increase the Rhodamine6G (used In LSCs) emission by energy transfer from Coumarin343. Both the dyes have high absorptivity as well as high quantum yield of emission. The host material PMMA has been chosen for its excellent properties for use in luminescent solar collectors (highly transparent and large refractive index)[25].

II. Theory

The overlap integral $J(\lambda)$ expresses the degree of spectral overlap between the donor emission and the acceptor absorption,

$$J(\lambda) = \frac{\int_0^{\infty} F_D(\lambda) \epsilon_A(\lambda) \lambda^4 d\lambda}{\int_0^{\infty} F_D(\lambda) d\lambda} \quad (1)$$

where $F_D(\lambda)$ is the corrected fluorescence intensity of the donor in the wavelength range λ to $\lambda + \Delta\lambda$ with the total intensity (area under the curve) normalized to unity. $\epsilon_A(\lambda)$ is the extinction coefficient of the acceptor at λ expressed in units of $M^{-1}cm^{-1}$ and λ is in nanometer, then $J(\lambda)$ is in units of $M^{-1}cm^{-1}nm^4$.

The Förster distance R_{0A} is expressed as

$$R_{0A} = 9.78 \times 10^3 [\kappa^2 n^{-4} Q_D J(\lambda)]^{1/6} \text{ (in } \text{Å)} \quad (2)$$

where Q_D is the quantum yield of the donor in the absence of acceptor; n is the refractive index of the medium and κ^2 is the molecular orientation factor. At the distance R_{0A} , one-half of the donor molecules decay by energy transfer and one-half decay by the usual radiative and nonradiative processes. It is the separation distance between donor and acceptor which yields 50% energy transfer efficiency. Another important parameter, the reduced concentration γ_{DA} is calculated by

$$\gamma_{DA} = \frac{[A_A]}{[A_{0A}]} \quad (3)$$

where $[A_A]$ is the acceptor concentration and $[A_{0A}]$ is the critical acceptor concentration given by

$$[A_{0A}] = 3000/2\pi^{3/2} N R_{0A}^3 \quad (4)$$

where N is Avogadro's number.

In case of pure donor and in the absence of energy diffusion within the donor molecules, the decay function of the donor is given by

$$I_D = I_0 \exp(-t/\tau_D) \quad (5)$$

In presence of acceptor the donor fluorescence follows Förster kinetics of energy transfer [1]

$$I_D = I_0 \exp(-t/\tau_D) \exp[-2\gamma_{DA}(t/\tau_D)^{1/2}] \quad (6)$$

where τ_D is the decay of the donor in the absence of acceptor. The first term in Eq.(6) indicates the first-order deactivation and corresponds to unquenched fluorescence lifetime of the donor, while the second term accounts for excitation energy transfer due to dipole-dipole interaction with acceptor molecules. Förster kinetics for energy transfer assumes no diffusion and is applicable in the limit of low donor and high acceptor concentration. Therefore, it does not account the excitation migration. The donor-acceptor interaction strength and the donor-donor interaction strength are given by (α) and (β) as follows[9,10]

$$\alpha = 1/\tau_D(R_{0A})^6 \text{ and } \beta = 1/\tau_D(R_{0D})^6 \quad (7)$$

where R_{0D} is the critical transfer distance for excitation energy migration among donors. This is obtained from the spectral overlap of the absorption and fluorescence spectra of the donor. In the limit of high donor concentration, when donor-acceptor interaction strength (α) is comparable to donor-donor interaction strength (β) ($\alpha \approx \beta$), donor decay kinetics is modified. According to the theories put forward by Huber[9,10] and LAF[16] when α and β are of comparable strength, the donor fluorescence decay follows the following expression

$$I_D = I_0 \exp[-(t/\tau_D) - 2(\gamma_{DD}/2^{1/2} + \gamma_{DA})(t/\tau_D)^{1/2}] \quad (8)$$

Where the parameter γ_{DD} is defined as

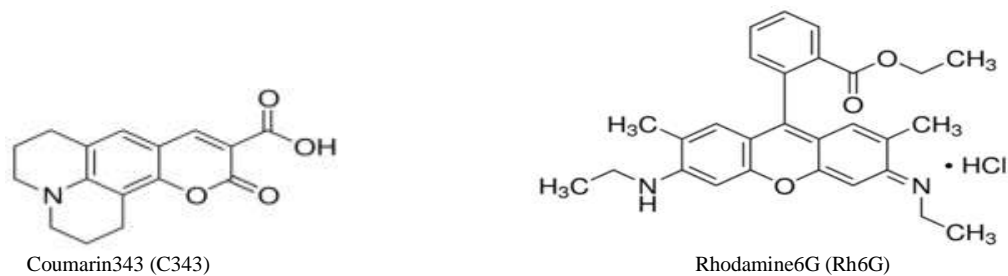
$$\gamma_{DD} = \frac{[A_D]}{[A_{0D}]} \quad (9)$$

