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Optical properties of Rh6G:C337 dye mixture in the sol-gel silica matrix

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The mixed dye method has received considerable attention because it is promising method to improve the efficiency of dye lasers and broaden the laser emission spectrum [1–3]. In a dye mixed solution, the excitation energy can be efficiently transferred from the donor to the acceptor. Two mechanisms involved in such an energy transfer are: (i) the radiative transfer, that is, the absorption of donor emission by an acceptor, and (ii) nonradiative transfer, that is, the excitation transfer takes place before the excited donor emits. The radiative energy transfer mechanism is found to be the dominant mechanism in dilute solutions [4, 5]. It is well known that the intermolecular energy transfer depends strongly on factors such as dye molecule concentrations, spectral overlap between the acceptor absorption spectra and the donor fluorescence spectra [6, 7]. Most of the studies conducted so far report investigation of energy transfer between dyes in liquid media. To our knowledge, no work has been reported on the energy transfer of Rh6G:C337 dye mixture in the sol-gel silica matrix. In this paper, the optical properties of Rh6G: C337 dye doped sol-gel silica matrices are investigated.

The dye doped sol-gel silica samples were prepared by hydrolysis and polycondensation of tetraethosysilane (TEOS), with formamide as a drying control chemical additive (DCCA) [8]. TEOS silica sol was prepared by keeping the molar ratio of TEOS/H_2O/Formamide/Ethanol at 1:4:4:4. Hydrochloric acid (HCl) of 0.1 ml served as a catalyst. After stirring the mixed solutions with above components for 30 min, dyes were added as molar concentrations. In all samples prepared this way the acceptor (Rhodamine 6G) molar concentration was kept constant at 1×10^{-4} M, whereas the donor (Coumarin 337) molar concentrations were varied (3×10^{-7}, 5×10^{-7}, 1×10^{-6}, and 5×10^{-6} M). After stirring the dye-doped solution for 60 min, it was poured into a teflon tube container and dried in a oven at 50 °C for 6 weeks. The dried samples were easily removed from the teflon tube container. The dimension of samples was 7.6 mm × 80 mm ($\phi \times L$). The absorption spectra of samples were recorded on a Varian CARY 5G UV-VIS-NIR Spectrometer. To record the fluorescence spectra, a linear Xenon flashlamp (EG&G) was used as the excitation source. The inner diameter of the flashlamp was 9 mm and arc length was 75 mm. The flashlamp was fired by a 1 μF capacitor allowing a maximum of 16 kV to apply to the flashlamp. The fluorescence spectra from samples were measured using ORIEL Multispc spectrograph. In order to study the dependence of

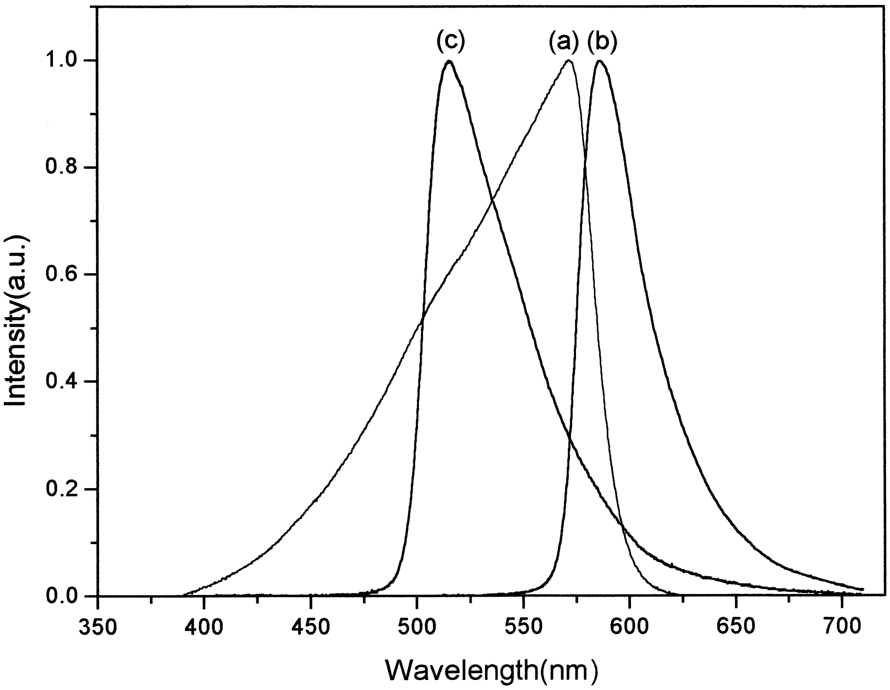

Figure 1 The absorption and the fluorescence spectra for Rhodamine 6G and Coumarin 337: (a) absorption spectrum of Rhodamine 6G, (b) fluorescence spectrum of Rhodamine 6G, and (c) fluorescence spectrum of Coumarin 337.

