

Supporting Information

Photocatalytic Chip Inspired from Photovoltaics of Polymer-immobilized Surfaces: Self-assembly and other factors†

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Section S1. Materials and Instrumentations

Rhodamine-B ($\geq 95\%$ pure, powder), Poly (3-Hexylthiophene) (P3HT) (99.995% trace metals basis, P-type), Phenyl-C₆₁-Butyric acid methyl ester (PCBM) (99.5% N-type), Ethanol and Chloroform were purchased from Sigma Aldrich. Glass films (75 mm × 25 mm × 1 mm) were purchased from blue star polar industrial corporation company, India. Distilled water was used for all dye degradation studies experiments. UV-Visible absorbance study of solutions for dye degradation studies was carried out on Shimadzu UV-2401 spectrophotometer. FESEM images of thin films were captured on F E I Quanta FEG 200 - High Resolution Scanning Electron Microscope. The wettability property of the films was measured through water contact angle measurement (Holmarc model HO-IAD-Cam-01B). Atomic force microscope (AFM) data were recorded for the P3HT-PCBM surfaces on glass through Nova 1.0.26 RC1 atomic force microscope in semi-contact mode with NT-MDT solver software. Transient photoluminescence spectroscopy analysis of P3HT-PCBM solutions (2mg/L concentration in chloroform) was measured through TCSPC – Fluorescence Spectrophotometer (Edinburg Instruments, FLS980) at 578nm emission wavelength and the lifetime of excitons were calculated by using bi-exponential model for fitting the photoluminescence decay curves. The photocatalytic dye degradation experiment was carried out by either sunlight (with light intensity of 186 w/m²) or 200W Hg (Xe) medium pressure lamp (66454, Newport Oriel instrument) with distance of 20 cm from lamp to sample at room temperature (30°C).

Section S2. Preparation of P3HT-PCBM films

We prepared P3HT-PCBM films on glass surfaces with different weight ratios of P3HT and PCBM compounds (weight percent in terms of PCBM contents) such as 1:0 (0%), 1:0.25 (20%), 1:0.5 (33.3%), 1:0.75 (42.9%), 1:1 (50%), 0.75:1 (57.1%), 0.5:1 (66.7%) and 0.25:1 (80%) by drop-casting method. In all the above cases, the total amount of P3HT-PCBM was 5 mg. The glass surfaces were cut into square slides with dimensions of 2.5 cm × 2.5 cm. Then the surfaces were cleaned by piranha solution treatment at 100°C for 2 hours followed by drying. Initially we prepared the stock solution of P3HT and PCBM materials and we made a P3HT-PCBM composites solution using stock solutions [Stock solution of P3HT: 50mg of P3HT was dissolved in 10 mL of chloroform (5mg/mL concentration) and Stock solution of PCBM: 50mg of PCBM was dissolved in 10mL of chloroform (5mg/mL concentration)]. In a vial, we made 5 mg of P3HT-PCBM composites solution from stock solution of P3HT and PCBM compounds in 2 mL of chloroform with respect to the below-mentioned weight ratio of P3HT and PCBM (shown in table S1). The solutions were subjected to 1 hour of stirring for obtaining a uniform mixture before drop-casting. The P3HT-PCBM solutions were drop-casted on glass surfaces at room temperature. The resulting P3HT-PCBM films were annealed at 110°C for 3 hours.

Table S1. Varying Ratios of P3HT-PCBM

Ratio of P3HT-PCBM	Ratio by weight interms of PCBM contents (%)	Amount of P3HT (mg)	Volume took from P3HT (5mg/mL) stock solution	Amount of PCBM (mg)	Volume took from PCBM (5mg/mL) stock solution	Dilution with CHCl₃ (upto 2mL)
1:0	0%	5	1000 μ l	0	0 μ l	1000 μ l
1:0.25	20%	4	800 μ l	1	200 μ l	1000 μ l
1:0.5	33.3%	3.4	680 μ l	1.6	320 μ l	1000 μ l
1:0.75	42.9%	2.95	590 μ l	2.05	410 μ l	1000 μ l
1:1	50%	2.5	500 μ l	2.5	500 μ l	1000 μ l
0.75:1	57.1%	2.05	410 μ l	2.95	590 μ l	1000 μ l
0.5:1	66.7%	1.6	320 μ l	3.4	680 μ l	1000 μ l
0.25:1	80%	1	200 μ l	4	800 μ l	1000 μ l

Section S3. Characterization of P3HT, PCBM and P3HT-PCBM composites films by fluorescence microscopy:

Fluorescence images of films were captured by LEICA DMi8 fluorescence microscope instrument with 40X objective lens with 460 nm wavelength excitation and provided below (S1). Z-stacked Fluorescence microscopic images of P3HT-PCBM films with different ratios have been provided separately in Fig. S2.