Here $[A_D]$ is the donor concentration and $[A_{0D}]$ is the critical donor concentration given by

$$[A_{0D}] = 3000/2\pi^{3/2} N R_{0D}^3 \quad (10)$$

III. Experimental Procedures

Materials. Coumarin343 and Rhodamine6G dyes were purchased from Sigma Aldrich USA and poly (methylmethacrylate) (PMMA) was purchased from Hi-media, India and were used without further purification. The solvent (DMF) used in this study is of spectroscopic grade. The molecular structural formulas of C343 and Rh6G are shown in Scheme1.



Sample Preparation. Coumarin343 and Rhodamine6G doped PMMA films were prepared by dissolving the required quantity of PMMA in dimethylformamide (DMF) and mixing it with the desired concentration of dyes dissolved in the same solvent. This homogeneous mass was slowly heated to a temperature of about 40°C and poured in a Polypropylene dish and allowed to dry for 2-3 days at room temperature to obtain transparent films. The films were then cut to required shape and size. The thickness of the films ($\approx 0.3\text{mm}$) was kept constant by using equal weight of PMMA for all the samples.

Steady-State and Time-Domain Fluorescence Measurements. Absorption and fluorescence spectra were recorded by using Perkin-Elmer Lambda-35 UV-Visible spectrometer and Perkin-Elmer LS-55 fluorescence spectrometer respectively. The fluorescence spectra were recorded under constant geometric conditions. Fluorescence decay times were recorded using Edinburgh Instrument (EI) FLA-199 time domain single photon counting spectrometer and analyzed by related software as described in ref.[18]. The excitation source was a thyratron gated hydrogen lamp. The excitation and emission wavelengths were positioned at 444nm and 495nm respectively for recording the decay. The instrumental /Lamp profile (IRF) was measured at the excitation wavelength using Ludox scatter. Photomultiplier tube (PMT) XP2020x was used as detector. The decay curves were fitted using single exponential fit and Förster fits by judging the χ^2 values, standard deviations and residuals.

IV. Results and Discussion

Steady state analysis. Absorption and emission spectra of coumarin343 (donor) and rhodamine6G (acceptor) in PMMA are shown in Fig1.(a) and Fig.(b) respectively and the positions of band maxima are given in table 1. For coumarin343 the absorption and emission maxima lie at 444nm and 496nm respectively. Rhodamine6G shows an absorption maximum around 535nm and a corresponding emission maximum at 600nm.

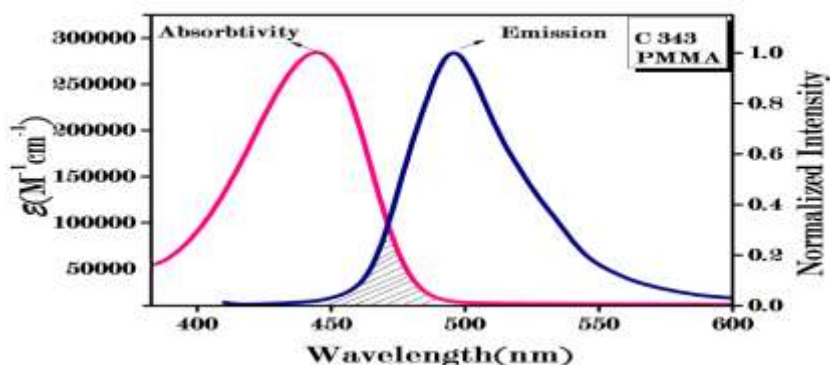


Fig1.(a).Absorption and emission spectra of coumarin343($1 \times 10^{-4}\text{M}$) in PMMA

