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Electronic supplementary information

General Methods

^{1}H and ^{13}C NMR spectra were recorded on a Bruker ARX 200 or ARX 300 MHz Brucker spectrometer. Chemical shifts for ^{1}H NMR spectra are referenced relative to residual protium in the deuterated solvent ($CDCl_3$ δ = 7.26 ppm, MeOD δ = 3.31 ppm, CD_3CN δ = 1.94 ppm). Mass spectra were recorded on a EI-MS HP 5989A spectrometer or on a JMS-700 (JEOL LTD, Akishima, Tokyo, Japan) double focusing mass spectrometer of reversed geometry equipped with electrospray ionization (ESI) source. Fast atom bombardement mass spectroscopy (FAB-MS) analyses were performed in m-nitrobenzyl alcohol matrix (MBA) on a ZAB-HF-FAB spectrometer. MALDI-TOF analyses were performed on an Applied Biosystems Voyager DE-STR spectrometer in positive linear mode at 20 kV acceleration voltage with a-cyano 4-hydroxycinnamic acide (CHCA) as matrix.

Thin-layer chromatography (TLC) was performed on aluminium sheets precoated with Merck 5735 Kieselgel $60F_{254}$. Column chromatography was carried out either with Merck 5735 Kieselgel 60F (0.040-0.063 mm mesh) or with SDS neutral alumina (0.05-0.2 mm mesh). Air sensitive reactions were carried out under argon in dry solvents and glassware. Chemicals were purchased from Aldrich and used as received. Compounds tetrakis(trisphenyl)phosphine palladium[1], 4-bromo-2,2'-bisthiophene[2], 2,6-di-2-pyridyl-4(1H)-pyridone[3], 4'-trimethylstannyl-2,2':6',2''-terpyridine[4], 2,2':6',.2"-terpyridine, 4'-diethylphosphonate-2,2':6',2''-terpyridine[5], 4'-diethylphosphonate-2,2':6',2''-terpyridine ruthenium (III) trichloride[5], and 4'-[[(trifluoromethyl)sulfonyl]oxy]-2,2':6',2''-terpyridine[3] were prepared according to literature methods.

Preparation of the sensitizers

3,4-dioctylthiophene (**2**)

2.2 g (90.6 mmol.) of magnesium turnings and 5 mL of dry diethylether were introduced into a three-necked flask equipped with a dropping funnel and a condenser. A few drops of pure 1-bromooctane (15 g, 78.2 mmol.) were added from the funnel. As soon as the reaction started, the remaining 1-bromooctane was diluted with 60 mL of dry diethylether and added dropwise. After complete addition, the medium was stirred at room temperature under nitrogen atmosphere for 1 hour. Then, the Grignard reagent was transferred by means of the double-ended needle technique to a solution of 5 g (20.5 mmol.) of 3,4-dibromothiophene **1** and 31 mg (0.056 mmol.) of [1,3-bis(diphenylphosphino)propane] nickel(II) dichloride in 95 mL of dry diethylether. After the addition, the mixture was stirred at room temperature under nitrogen atmosphere for 12 hours. Magnesium salts were precipitated with a large excess of petroleum ether. After filtration, the solvent was evaporated and the residue was quickly eluted on a silica gel column with petroleum ether. The product was distilled in vacuum (bp. 150°C/0.4 mmHg) to give 4.1 g (64%) of 3,4-dioctylthiophene.

^{1}H RMN ($CDCl_3$, 300 MHz) : δ = 0.94 (m, 6H), 1.33 (m, 20H), 1.66 (m, 4H), 2.55 (t, 4H), 6.92 (s, 2H).

^{13}C RMN ($CDCl_3$, 300 MHz) : δ = 14.26, 22.86, 28.98, 29.48, 29.69, 29.82, 32.08, 120.01, 142,16

MS (EI): *m/z* (%)= 309 ($M+H^+$) (100).

2,5- dibromo-3,4-dioctylthiophene (**3**)

In absence of light and at 0 °C, a solution of NBS (1.9 g, 10.5 mmol.) in degassed and dry DMF (15 mL) was added dropwise to a solution of 3,4-dioctylthiophene (1.3 g, 4.2 mmol.) in degassed and dry DMF (20 mL). After 5 hours at room temperature, the mixture was poured onto ice and extracted with diethylether. The organic layers were combined, washed with a saturated aqueous $NaHCO_3$ solution, with water and dried over magnesium sulfate. Purification was performed by column chromatography on silica gel with petroleum ether as eluent and afforded 1.5 g (77%) of a colourless liquid.