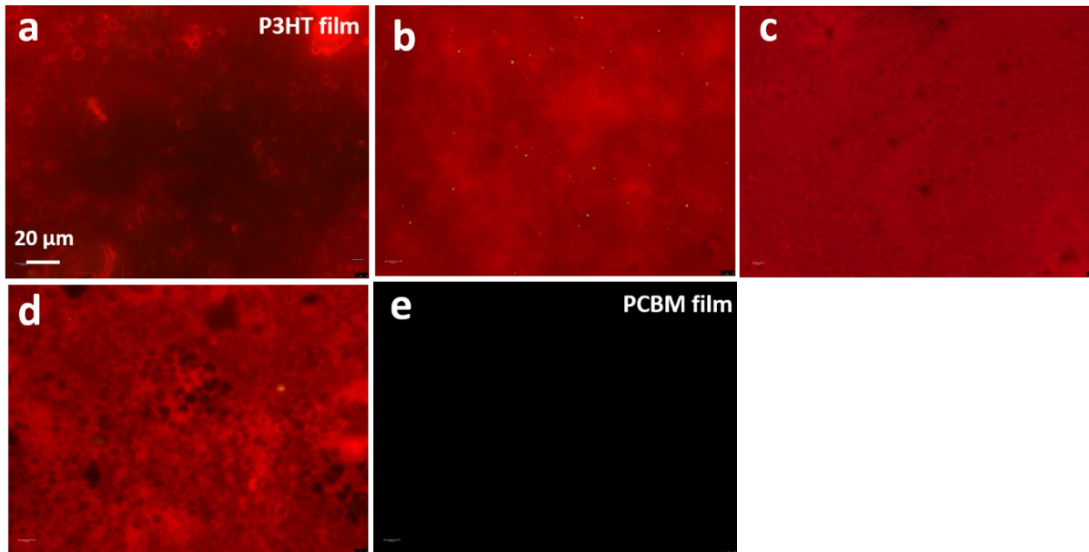


Fig. S1 Fluorescence microscopic images of P3HT-PCBM films with different ratio a) 1:0, b) 1:0.25, c) 1:0.5, d) 1:0.75 and e) 0:1

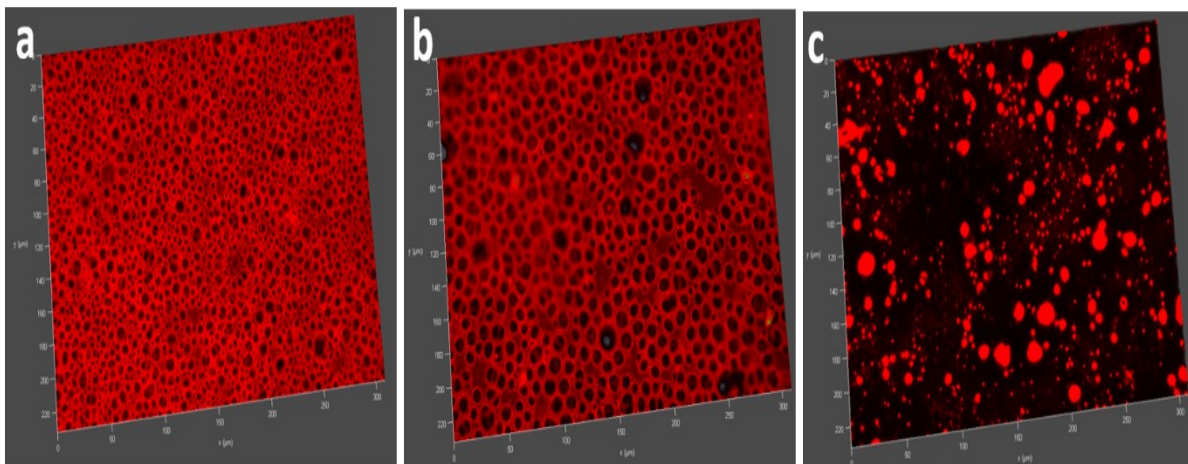


Fig. S2 Z-stacked Fluorescence microscopic images of P3HT-PCBM films with different ratio a) 0.75:1, b) 0.5:1, and c) 0.25:1

Section S4. Spectroscopic studies of P3HT, PCBM, and different ratios of P3HT-PCBM:

After taking UV-Visible absorption spectra of P3HT, PCBM, and different ratio of P3HT-PCBM in solutions (Fig S3), we have studied aggregation behaviour of P3HT-PCBM solution by changing the PCBM concentration after 24 hours of time with photoluminescent analysis (excitation at 449nm) and represented in Figure S4a, S4b, and S4c. Li et al reported that, ordered aggregation of P3HT-PCBM can be formed in solvent mixture and ageing time can be calculated by optical analysis.[1] In our case, as demonstrated in Fig. S4a, the distinguishable vibronic emission peaks of P3HT polymer appeared in P3HT:PCBM at 578nm and 625nm plausibly due to the transfer/recombination of the electron and holes.

When P3HT is mixed with different amount of PCBM in solution form, significant quenching was observed in P3HT emission intensity of P3HT-PCBM solutions (1:0 to 1:0.75) (Fig) attributed to the intermolecular charge transfer, rather than produce emission.[2] But beyond the 0.75:1 ratio solution the PL intensity increased, especially in 0.5:1 ratio solution (from 512 to 604 showed in Fig S4c, PL intensity plot), plausibly indicating the higher degree of phase separation and increased aggregation of P3HT-PCBM due to the large amount of PCBM.

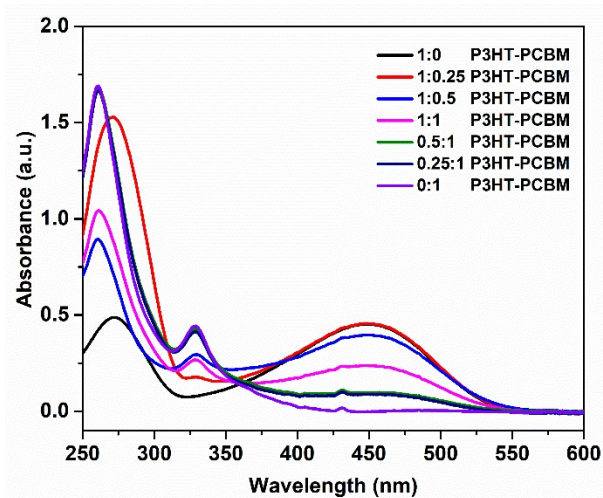


Fig. S3 UV-Visible absorption spectra of P3HT, PCBM, and different ratio of P3HT-PCBM solutions