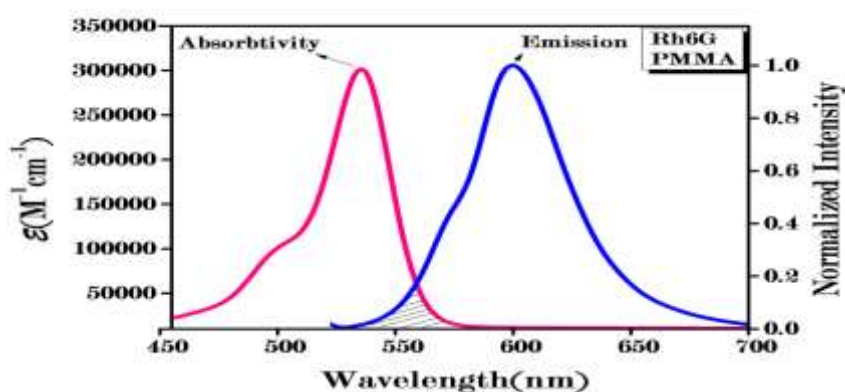


Fig1.(b)Absorption and emission spectra of rhodamine6G(1×10^{-4} M) in PMMA.

Table 1 Photo physical parameters for Coumarin343-Rhodamine6G system in PMMA

Donor		Acceptor			$J(\lambda)(M^{-1}cm^3)$	$R_0(\text{Å})$	$J_{DD}(\lambda)(M^{-1}cm^3)$	$R_{DD}(\text{Å})$	(α/β)
λ_{abs}	λ_{em}	λ_{abs}	λ_{em}						
444nm	496nm	535nm	600nm	2.2×10^{-13}	52.6	1.5×10^{-13}	49.8	1.3	

λ_{abs} wavelength of absorption maximum; λ_{em} wavelength of emission maximum; $J(\lambda)(M^{-1}cm^3)$ overlap integral for donor-acceptor interaction; $R_0(\text{Å})$ critical transfer distance between donor-acceptor molecules; $J_{DD}(\lambda)(M^{-1}cm^3)$ overlap integral for donor-donor interaction; $R_{DD}(\text{Å})$ critical transfer distance between donor-donor molecules; (α/β) ratio of donor-acceptor and donor-donor interaction parameters.

The absorption and emission spectra of coumarin343 and rhodamine6G do not show any concentration dependence up to the concentration used in this study and it rules out the possibility of the formation of dimers (or) higher aggregates. No new absorbing species is formed when coumarin343 and rhodamine6G are mixed together in PMMA matrix which is evident from the fact that the spectra remain unaffected upon change in the concentrations of the dyes. The overlapping of the absorption spectrum of rhodamine6G and emission spectrum of coumarin343 is shown in Fig.2 (a).The overlapping condition is an important factor for an efficient energy transfer from donor to acceptor as envisaged according to Förster theory[1]. In addition, there is also a significant overlap between the absorption and fluorescence spectra of the donor itself as evident from Fig1(a) indicating the possibility of donor-donor migration of excitation energy. In the present case ,therefore the possibilities of both types of energy transfer viz. long range Förster energy transfer and short range migration should be present. The values of overlap integral $J(\lambda)$ for donor-acceptor system using Eq.(1) is 2.2×10^{-13} and the critical transfer distance R_0 between donor-acceptor using Eq.(2)(using measured values of $Q_D=0.63$ [23] and $n=1.4$ [26] is $52.6(\text{Å})$. On the other hand the values of overlap integral $J_{DD}(\lambda)$ and critical transfer distance R_{DD} for donor-acceptor interaction are 1.5×10^{-13} and $49.8(\text{Å})$ respectively (Table1). The ratio of donor-acceptor and donor-donor interaction parameters (α/β) is 1.3 which suggests a higher dipole-dipole energy transfer rate.

The fluorescence spectra of coumarin343 in the prescence of different concentrations of acceptor (rhodamine6G) are shown in Fig.2(b). In all spectra, the concentrartion of coumarin343 has been kept constant (1×10^{-4} M) and the concentration of rhodamine6G has been varied from 0M to 6×10^{-4} M. For all the samples the geometrical conditions were kept strictly identical . The excitation wavelength was kept at 444nm as at this wavelength the absorption by rhodamine6G is very small. It is evident from these spectra that with the increase in the concentration of rhodamine6G succsesive quenching of coumarin343 fluorescence occurs accompanied by succsesive enhancement in the fluorescence intensity of rhodamine6G. Since rhodamine6G is not directly excited at the wavelength of excitation(444nm), the enhancement of rhodamine6G fluorescence is due to energy transfer from coumarin343,which depends on concentration of rhodamine6G. A red shift is observed in the acceptor fluorescence maxima with increase in the acceptor concentration. This can be attributed to the spectral overlap between the donor and acceptor fluorescence which recodes with increasing acceptor concentration. There is also an apperent blue shift in the donor emission possibly due to self absorption. However, the steady state data qualitatively indicate the presence of energy transfer and no quantitave analysis is possible only on their basis. For a clear understanding a time domain analysis is necessary. In the present study the steady state analysis has been used only to evaluate the overlap integral from the spectra and the critical transfer distance with decrease in the intensity of the donor emission upon increase in the acceptor concentration.