^{1}H RMN ($CDCl_3$, 300 MHz) : δ = 0.90 (m, 6H), 1.20-1.60 (m, 24H), 2.53 (t, 4H).

^{13}C RMN ($CDCl_3$, 300 MHz) : δ = 14.28, 22.86, 29.12, 29.42, 29.50, 29.72, 32.06, 107.99, 141,52

MS (EI): *m/z* (%) = 466 (M^+) (28), 387 (33), 289 (62), 191 (100).

3',4'-dioctyl-2,2':5',2''-terthiophene (**4**)

0.27 g (11 mmol.) of magnesium turnings and 1 mL of dry diethylether were introduced into a three-necked flask equipped with a dropping funnel and a condenser. A few drops of pure 2-bromothiophene (1.33 g, 8.2 mmol.) were added from the funnel. As soon as the reaction started, the remaining 2-bromothiophene was diluted with 45 mL of dry diethylether and added dropwise. After complete addition, the medium was stirred at reflux under nitrogen atmosphere for 1 hour. Then, the Grignard reagent was transferred by means of the double-ended needle technique to a cold (-78 °C) solution of 0.93 g (2 mmol.) of 2,5-dibromo-3,4-dioctylthiophene and 30 mg (0.04 mmol.) of [1,1'-bis(diphenylphosphino)ferrocene] palladium (II) dichloride in 30 mL of dry diethylether. After the addition, the mixture was stirred at room temperature under nitrogen atmosphere for 20 hours. Then, the mixture was cooled to 5 °C and a diluted solution of HCl was added. The aqueous layer was extracted with diethylether. The organic layers were combined, washed with water and dried over magnesium sulfate. The crude product was purified by column chromatography on silica gel and eluted with petroleum ether. A second column chromatography on reverse silica gel with petroleum ether was yielded 0.67 g (70%) of pure 3',4'-dioctyl-2,2':5',2''-terthiophene.

^{1}H RMN ($CDCl_3$, 300 MHz) : δ = 0.92 (m, 6H), 1.20-1.60 (m, 24H), 2.75 (t, 4H), 7.08 (dd, 2H, ^3J = 3.6 Hz, ^3J = 5.1 Hz), 7.16 (dd, 2H, ^3J = 3.6 Hz, ^4J = 1.2 Hz), 7.33 (dd, 2H, ^3J = 5.1 Hz, ^4J = 1.2 Hz).

^{13}C RMN ($CDCl_3$, 300 MHz) : δ = 14.13, 22.70, 28.14, 29.27, 29.91, 30.77, 31.89, 125.27, 125.84, 127.33, 129.81, 136.23, 140.11

MS (EI): *m/z* (%)= 472 (M^+) (100).

5-bromo-3',4'-dioctyl-2,2':5',2''-*ter*thiophene (**5**)

In absence of light and at –20 °C, a solution of NBS (0.14 g, 0.79 mmol.) in chloroform (20 mL) was added dropwise, under argon, over 1 hour to a solution of 3',4'-dioctyl-2,2':5',2''-terthiophene (0.47 g, 0.99 mmol.) in degassed CS_2 (20 mL). The mixture was stirred at this temperature for 1 h 30 and to 0 °C for 30 min. The medium was poured onto cold ammonium chloride solution and extracted with dichloromethane. The organic layers were combined, washed with a saturated aqueous $NaHCO_3$ solution, with water and

dried over magnesium sulfate. Purification was performed by column chromatography on silica gel with petroleum ether and afforded 0.29 g (52%) of a yellow pale liquid.
^{1}H RMN ($CDCl_3$, 300 MHz) : δ = 0.90 (m, 6H), 1.20-1.60 (m, 24H), 2.66 (m, 4H), 6.85 (d, 1H, ^3J = 3.8 Hz), 6.98 (d, 1H, ^3J = 3.8 Hz), 7.03 (dd, 1H, ^3J = 3.6 Hz, ^3J = 5.1 Hz), 7.11 (dd, 1H, ^3J = 3.6 Hz, ^4J = 1 Hz), 7.28 (dd, 1H, ^3J = 5.1 Hz, ^4J = 1 Hz).
^{13}C RMN ($CDCl_3$, 300 MHz) : δ = 14.19, 22.75, 28.14, 29.30, 29.90, 30.79, 31.94, 111.77, 125.48, 126.05, 127.38, 127.77, 128.84, 130.18, 130.43, 135.98, 137.81, 140.12, 140.61
MS (EI): *m/z* (%)= 550 (M^+) (100).