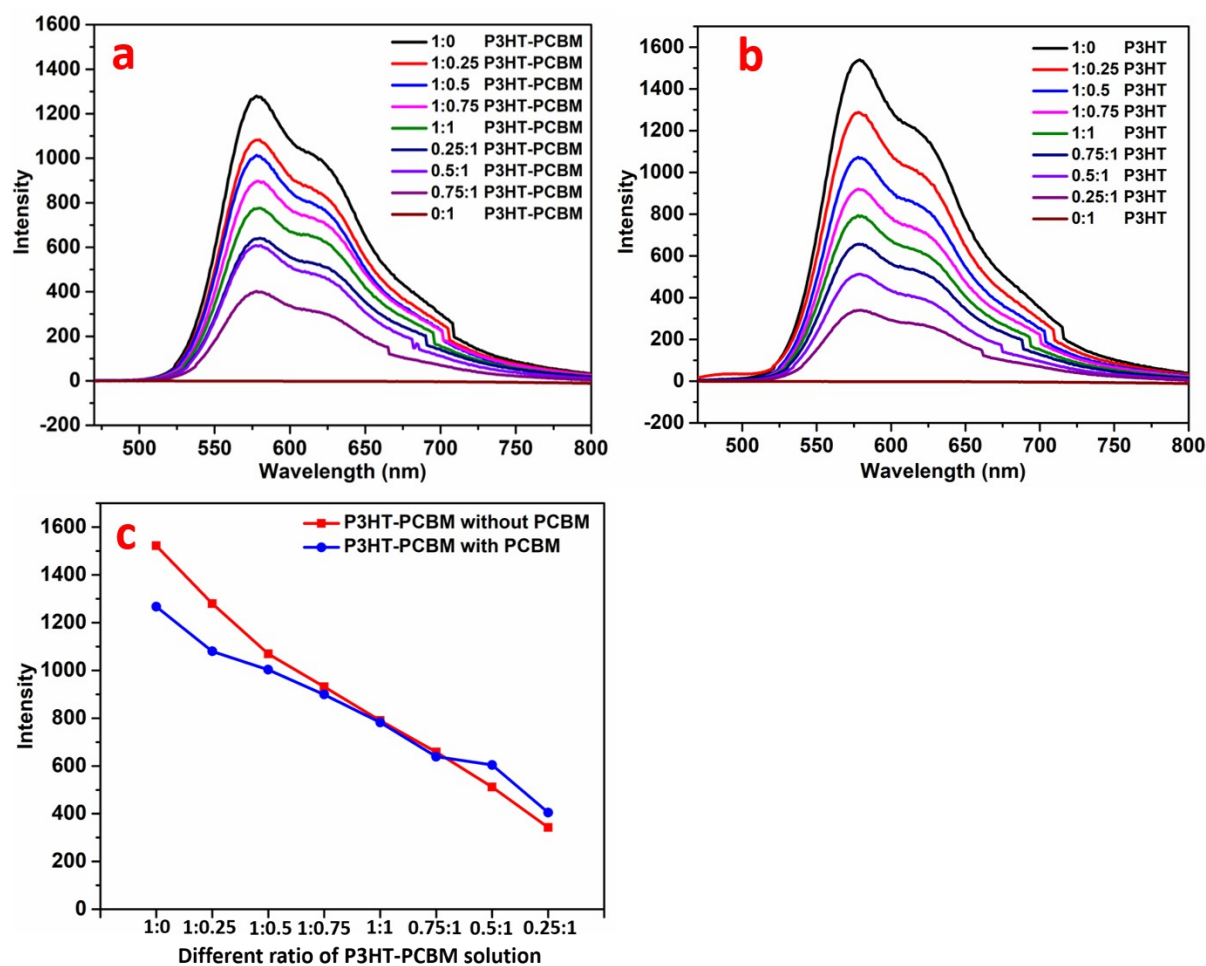
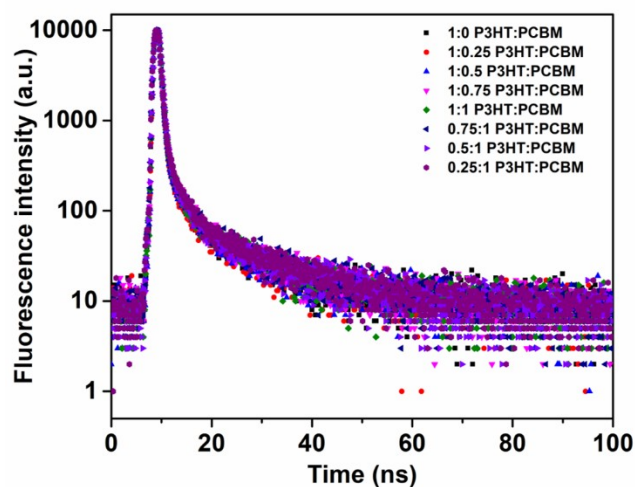


Fig. S4 (a) Photoluminescence spectra of P3HT-PCBM solutions after 24 hours time from preparation time (b) Photoluminescence spectra of P3HT-PCBM solutions without PCBM and (c) PL intensity plot of varying ratio of P3HT-PCBM solution (2mg/L concentration in chloroform) without and with PCBM



S.No	P3HT-PCBM solution	Emission at 578nm	
		C1 (ns), (%)	C2 (ns), (%)
1	1:0	0.47 ns, (93.7%)	10.5 ns, (6.2%)
2	1:0.25	0.47 ns, (94.4%)	11.47 ns, (5.5%)
3	1:0.5	0.47 ns, (93.5%)	11.4 ns, (6.4%)
4	1:0.75	0.48 ns, (93.0%)	10.35 ns, (7.0%)
5	1:1	0.47 ns, (93.0%)	10.91 ns, (7.0%)
6	0.75:1	0.47 ns, (93.0%)	10.62 ns, (6.9%)
7	0.5:1	0.49 ns, (94.3%)	12.29 ns, (5.6%)
8	0.25:1	0.50 ns, (93.5%)	10.40 ns, (6.4%)

Figure S5. Transient photoluminescence spectra of P3HT-PCBM solutions (left) and Table S2: lifetime values of P3HT-PCBM solutions (2mg/L concentration in chloroform) with different ratio(right)

Section S5: High resolution microscopic studies of P3HT, PCBM, and different ratios of P3HT-PCBM: To find out detailed morphology on the surfaces of the composites after drop-casting, scanning electron microscopy (SEM) and atomic force microscopy (AFM) were used separately and provided below.

