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Photoluminescence Quenching Study of Composites Comprising Novel Fullerene-Based Acceptors and MDMO-PPV

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In this work, some novel methanofullerene derivatives of the formula [6,6]-phenyl–C_{61}–$(CH_2)_n$–COOR, where n = 2, R=C_4H_9, C_3H_7, C_2H_5, CH_3, $CH_2C_6H_5$, $CH(CH_3)_2$, have been studied together with the reference compound PCBM (n = 3 and R=CH_3). We have measured the dependence of polymer photoluminescence (PL) in the blends of poly(2-methoxy-5-(3′7′-dimethyloctyloxy)-1,4-phenylene-vinylene) (MDMO-PPV) and new fullerene derivatives, as well as PCBM and C_{60}, on the concentration of fullerene. Strong PL quenching due to the photoinduced electron transfer was observed for all fullerenes studied. The derivatives with R=C_4H_9, C_3H_7, and C_2H_5 showed a more effective quenching than PCBM and C_{60}. This means that the novel metanofullerens may be selected as good acceptor materials for organic photovoltaic cells.

Keywords: electron transfer; fullerene derivatives; photoluminescence quenching; photovoltaics

INTRODUCTION

The design and synthesis of new materials for the organic photovoltaic (PV) cells based on the polymer-fullerene derivative composites are intensively investigated nowadays [1,2]. Due to the effective

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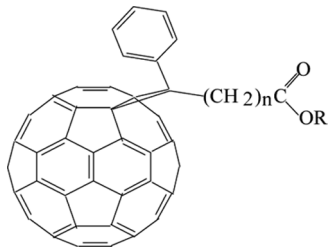
photoinduced electron transfer between polymer and fullerene [3–5], solar cells with high power conversion efficiency were prepared by using these systems. The photoinduced electron transfer results in the polymer photoluminescence quenching, so such quenching is the necessary condition of the applicability of the material for photovoltaic devices [2,6].

Different fullerene derivatives were proposed as acceptors in polymer/fullerene composites for photovoltaic applications [7,8]. The methanofullerene derivative [6,6]-phenyl$-C_{61}-$butyric acid methyl ester (PCBM) is most often used in such systems [9,10]. Poly(2-methoxy-5-(3′7′-dimethyloctyloxy)-1,4-phenylene-vinylene) (MDMO-PPV) in composites with PCBM results in solar cells with high-power conversion efficiency [1,2].

In this work, we report some new soluble methanofullerene derivatives of the formula [6,6]-phenyl$-C_{61}-(CH_2)_n-$COOR where n = 2, R=C_4H_9, C_3H_7, $CH_2C_6H_5$, $CH(CH_3)_2$, C_2H_5, CH_3 (the compound with n = 3 and R=CH_3 is the well-known PCBM).



We have measured the dependence of the polymer photoluminescence (PL) in the blends of MDMO-PPV and fullerene derivatives on the concentration of fullerene in order to evaluate the potential of our compounds as acceptors in the process of photoinduced electron transfer. Blends of MDMO-PPV with PCBM and C_{60} were also studied for comparison.

EXPERIMENTAL

New methanofullerenes were synthesized as described elsewhere [11]. MDMO-PPV was a commercial product of Aldrich.

Films of MDMO-PPV/fullerene composites were prepared by casting the chlorobenzene solutions containing MDMO-PPV and fullerene in defined ratios onto a glass substrate.

The absorption and PL spectra of composite films were measured with an Ocean Optics PC1000 plug-in spectrometer. The excitation

line for PL measurements, $\lambda_{exc} = 450$ nm, was obtained from an In-Ga light-emitting diode. The absorption spectrum of MDMO-PPV has a maximum at about 490–500 nm. The absorbance of all the films at 450 nm exceeded 2.5, which means that more than 99% of the excited light was absorbed in the film during PL measurements.

RESULTS AND DISCUSSION

We have measured PL spectra of the films containing blends of MDMO-PPV with fullerene C_{60} or with methanofullerene with the concentration of the fullerene component of 0, 0.26, 0.5, 1, 2, 4, and 7.7% by weight.