4'-[(2,2'-bisthiophen)-5-yl]-2,2':6',2''-terpyridine (**7**)
A Schlenk tube was charged with 100 mg (0.25 mmol.) of 4'-trimethylstannyl-2,2':6',2''-terpyridine and 93 mg (0.37 mmol.) of 5-bromo-2,2'-bisthiophene in 15 mL of dry dioxane. The mixture was degassed by pumping and flushing with argon on the vacuum line and then 26 mg (0.025 mmol.) of tris(dibenzylideneacetone)dipalladium, 85 mg (0.55 mmol.) of cesium fluoride and 18 mg (0.09 mmol.) of tri-*tert*-butylphosphine was added. The mixture was heated to 130 °C for 16 hours. After cooling to room temperature, the solution was diluted with ethyl acetate and washed with diluted ammoniac solution and water. The organic layer was dried over $MgSO_4$, filtered and evaporated in vacuum. Purification was performed by column chromatography on silica gel with dichloromethane/diethylether (100/0 to 80/20) as eluent and afforded 72 mg (72%) of an orange solid.
^{1}H RMN ($CDCl_3$, 300 MHz) : δ = 7.06 (dd, 1H, ^3J = 4.8 Hz, ^{3}J = 3.6 Hz), 7.26 (m, 3H), 7.37 (ddd, 2H, ^3J = 7.8 Hz, ^3J = 4.8 Hz, ^4J = 1.8 Hz), 7.70 (d, 1H, ^3J = 3.6 Hz), 7.88 (ddd, 2H, ^3J = 7.8 Hz, ^3J = 7.2 Hz, ^4J = 1.8 Hz), 8.65 (d, 2H, ^3J = 7.8 Hz), 8.67 (s, 2H), 8.75 (m, 2H).
^{13}C RMN ($CDCl_3$, 300 MHz) : δ = 116.62, 121.31, 123.91, 124.25, 124.67, 125.03, 126.55, 128, 136.84, 137.06, 139.05, 140.25, 142.98, 149.12, 155.97, 156.04
MS (EI): *m/z* (%) = 397 (M^+) (100).

4'-[(3',4'-dioctyl-2,2':5',2''-*ter*thiophen)-5-yl]-2,2':6',2''-terpyridine (**8**)
A Schlenk tube was charged with 100 mg (0.25 mmol.) of 4'-trimethylstannyl-2,2':6',2''-terpyridine and 200 mg (0.38 mmol.) of 3',4'-dioctyl-2,2':5',2''-terthiophene in 15 mL of dry dioxane. The mixture was degassed by pumping and flushing with argon on the vacuum line and then 26 mg (0.025 mmol.) of tris(dibenzylideneacetone)dipalladium, 85 mg (0.55 mmol.) of cesium fluoride and 18 mg (0.09 mmol.) of tri-*tert*-butylphosphine was added. The mixture was heated to 130 °C for 16 hours. After cooling to room temperature, the solution was diluted with ethyl acetate and washed with diluted ammoniac solution and then with water. The organic layer was dried over $MgSO_4$, filtered and evaporated in vacuum. The crude product was purified by column chromatography on silica gel with dichloromethane/diethylether (100/0 to 80/20) as eluent and afforded 143 mg (80%) of an orange solid.
^{1}H RMN ($CDCl_3$, 300 MHz) : δ = 0.87 (m, 6H), 1.28-1.62 (m, 24H), 2.71 (t, 4H), 2.78 (t, 4H), 7.07 (dd, 1H, ^3J = 3.6 Hz, ^3J = 5.1 Hz), 7.17 (dd, 1H, ^3J = 3.6 Hz, ^4J = 1.2 Hz), 7.19 (d, 1H, ^3J = 3.9 Hz), 7.33 (dd, 1H, ^3J = 5.1 Hz, ^4J = 1.2 Hz), 7.37 (ddd, 2H, ^3J = 7.5 Hz, ^3J = 4.8 Hz, ^4J = 1.2 Hz), 7.74 (d, 1H, ^3J = 3.9 Hz), 7.88 (ddd, 2H, ^3J = 7.5 Hz, ^3J = 7.8 Hz, ^4J = 1.5 Hz), 8.62 (d, 2H, ^3J = 7.8 Hz), 8.68 (s, 2H), 8.74 (m, 2H).
^{13}C RMN ($CDCl_3$, 300 MHz) : δ = 14.09, 22.67, 28.12, 29.26, 29.89, 30.61, 30.70, 31.86, 116.70, 121.28, 123.84, 125.41, 125.92, 126.22, 126.60, 127.37, 129.50, 130.42, 136.05, 136.78, 138.17, 140.29, 140.68, 140.94, 143.09, 149.08, 156.01
MS (EI): *m/z* (%) = 703 (M^+) (71).