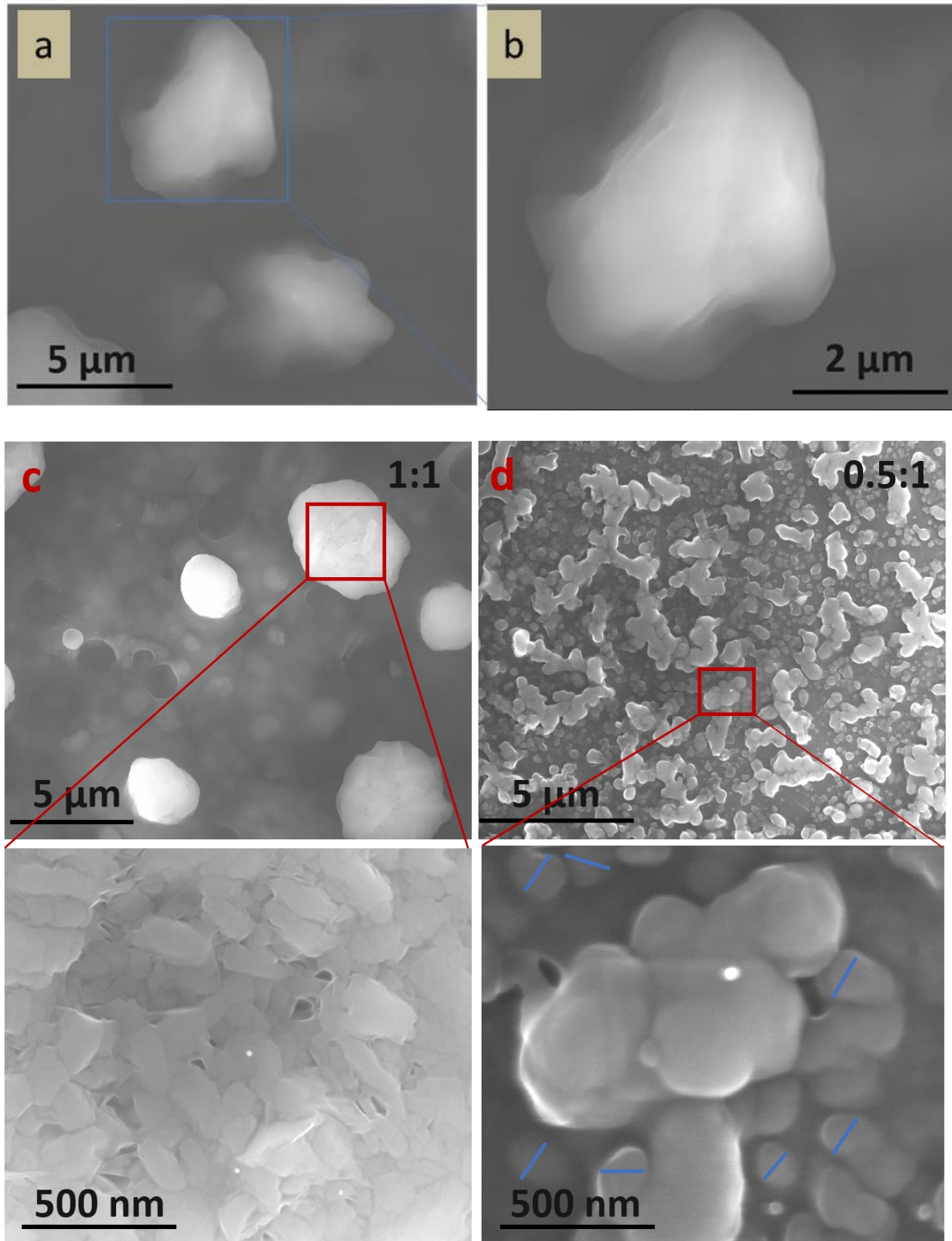


Fig. S6 (a) SEM image of pristine P3HT film in lower magnification (b) SEM image of pristine P3HT film in lower magnification different magnification SEM images of P3HT-PCBM films in a different ratio (c) 1:1 and (d) 0.5:1; Corresponding zoomed images have been shown below them

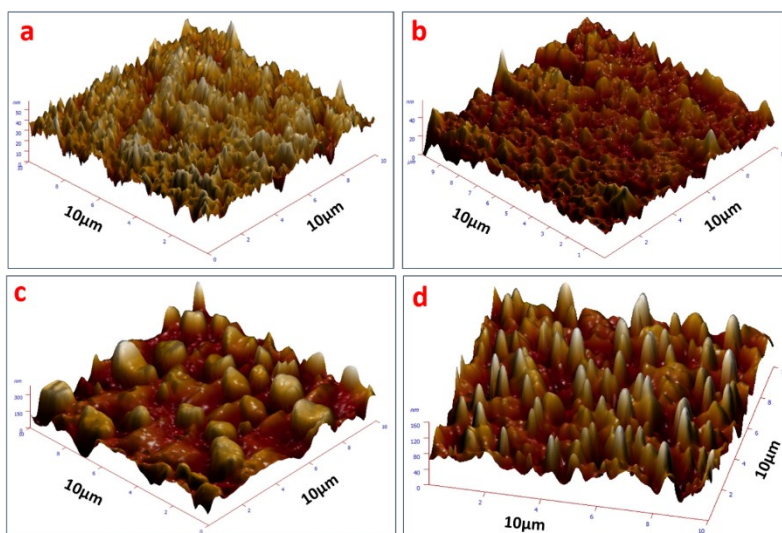


Fig. S7 AFM topographic 3D images of a) 1:0 (P3HT film), b) 1:1 ratio, c) 0.5:1 ratio P3HT-PCBM films and d) 0:1 (PCBM film)

Section S6: X-Ray diffraction analysis (XRD): XRD analysis of P3HT-PCBM films was carried out by PANalytical XRD instrument to find out the crystalline sizes and some of the data were provided below.