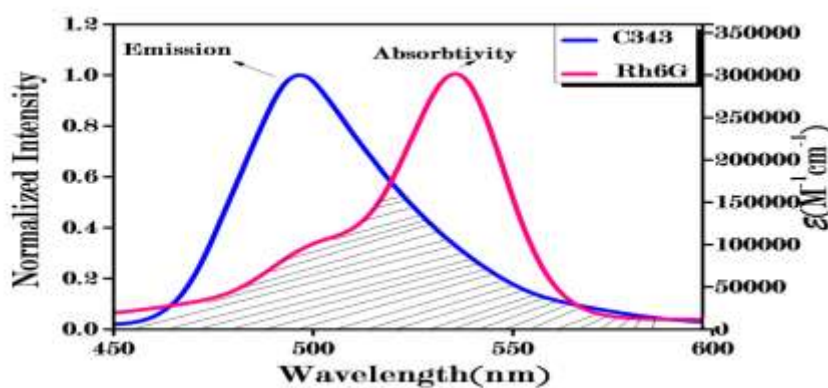


Fig.2(a) .Spectral overlap of the absorption spectrum of rhodamine6G and the emission spectrum of coumarin343 in PMMA.

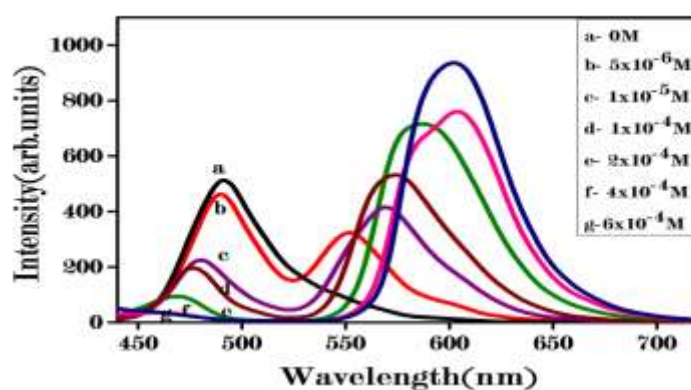


Fig.2(b).Variation in emission intensity of donor (C343 fixed at $1 \times 10^{-4} \text{M}$) on increasing acceptor (Rh6G) concentration (from 0M to $6 \times 10^{-4} \text{M}$).

V. Time Domain Analysis.

Donor Dynamics

Life time of the pure donor C343 ($1 \times 10^{-4} \text{M}$) is 5.3ns. The decay is single exponential with χ^2 value equals 1.1 and the fit is very good as the data points are properly distributed (Fig.3(a)). Addition of rhodamine6G results in deviation from single exponential decay to non-exponential (Fig.3(b)) and the lifetime of donor fluorescence decreases (Table2). The deviation from single exponential becomes more pronounced with the increase in the acceptor concentration and χ^2 shows a gradual increase. The decay curves at higher acceptor concentrations fit well into the Förster's equation of energy transfer (Eq.(6)). The decay parameters obtained from the fitting of decay curves with single exponential and Förster function for different combination of donor-acceptor concentration are given in Table 2. Experimentally observed data can be divided into two categories: (i) low acceptor concentrations and (ii) high acceptor concentrations.

(i) Donor Dynamics at Low Acceptor Concentration The decay curves for coumarin343(donor)($1 \times 10^{-4} \text{M}$) in the presence of low concentrations of rhodamine6G do not significantly deviate from single exponential although good fit is obtained with the help of the Förster fit as evident from the χ^2 values and standard deviations. The decay parameters obtained from the fitting of decay curves with single exponential and Förster fit for different combinations of donor-acceptor concentration are given in Table 2. There is a regular increase in the value of γ with increase in the acceptor concentration consistently in accordance with the Förster theory. In order to compare these γ values with theoretical data they have been reproduced in Table 3. The values of the critical acceptor concentration $[A_{0A}]$ and the critical transfer distance R_{0A} for all donor-acceptor concentrations are also given in Table 3. Values of R_{0A} and γ_{DA} have been calculated with the help of Eqs. (3) and (4). A comparison of γ_{obs} and γ_{DA} has been made in order to find the agreement between the experimental and theoretically obtained values. It can be seen that on increasing acceptor concentration the observed value of the reduced concentration (γ_{obs}) shows a regular increase. However, it is close to the observed value only at higher concentration. Also, at low acceptor concentrations up to $1 \times 10^{-5} \text{M}$ the observed values of critical transfer distance R_{0A} is greater than the value of critical transfer distance R_0 obtained from steady-state spectral measurements (52.6Å). Thus at low acceptor concentrations fitting of data with Förster's function (up to $1 \times 10^{-5} \text{M}$) seems inconsistent. This