fluorescence spectrum on the excitation energy, fluorescence spectra were measured under various excitation energies of 30, 50, 70, and 90 J.

The absorption and fluorescence spectra of the sample are shown in Fig. 1, where (a) represent the absorption spectra of the pure Rhodamine 6G dye molecule in the sol-gel silica matrix. In Fig. 1 curves (b) and (c) represent the fluorescence spectra of the pure Rhodamine 6G and Coumarin 337 dye molecules in the sol-gel silica matrix, respectively. Good overlap between the absorption spectra of Rhodamine 6G and the fluorescence spectra of Coumarin 337 suggests that the energy transfer from the donor (Coumarin 337) to the acceptor (Rhodamine 6G) may occur. The fluorescence spectra of Coumarin 337 peaked at 485 nm in ethanol and 514 nm in sol-gel silica, representing a red-shift of 29 nm. Fig. 2 shows the fluorescence spectra of mixed dye samples in the sol-gel silica matrix with donor concentration of 3×10^{-7}, 5×10^{-7}, 1×10^{-6}, and 5×10^{-6} M, at a fixed acceptor concentration of 1×10^{-4} M and for an excitation energy of 30 J. All spectra were measured under the same experimental

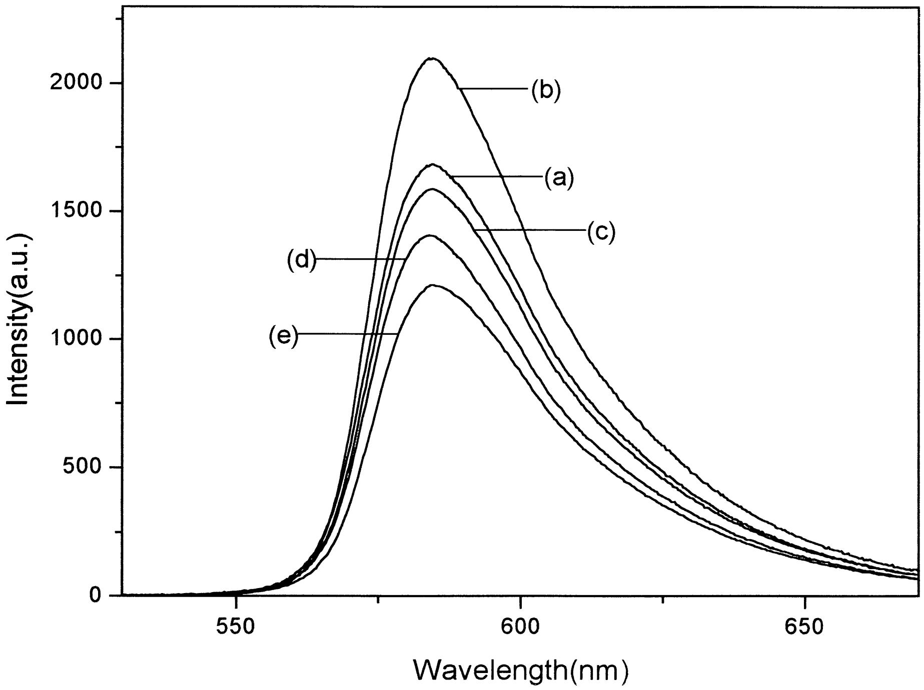

Figure 2 The fluorescence spectra intensity of mixed dye samples in sol-gel silica matrix with various donor concentrations for an excitation energy of 30 J: (a) 3×10^{-7}, (b) 5×10^{-7}, (c) 1×10^{-6}, and (d) 5×10^{-6} M, the sample with only the acceptor (Rh 6G) of 1×10^{-4} M is also plotted in (e).