In order to analyse the microstructure of P3HT polymers by changing the PCBM loads in P3HT-PCBM blends, we performed the X-ray diffraction analysis (Fig. S8 (a) and (b) and table S3). The pristine P3HT polymer film shows two sets of diffraction patterns due to the lamellar stacking of P3HT backbones (5.4° (100), 11.03° (200), 16.3° (300)) and π - π stacking of polythiophene (23.4° (010)). Earlier it was observed that the crystallinity of P3HT in P3HT-PCBM films was decreased by increasing the PCBM concentration. In our case also (1:1 ratio) similar observations were noticed with the reduction of peak intensity of P3HT peak at 10.57° (200 plane). But In the case of P3HT-PCBM film with a 0.5:1 ratio, the increment of peak intensity (1336) as well as higher crystallite size (24.63 nm) of P3HT was observed belongs to peak at 10.53° (200 plane) compared to 1:1 ratio (intensity is 855 and crystallite size is 16.42 nm) (Fig. S8 (b) inset picture) indicating the high crystallinity due to the alignment of P3HT polymers induced by PCBM molecules of particular concentration. This property can influence the transportation of charges as well as catalytic efficiency.

Table S3. X-Ray diffraction analysis parameters of P3HT-PCBM composite films (1:0, 1:1 and 0.5:1 ratio)

P3HT-PCBM	Plane	Two Theta	Intensity	FWHM	d	Crystallite
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Surfaces		position (°)	height		spacing	size (nm)
1:0	200	11.03	1685	0.6770	8.01	12.32
1:1	200	10.57	855	0.5077	8.36	16.42
0.5:1	200	10.53	1336	0.3385	8.39	24.63

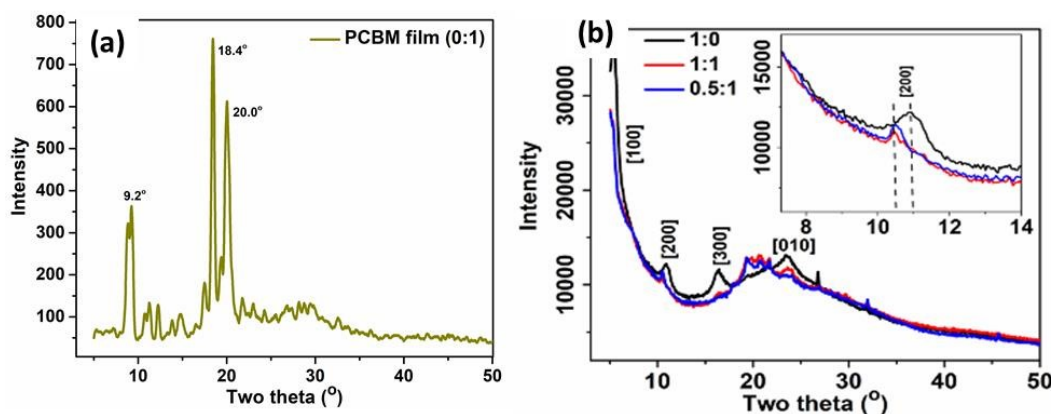


Fig. S8 XRD pattern of (a) PCBM film and (b) combined XRD pattern of P3HT-PCBM films (1:0, 1:1 and 0.5:1 ratio)

Section S7: Wettability studies: Contact angle meter was used to study the hydrophobicity or hydrophilicity of the surfaces and the data were provided below. The surface wettability of P3HT-PCBM films with different ratios was studied by a water contact angle meter and represented in ESI Fig. S9. It can be noted that pure P3HT (1:0) and PCBM (0:1) exhibit a contact angle of 106.4° and 83.6° respectively. These values indicated that PCBM has high surface energy to exhibit hydrophilic character resulting from the presence of polar ester group whereas P3HT possesses hydrophobic nature due to the presence of the alkyl chains and thiophene rings. In this context, it may be noted that high catalytic efficiency can be achieved by reducing hydrophobicity (86.6°) to enhance the surface contact between dye molecules and catalyst sites. Hence we observed that at a ratio of 0.5:1 (P3HT-PCBM) film possessed greater hydrophilic property due to the high contents of PCBM to introduce better catalytic activity.