inconsistency is due to the fact that Förster theory assumes low donor and high acceptor concentration. At low acceptor concentrations the rate of donor-donor migration is prominent and the donor-acceptor transfer rates are comparable only at higher concentrations. Therefore, the role of donor-donor excitation migration becomes significant as suggested by Huber[9,10] and LAF[16]. The modified decay functions according to Huber[9,10] and LAF[16] are given by Eq.8 and assumes the same form as the Förster function with γ_{DA} replaced by $\gamma_{cal} (= \gamma_{DD}/2^{1/2} + \gamma_{DA})$, γ_{DD} is related to the donor-donor interaction and can be calculated by using the relation $\gamma_{DD} = [A_D]/[A_{0D}]$. Values of γ_{cal} obtained from the values of γ_{DD} and γ_{DA} are quite in agreement with γ_{obs} . This agreement can be better realized from the corresponding values of R_{obs} also given in Table3. Thus at low acceptor concentrations the theoretical models of LAF and Huber satisfactorily explain the energy migration. The value of γ_{DD} which is equal to 0.0263 is more than the γ_{DA} values at low acceptor concentrations. It means a higher contribution of the migration at low acceptor concentrations.

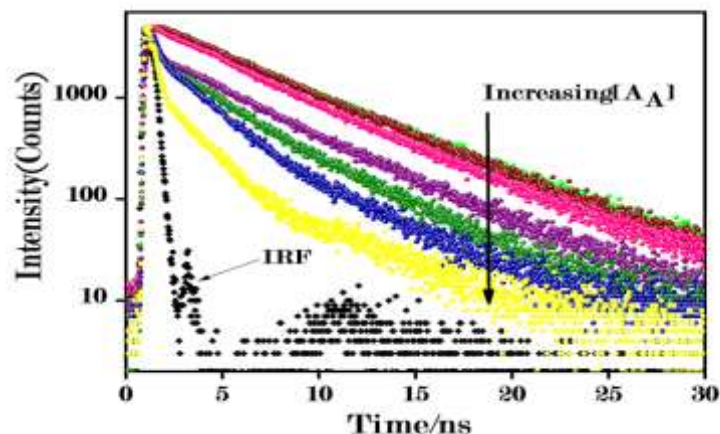


Fig.3(a). Fluorescence decay curve of C343 (pure donor at 1×10^{-4}) in thin film of PMMA at 495nm (λ_{exc} :444nm) with varying concentrations of Rh6G(acceptor). Rh6G concentrations are (i)0M,(ii) 5×10^{-6} M,(iii) 1×10^{-5} M,(iv) 1×10^{-4} M,(v) 2×10^{-4} M,(vi) 2×10^{-4} M,(vii) 4×10^{-4} M and (viii) 6×10^{-4} M.