The typical PL spectra of MDMO-PPV:methanofullerene films prepared using different ratios of the components are shown in Figure 1. The PL spectra exhibit a double band with maxima at 580 and 625 nm due to MDMO-PPV photoluminescence. When the fullerene concentration increases, the intensity of the PL band decreases. The quenching of the polymer luminescence by fullerene is due to the electron transfer from the photoexcited polymer to fullerene molecules [3–6]. Similar PL spectra showing the polymer PL quenching due to the photoinduced electron transfer were observed for composites of C_{60} with poly(2-methoxy-5-(2′-ethylhexyloxy)-1,4-phenylene-vinylene) (MEH-PPV) [6].

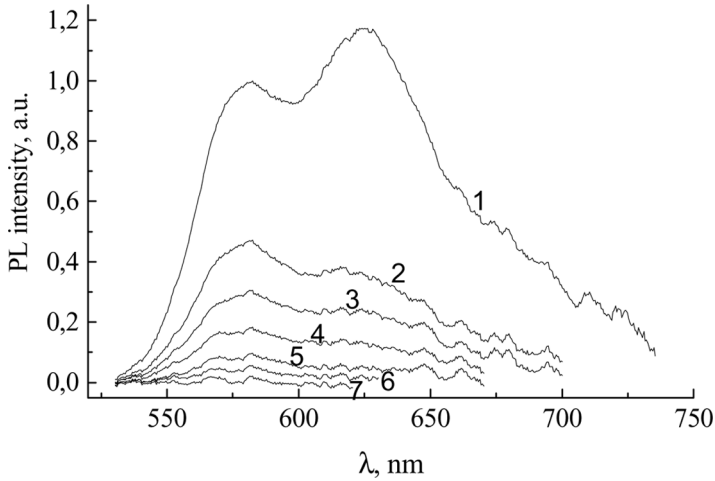


FIGURE 1 PL spectra ($\lambda_{exc} = 450$ nm) of a MDMO-PPV film (1) and MDMO-PPV:F films (F is fullerene derivative [6,6]-phenyl$-C_{61}-(CH_2)_2-COOC_2H_5$) for the concentrations of F equal to 0.26 (2), 0.5 (3), 1 (4), 2 (5), 4 (6), and 7.7 (7)% by weight.

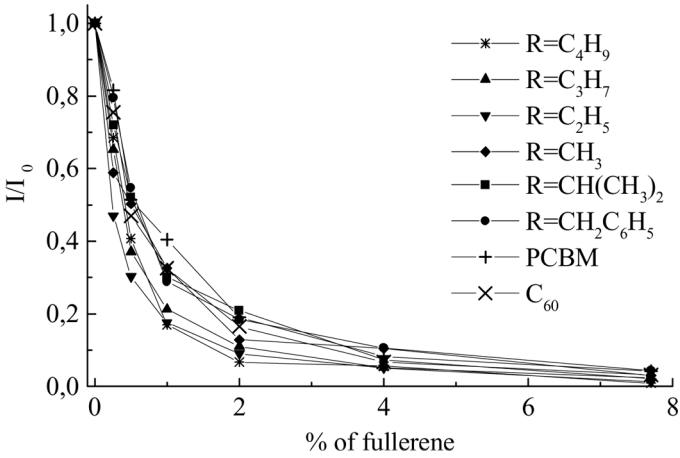


FIGURE 2 Dependences of the relative intensities of PL at 580 nm for MDMO-PPV:F films (F corresponds to C_{60}, PCBM, and fullerene derivatives [6,6]-phenyl–C_{61}–$(CH_2)_2$–COOR with R=C_4H_9, C_3H_7, C_2H_5, CH_3, $CH_2C_6H_5$, $CH(CH_3)_2$) on the concentration of F.

For a pure MDMO-PPV film, the 625-nm PL component is more intense. For composite films, the 580-nm component is the most intense. For the quantitative estimation of the effect of fullerene concentration on the PL intensity, we used the 580-nm band.

The dependence of the relative intensity I/I_0 (where I_0 is the intensity for a pure MDMO-PPV film) of the 580-nm maximum on the fullerene concentration for all fullerens studied is shown in Figure 2. The corresponding numerical data are given in Table 1.