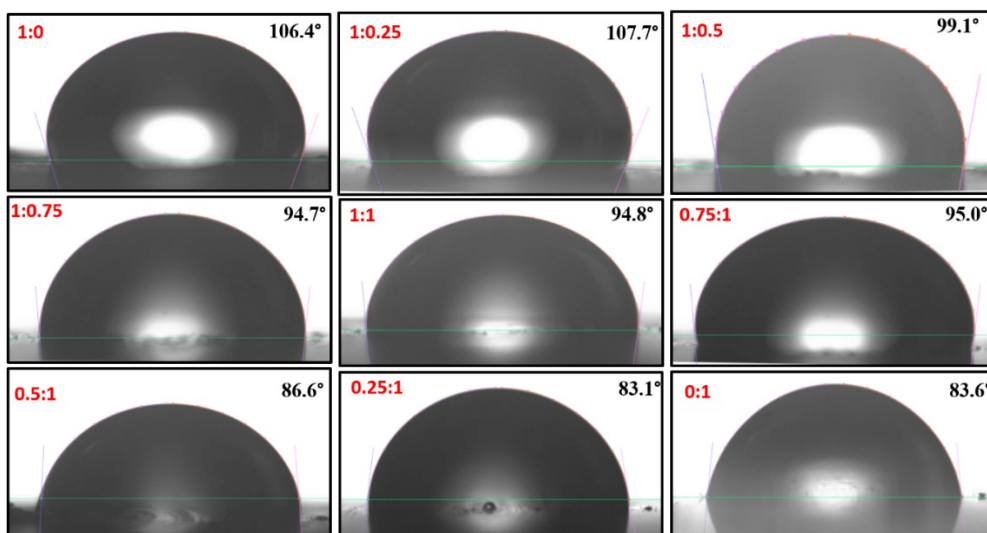


Fig. S9 Water droplet images and water contact angle values of P3HT, PCBM and P3HT-PCBM films (different ratios)

Section S8: Photocatalytic experiments: photocatalytic dye degradation experiments were performed under sunlight as well as using mercury xenon lamp of 200 watts for dyes like rhodamine B and methylene blue and some of the detailed results are presented below.

Preparation of Rhodamine B (Rh-B) dye solution:

The Rhodamine B dye solution was prepared by using this calculation and the different concentration of dye solution has been obtained.

$$V_1N_1=V_2N_2; \quad V_2=(V_2N_2)/N_1$$

Where, V_1 = initial volume; V_2 = Final volume

N_1 = Initial concentration; N_2 = Final concentration

120 mg of Rh-B dye was dissolved in 200 mL of distilled water to give 500 ppm of Rh-B dye solution. From the 500 ppm stock solution, 20 mL of Rh-B was made up to 100 mL in a standard metric flask using distilled water to give 100 ppm stock solution.

Standard curve of Rh-B dye solution:

Rhodamine-B dye solutions of concentrations varying from 0.125,0.25,0.5,1-12ppm was subjected to UV analysis to determine the absorption. All UV-visible absorption measurements were performed with Shimadzu, UV-2450, UV-VIS Spectrophotometer in the wavelength range of 200–800 nm. The standard curve of Rh-B dye was plotted using Origin Pro 8.5 and it is represented in Fig. S10.

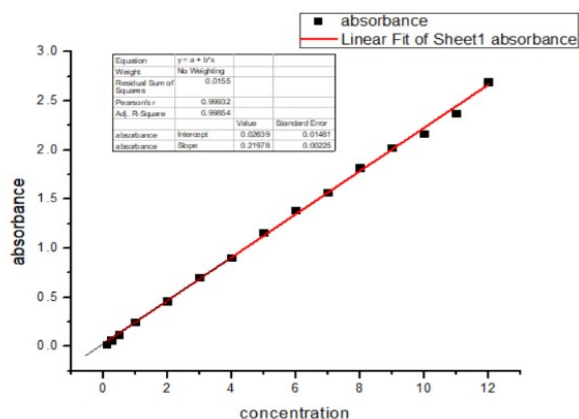


Fig. S10 Standard curve of Rh-B dye

Photocatalytic experiments

For evaluating the photocatalytic activity of P3HT-PCBM films, we have chosen rhodamine B dye as a model organic pollutant. In a beaker, we took 5 mL of dye solution (2 ppm), and the P3HT-PCBM film (5 mg catalyst) was kept inside the beaker containing the dye solution. The reaction medium was kept in dark conditions for 2 hours to achieve equilibrium between dye molecules and P3HT-PCBM film surfaces. Then the dye solution was kept under sunlight irradiation (with light intensity of 186 w/m²) or Mercury xenon lamp (200W) for respective hours. After irradiation the concentration of dye solutions was determined by UV-Visible spectrophotometric method and calculated the removal of dye in-terms of percentage by the following equation: percentage removal = $(C_i - C_f) / C_i \times 100$ Where, C_i = initial concentration of dye, C_f = final concentration of dye.

The photocatalytic degradation of Rh-B was carried out under different optimization conditions: (a) Effect of initial concentration of Rh-B (b) By varying ratio of P3HT-PCBM composite film (c) By varying the time period of irradiation.

Effect of initial concentration of Rh-B:

From the 100 ppm stock solution, the Rh-B solutions of 1, 2, 3, 4, 5 and 6 ppm were prepared in a 25 mL standard metric flask using distilled water. The photocatalytic degradation was carried out with 5 mL of Rh-B dye solution in 50 mL beaker. P3HT-PCBM (1:1) film (It contains 5 mg catalyst) photocatalyst was immersed in a dye solution. The dye solution along with the P3HT-PCBM films were exposed to the sunlight for the time period of 2 hours. After 2 hours, the dye solution was removed and measured the absorption value to find the

final concentration of dye solution. The we calculated the removal of dye (%) by using the above-mentioned equation.

Table S4. Removal of dye (%) of Degradation of Rh-B dye with varying concentration using P3HT:PCBM films

S.No	Initial concentration of dye (ppm)	Removal of dye (%)
1	1	76.9
2	2	52.9
3	3	49.7
4	4	46.7
5	5	35.5
6	6	29.5
7	Control-2ppm	15

Effect of varying ratio of P3HT-PCBM:

Rh-B dye solution of 2 ppm were prepared in 25 mL standard metric flask. 5mL of Rh-B dye solution was treated with P3HT-PCBM films of varying ratios 1:0, 1:0.25, 1:0.5, 1:0.75, 1:1, 0.75:1, 0.5:1 and 0.25:1. The P3HT-PCBM films was immersed in dye solution and it was exposed to different condition like, Mercury xenon lamp (200W) or natural sunlight for the time period of 2 hours. Finally, we calculated the removal of dye (%) by measuring the final concentration of dye solution after 2 hours of irradiation.