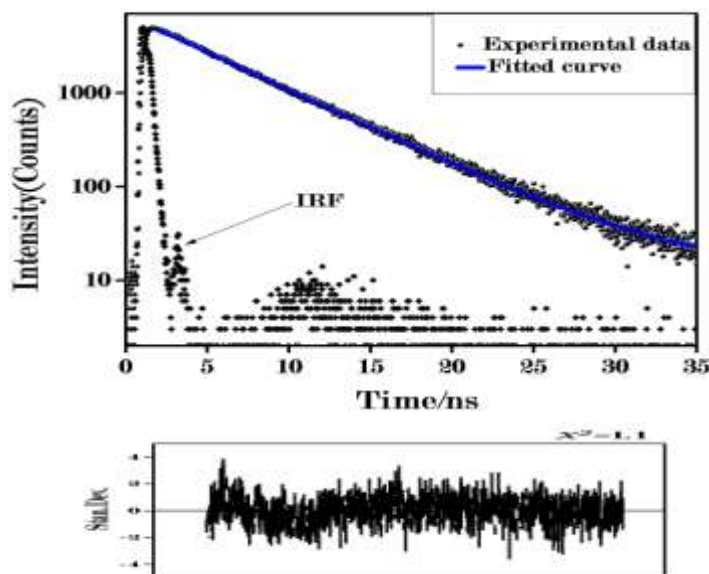
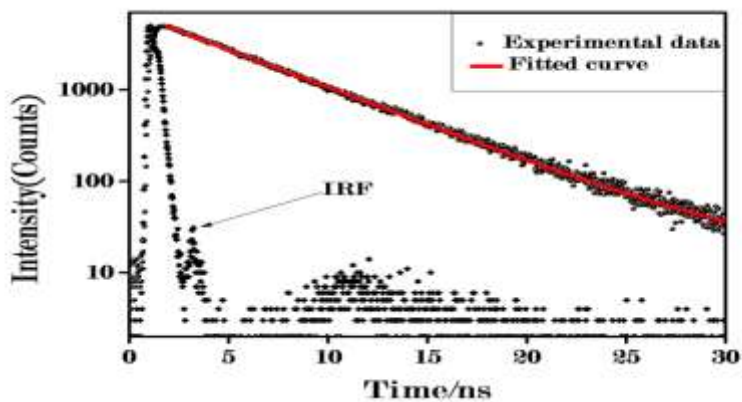
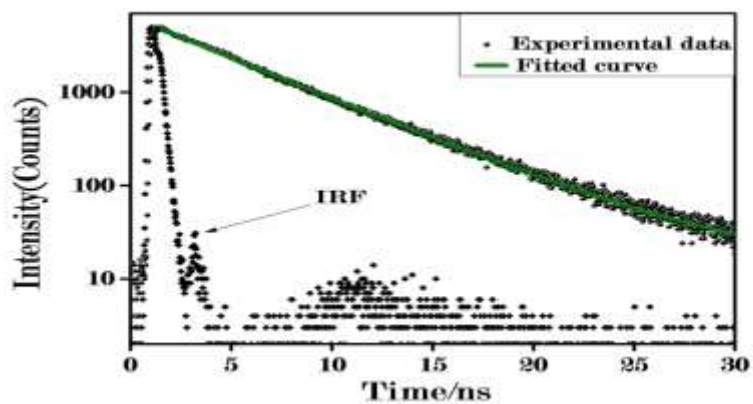


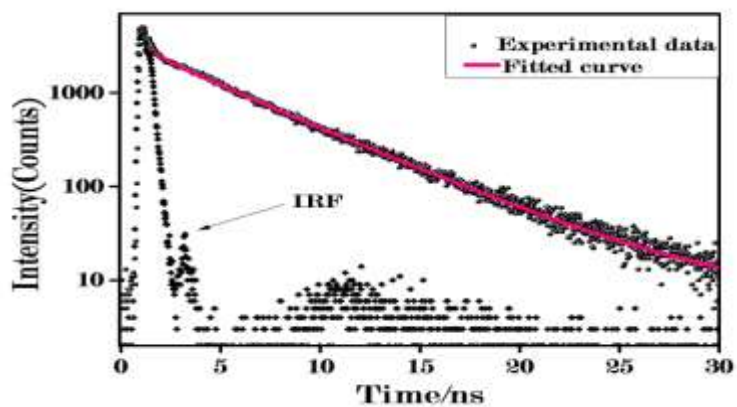
Fig.3(b). Fluorescence decay curve of C343(pure donor at 1×10^{-4})in thin film of PMMA at 495nm(λ_{exc} : 444nm). Fitted with a single exponential function.



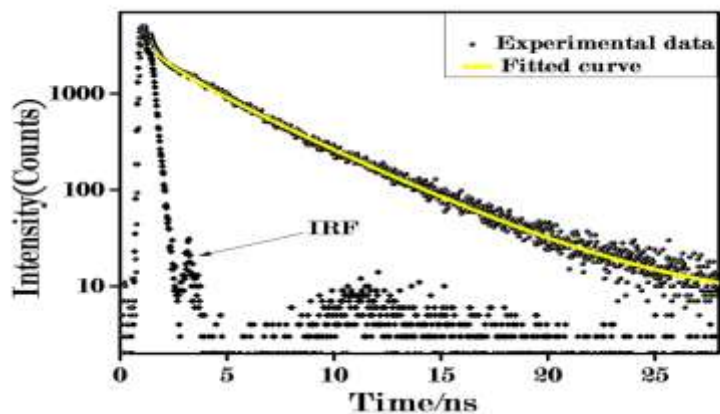
(i)



(ii)



(iii)



(iv)