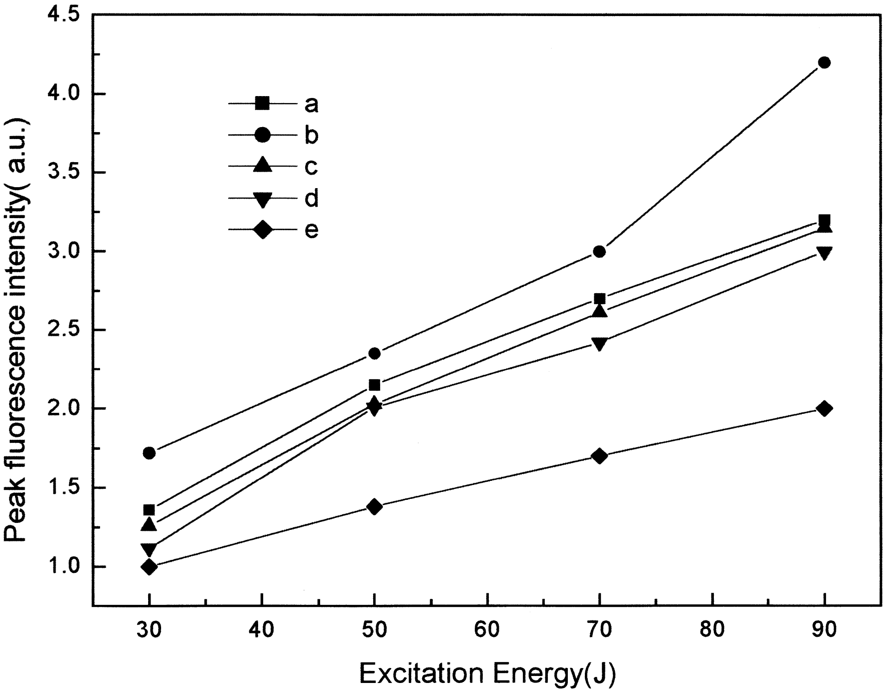

Figure 3 Variation of the peak fluorescence intensity under several excitation energies for various donor concentrations: (a) 3×10^{-7}, (b) 5×10^{-7}, (c) 1×10^{-6}, and (d) 5×10^{-6} M, the sample with only the acceptor (Rh 6G) of 1×10^{-4} M is also plotted in (e).

conditions. The maximum of the peak fluorescence intensity increases with increasing donor (Coumarin 337) concentrations to 5×10^{-7} M. A considerable enhancement of the peak fluorescence intensity of 170% were observed in the present dye mixture system, compared with pure Rhodamine 6G in sol-gel silica matrices. This fact indicates that good excited states for an efficient energy transfer from Coumarin 337 to Rhodamine 6G exist. Above the concentration of 5×10^{-7} M, the peak fluorescence intensity decreases with increasing the donor concentrations, suggesting that a good energy transfer to the acceptor will not be expected. Fig. 3 shows the variation of peak fluorescence intensity of the mixed dye samples in a sol-gel silica matrix under several excitation energies for various donor concentrations. Let us define the efficiency of the system as the ratio of the peak fluorescence intensity to the excitation energy. By adding the donor (Coumarin 337) molecules to the acceptor (Rhodamine 6G) dye, efficiency of Rhodamine 6G and Coumarin 337 dye doped samples increases considerably and maximum efficiency is obtained at the donor concentration of 5×10^{-7} M. The efficiency of the pure Rhodamine 6G sample is not so effective under high excitation energy. Therefore, the donor concentration of 5×10^{-7} M can be considered as the optimum concentration in the present sol-gel silica matrix consisting of Rh6G and C337.

In conclusion, we studied the fluorescence characteristics for Rh6G:C337 dye mixture in a sol-gel silica matrix for various donor (C337) concentrations for the first time. The present investigations of the energy transfer between Coumarin 337 (donor) and Rhodamine 6G (acceptor) showed that the fluorescence intensity increases with the donor concentrations up to the critical concentration (5×10^{-7} M). In addition, a significant enhancement of the fluorescence intensity was observed. Moreover, when the Coumarin 337 molecules are added to the Rhodamine 6G dye, the efficiency of the mixed dye samples in a sol-gel silica matrix increases considerably and the maximum efficiency is obtained at the donor concentration of 5×10^{-7} M.

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