Table S5. Removal of dye (%) plot of Rh-B dye degradation with using varying ratio of P3HT:PCBM films

S.No	Ratio of P3HT-PCBM films	Removal of dye (%)	
		Mercury Xenon lamp irradiation	Sunlight irradiation
1	1:0	43.9	60
2	1:0.25	39.8	44.4
3	1:0.5	44.6	48.6
4	1:0.75	38.4	34.1
5	1:1	41.9	49.8
6	0.75:1	48.1	77.8
7	0.5:1	63.5	95.1
8	0.25:1	47.3	36.3

Effect of Time Period :

5 mL of Rh-B dye solution was taken in 50 mL beaker and the P3HT-PCBM (1:1 ratio or 0.5:1 ratio) films were immersed in the dye solution. It was exposed to natural sunlight for the time period of 30, 60, 90, 120, 150 and 180 mins. After that, the dye solution was removed and calculated the C_t/C_0 values.

Table S6. Removal of dye (%) plot of Rh-B dye degradation with varying irradiation time using P3HT:PCBM films

S.No	Irradiation time (min)	C_t/C_0 value	
		1:1 ratio P3HT-PCBM film	0.5:1 ratio P3HT-PCBM film
1	0	1	1
2	30	0.803	0.664
3	60	0.72	0.305
4	90	0.616	0.168
5	120	0.492	0.072
6	150	0.436	0.031
7	180	0.243	0.005

kinetic study of degradation reaction

The kinetic studies of photocatalytic degradation of Rh-B was helpful to determine the reaction rate with respect to irradiation time using the Langmuir–Hinshelwood (L–H) equation,

Pseudo first order reaction rate:

$$\ln \frac{(C_0)}{(C_f)} = kt$$

Where C_0 = Initial concentration of dye (ppm)

C_f = Final concentration of dye (ppm)

K = Rate constant

T = Time (min)

Table S7. Kinetic study data

S.No	Irradiation time (min)	Ln[C ₀]/[C _f] value	
		1:1 ratio P3HT-PCBM film	0.5:1 ratio P3HT-PCBM film
1	0	0	0
2	30	0.219	0.409
3	60	0.327	1.187
4	90	0.484	1.783
5	120	0.709	2.624
6	150	1.059	3.473
7	180	1.414	5.298

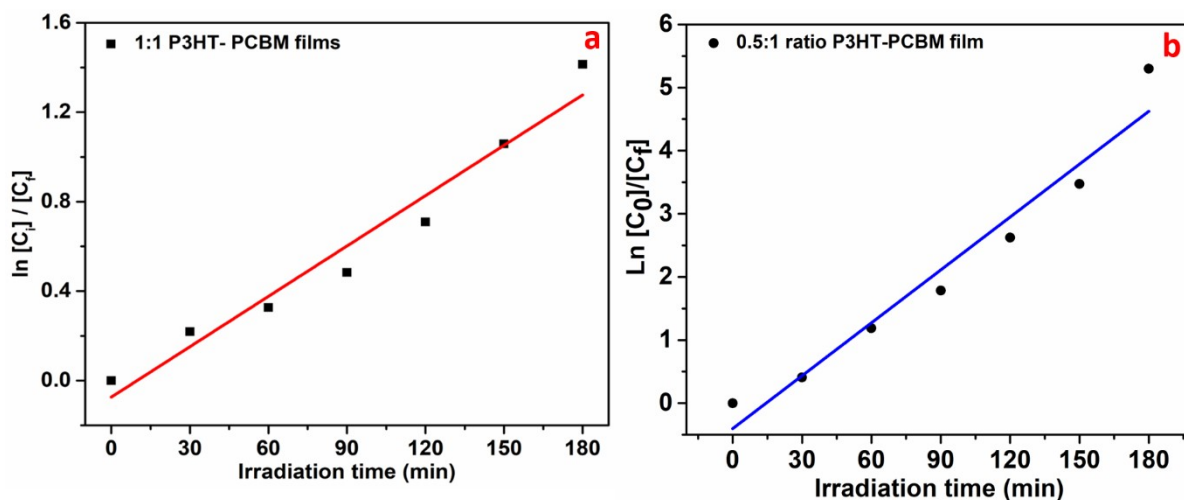


Fig. S11 Kinetic plot of RhB dye degradation using P3HT-PCBM films (a) 1:1 ratio and (b) 0.5:1 ratio

Recyclability study:

The recyclability experiment of dye degradation using P3HT-PCBM film (0.5:1 ratio) were performed upto three cycles. For first cycle, initially we took it 5mL of Rh-B dye solution in a beaker and immersed the P3HT-PCBM film with 0.5:1 ratio. Then it was

exposed to natural sunlight for the time period of 2 hours. Finally, we calculated the removal of dye (%) by measuring the final concentration of dye solution after 2 hours of irradiation. Then the film catalyst was washed with water in three times and directly used for further cycles. This same procedure was followed for second and third cycle of dye degradation experiment.