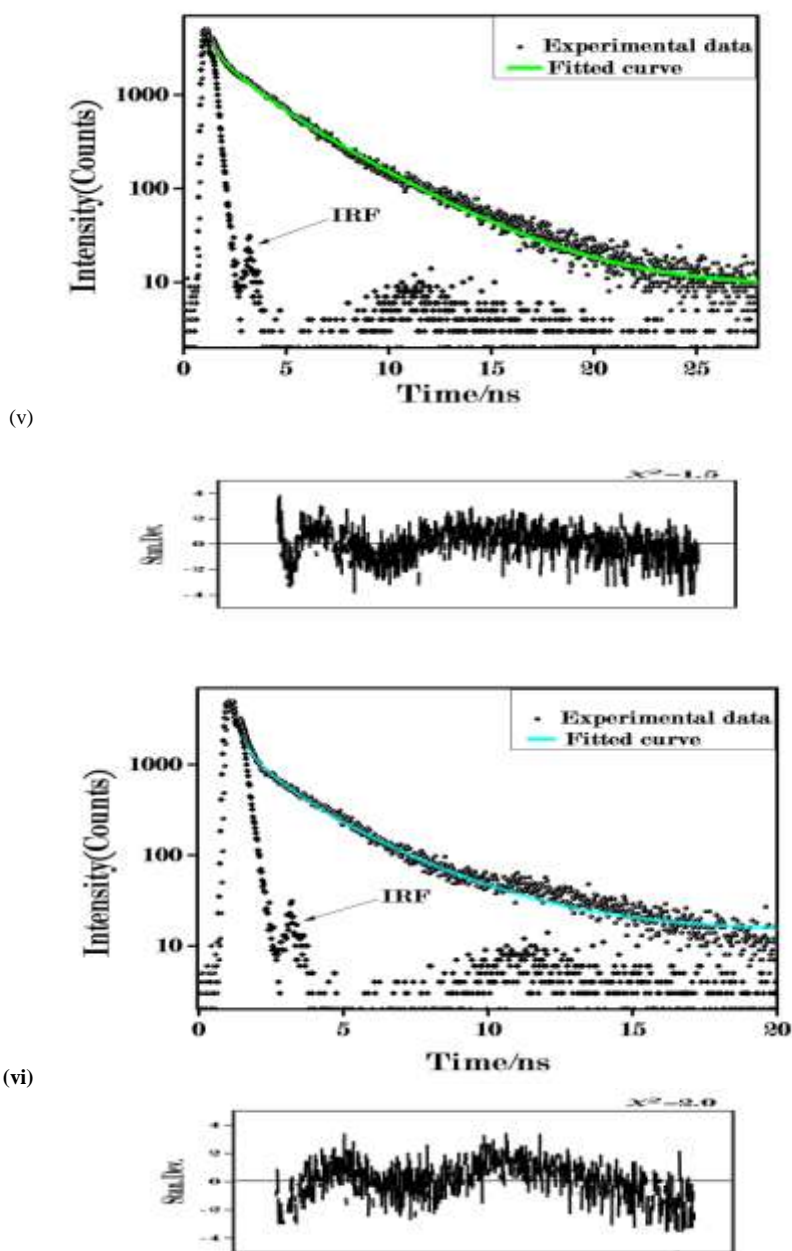


Fig.3 (c).Standard deviations for Förster function fitted decay curves of C343(donor at 1×10^{-4}) in thin film of PMMA at 490nm(λ_{exc} :444nm) with varying concentration of Rh6G(acceptor).Rh6G concentrations are (i) 5×10^{-6} M,(ii) 1×10^{-5} M,(iii) 1×10^{-4} M,(iv) 2×10^{-4} M, (v) 4×10^{-4} M and (vi) 6×10^{-4} M. Plotted are the experimental data(points) and the fitted data(colorful lines). Residuals and χ^2 values are also shown for each donor-acceptor combinations.

Table 2 Experimentally obtained parameters for C343-Rh6G system in PMMA for single exponential and Försters fit (λ_{exc} : 444nm)

Acceptor concentration [A](M)	Single exponential fit		Förster fit		
	τ (ns)	χ^2	$(\gamma)_{obs}$	τ (ns)	χ^2
0	5.3	1.1	-	5.3(fixed)	1.1
5×10^{-6}	5.2	1.2	0.025	..	1.2
1×10^{-5}	4.9	1.9	0.030	..	1.3
1×10^{-4}	4.4	3.7	0.080	..	1.1
2×10^{-4}	3.4	9.4	0.087	..	1.3
4×10^{-4}	2.5	10.2	0.132	..	1.5
6×10^{-4}	1.8	11.6	0.201	..	2.0

τ (ns) fluorescence lifetime of donor ; $(\gamma)_{obs}$ reduced concentration obtained from Förster fitting.