TABLE 1 Relative Intensities of the 585-nm PL Band of MDMO-PPV in Composite Films for Different Fullerene Concentrations

	Fullerene							
	R in [6,6]-phenyl–C_{61}–$(CH_2)_2$–COOR							
% of fullerene	$CH(CH_3)_2$	$CH_2C_6H_5$	C_4H_9	C_3H_7	C_2H_5	CH_3	PCBM	C_{60}
0	1	1	1	1	1	1	1	1
0.26	0.720	0.795	0.685	0.652	0.470	0.589	0.815	0.754
0.5	0.522	0.547	0.407	0.371	0.303	0.503	0.515	0.470
1	0.302	0.289	0.170	0.213	0.176	0.326	0.405	0.326
2	0.209	0.185	0.067	0.110	0.090	0.129	0.190	0.165
4	0.073	0.106	0.056	0.052	0.051	0.105	0.083	0.067
7.7	0.022	0.044	0.009	0.024	0.013	0.030	0.042	0.032

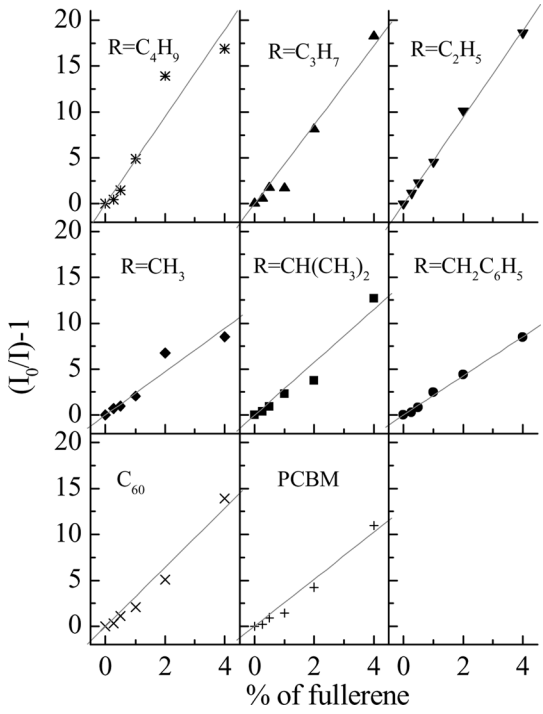


FIGURE 3 Linear fits for the dependences of the quenching factor $(I_0/I) - 1$ on the fullerene concentration for the data of Figure 2.

For all the fullerenes studied, the polymer PL is almost completely quenched at high concentrations (more than 7.7%). Individual quenching abilities of the fullerenes may be evaluated from the PL spectra intensities at low concentrations (less than 4%). At low concentrations, some fullerenes exhibit a stronger quenching effect than others, which may probably be a merit of their acceptor ability in the process of photoinduced electron transfer.

For the quantitative evaluation of the quenching properties of the fullerenes studied, we have used a linear fit of the Stern-Volmer type for the quenching factor $Q = (I_0/I) - 1$, where I_0 and I are the PL intensities for a pure polymer film and for a composite film, respectively. The corresponding data are given in Figure 3. The linear fit is described by the equation $Q = kC$ where C is the fullerene concentration in weight %. The slope k of a straight line representing such linear fit may be a measure of the quenching rate. The slopes of linear fits obtained from the data of Figure 3 for all the fullerenes studied are

TABLE 2 Slopes of the Straight Lines Representing Linear Fits of Figure 3.

Fullerene	k	Fullerene	k
R=C_4H_9	4.75	R=$CH(CH_3)_2$	2.87
R=C_3H_7	4.30	R=$CH_2C_6H_5$	2.13
R=C_2H_5	4.72	C_{60}	3.21
R=CH_3	2.36	PCBM	2.56

listed in Table 2. The data of Table 2 show that ethyl, propyl, and butyl derivatives exhibit the quenching rate nearly two times higher than that of PCBM and sufficiently higher than that of C_{60}. Other derivatives demonstrate quenching rates of the same order as C_{60} and PCBM.

It follows from the obtained data that the acceptor abilities of our compounds are not worse or even better than those for C_{60} and PCBM.

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