Table S8. Removal of dye (%) plot of Rh-B dye degradation in recyclability experiment using 0.5:1 ratio P3HT: PCBM film

S.No	Cycles	Removal of dye (%)
1	1 st	95.1%
2	2 nd	93.8%
3	3 rd	79.8%

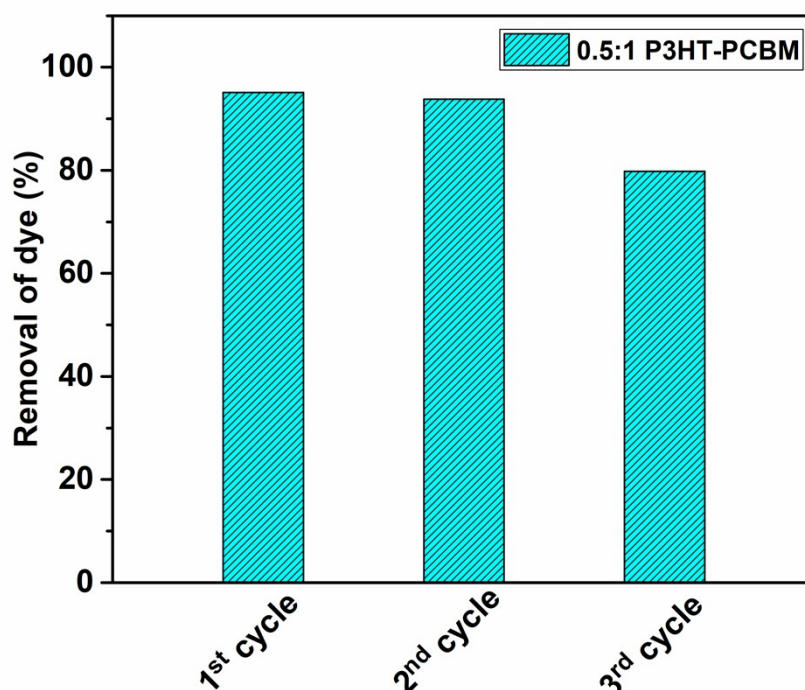


Fig. S12 percentage removal of dye in a recyclability experiment of dye degradation using 0.5:1 ratio of P3HT-PCBM film

Table S9. Comparison of photocatalytic efficiency (from our results) and photovoltaic efficiency (from previous literature report) of P3HT-PCBM films with different weight ratio of P3HT and PCBM content

From Our work: Photocatalytic efficiency	Photovoltaic efficiency from Literature (F.C.Chen et al) [3]
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In terms of PCBM content (%)	Photocatalytic efficiency (%)	In terms of PCBM content (%)	V _{oc} (V)	J _{sc} (mA/cm ²)	FF (%)	Photovoltaic efficiency PCE (%)
0	60					
20	44.4	25	0.60	4.33	42.8	1.11
33.3	48.6	40	0.60	10.10	53.1	3.22
42.9	34.1	50 (1:1)	0.60	9.53	64.0	3.66**
50	49.5	60	0.60	6.87	60.7	2.50
57	77.8	75	0.60	2.48	45.1	0.67
66.7(0.5:1)	95.1*					
80	36.3					

**Highest efficiency in photocatalysis*

***Highest efficacy in photovoltaics*

Table S10. Literature survey for metal free-polymer based photocatalyst systems for degradation of organic pollutants:

S.No	Polymer catalysts	Application in degradation of organic pollutants	conditions	output	reference
1	Conjugated polyimide polymers (PI-1, PI-2 & PI-3) from melamine and three dianhydride molecules	Methyl orange dye	Visible light irradiation ($\lambda=420$ nm)	Complete degradation ($\approx 100\%$) within 3 hours	[4] Chu et al, 2014
2	Nano and bulk structures of Poly(diphenylbutadiene)(PDPB)	Methyl orange & phenol	UV light & visible light	82% degradation rate with 270 min using nano PDPB	[5] Ghosh et al, 2015
3	poly(3,4-ethylenedioxythiophene) (PEDOT)	Methyl orange & phenol	UV light & visible light	100% degradation rate with 240 min using nano PEDOT under visible light irradiation	[6] Ghosh et al, 2015
4	Graphitic carbon nitride (g-C ₃ N ₄) polymer by dicyandiamide and melamine	Methylene blue	Visible light	Above 90% within 120 min	[7] Xu et al, 2015
5	Perylene diimide based porous conjugated polymer	Methylene blue	UV light	High rate (≈ 0.25 c/co) using PCP2-100%PDI catalyst	[8] Li et al, 2016
6	Naphthalene based polyimide	RhB dye	LED light & sunlight	Complete degradation ($\approx 100\%$) within	[9] Liu et al, 2017

				30 min	
7	poly(1,3,4-oxadiazole)s (POD)	Methyl orange, Methylene blue & RBB	Hg lamp	Complete degradation of RBB dye within 15 min	[10] Ran et al, 2018
8	polydiacetylene based conjugated nanoporous covalent organic polymer	Methyl orange	UV light & visible light	92% degradation with in 120 min under visible light	[11] Bhowmik et al, 2018
9	triazine-based covalent organic polymer	Methyl orange, Methylene blue & RhB	Visible light	67% MO, 78% RhB and 57% MB degradation under visible light	[12] Xu et al, 2018
10	Carbonitride grafted polyaniline	Methylene blue	Visible light	Complete degradation of MB dye within 30 min	[13] Vellaichamy et al, 2018
11	Polypyrrole nanostructures	Phenol	UV and Visible light	Complete degradation of RBB dye in 4.5 hours	[14] Yuan et al, 2018
12	Triptycene based imine linked polymer	RhB dye	Sunlight	95% degradation within 160 min	[15] Preet et al, 2019
13	Novel conjugated polymers with sulfone, sulphur & thioether bonds	RhB, Methylene blue, & Rh6G	Visible light	Rhodamine B (RhB) dye degradation is ~100% with TpSD	[16] Cao et a, 2019
14	Porphyrin containing polyimide	Methyl orange	Visible light	Maximum 84% degradation in 8 hours	[17] Cui et al, 2019
15	P3HT-PCBM film (0.5:1 ratio)	RhB dye	Sunlight & mercury-xenon lamp	95.1% degradation after 120 min	This work

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