Table 3 Experimentally observed and spectroscopically calculated values of various parameters in C343-Rh6G system.

[A](M)	Observed values			Spectroscopic values		
	γ_{obs}	$[A_{0A}](\times 10^{-3}M)$	$R_{0A}(\text{Å})$	γ_{DA}	γ_{DD}	$\gamma_{cal}(=\gamma_{DD}/2^{1/2} + \gamma_{DA})$
5×10^{-6}	0.025	0.20	130.1	0.0016	0.0263	0.020
1×10^{-5}	0.030	0.62	89.2	0.0032	0.0263	0.021
1×10^{-4}	0.080	2.29	58.0	0.0326	0.0263	0.051
2×10^{-4}	0.087	2.82	54.1	0.0653	0.0263	0.083
4×10^{-4}	0.132	3.01	52.9	0.1307	0.0263	0.149
6×10^{-4}	0.201	3.14	52.2	0.1960	0.0263	0.214

$[A_{0A}]$ observed value of critical acceptor concentration ; R_{0A} observed value of critical transfer distance between a donor-acceptor pair ; γ_{DA} reduced concentration obtained from spectroscopic measurements ; γ_{DD} value of reduced concentration for donor-donor interaction ; $\gamma_{cal}(=\gamma_{DD}/2^{1/2} + \gamma_{DA})$

(ii) Donor Dynamics at High Acceptor Concentrations The deviation of the decay of donor from exponential behavior to non-exponential behavior becomes more prominent as the concentration of the acceptor is increased. Decay of coumarin343 in the presence of high acceptor concentration with respect to the donor concentration fitted with Förster function is also shown in Fig.3(c). As mentioned before, the decay data fit well with the Förster expression as evidenced by the good values of the χ^2 . The observed values of γ_{obs} increases with increasing acceptor concentration and as the concentration of rhodamine6G increases, the observed values of the critical transfer distance decreases and finally attains a constant value that match well with spectroscopically calculated value (52.6Å) . This indicates that the Förster [1] mechanism dominates over migration at higher acceptor concentrations.

The influence of energy migration in energy transfer in a dye pair acriflavin-rhodamineB and acriflavin-malachite green in PMMA has been studied earlier [25-28]. Pandey et al.[25-26] studied the excitation energy migration and transfer in a dye pair of acriflavin and rhodamine6G in PMMA. Tripathi et al.[27] studied the migration and diffusion modulated excitation energy transfer in a dye pair 7-diethylamino-4-methylcoumarin and 3,3'-dimethyloxycarbocyanine iodide. Excitation energy transfer between the dye pair acriflavin to rhodamine6G in various polymers PVA, cellulose acetate and PMMA has also been studied [28]. Our results for the C343 to Rh6G donor acceptor system are in agreement to the view that the migration of energy between donors is also an important pathway of energy transfer particularly at low acceptor concentrations. However, at high acceptor

concentrations the migration effect is masked by highly efficient Förster dipole-dipole energy transfer between donor and acceptor molecules.

Coumarin6 and its derivatives, rhodamine6G and other dyes are used in luminescent solar collectors(LSC) since Zewail and coworkers have proposed their importance in increasing the efficiency of solar concentrators [29-31]. Absorption of rhodamine6G extends mainly in the spectral range 470nm-550nm Fig.1(b). On other hand Coumarin343 absorbs in the spectral range 400nm-470nm. A mixture of coumarin343 and rhodamine6G will enable absorption within the spectral range 400nm-550nm. Energy transfer from coumarin343 to rhodamine6G will increase the intensity of emission of rhodamine6G. As both migration and dipole-dipole mechanisms are responsible for energy transfer, by optimizing both donor as well as acceptor concentrations an efficient energy transfer can be achieved for proper use in LSCs.

VI. Conclusion

In the polymer matrix PMMA, at higher acceptor concentrations with respect to the donor concentration, direct energy transfer can be explained on the basis of the Förster [1] theory of fluorescence resonance energy transfer. At low acceptor concentrations with respect to the donor concentration, excitation migration among donor molecules influences the transfer kinetics, and the results are satisfactorily explained on the basis of theories given by Huber [9,10] and LAF[16]. However, there is an efficient transfer from Coumarin343 to Rhodamine6G at higher acceptor concentration and the pair may be well suited for use in LSCs.

Acknowledgments

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