4'-phosphonic acid-2,2':6',2''-terpyridine-(2,2':6',2''-terpyridine) ruthenium (II) di-hexa fluorophosphate (**13**)
40 mg (0.076 mmol.) of 4'-phosphonic acid-2,2':6',2''-terpyridine ruthenium (III) trichloride, 20 mg (0.084 mmol.) of terpyridine and 0.1 mL (0.4 mmol.) of triethylamine was heated to reflux in ethanol (10 mL) and water (2.5 mL) for 50 minutes under argon. After cooling to room temperature, the solvent was removed and the residue was dissolved in a minimum of ethanol. Acetone was added to precipitate a red solid. The product was redissolved in water and treated with aqueous HPF_6 solution to precipitate complex **13** as a red solid (50 mg, 85%).
^{1}H RMN (MeOD, 300 MHz) : δ = 7.16 (m, 4H), 7.37 (2d, 4H, ^3J = 5.5 Hz), 7.90 (m, 4H), 8.40 (t, 1H, ^3J = 8.4 Hz), 8.62 (m, 2H), 8.88 (d, 2H, ^3J = 8.1 Hz), 9.02 (d, 2H, ^3J = 12 Hz).
^{13}C RMN (CD_3CN/MeOD (90 :10), 300 MHz) : δ = 124.35, 125.04, 128.00, 128.06, 138.62, 138.76, 152.96, 155.97, 158.70
MS-FAB (*m/z*) : calcd for $C_{30}H_{23}P_3F_{12}N_6O_3PRu$ = 937.99 found = 938.0.

4'-diethylphosphonate-2,2':6',2''-terpyridine-(4'-[(2,2'-bisthiophen)-5-yl]-2,2':6',2''-terpyridine) ruthenium (II) di-tetrafluoroborate (**11**)
A suspension of 4'-diethylphosphonate-2,2':6',2''-terpyridine ruthenium (III) trichloride (48 mg, 0.08 mmol.) and $AgBF_4$ (45 mg, 0.47 mmol.) was heated to 60 °C for 1 h 30 in a degassed mixture of ethanol (10 mL) and dry DMF (2.5 mL). After cooling to room temperature, the solution was filtered to remove AgCl and the filtrate was placed in a round bottom flask. The solution was degassed and 22 mg (0.07 mmol.) of 4'-[(2,2'-bisthiophen)-5-yl]-2,2':6',2''-terpyridine in 2 mL of dry DMF was added dropwise with 47 mg (0.4 mmol.) of hydroquinone. The resulting mixture was refluxed for 5 hours. The desired complex was precipitated by addition of petroleum ether to the mixture, filtered and washed with petroleum ether. The crude product was purified by column chromatography on silica gel, eluted with pure acetone and then with increasing gradients of water and saturated KNO_3 solution in acetone (starting from acetone/H_2O/KNO_3 90:10:0, until 79:20:1), to give a red product. The solid was dissolved in methanol and treated with $NaBF_4$ solution to complex **11** as a red solid (36 mg, 64%).
^{1}H RMN (CD_3CN, 300 MHz) : δ = 1.49 (t, 6H), 4.42 (m, 4H), 7.17 (m, 3H), 7.24 (dd, 2H, ^3J = 6.6 Hz, ^3J = 7.4 Hz), 7.32 (d, 2H, ^3J = 5.1 Hz), 7.47 (d, 1H, ^3J = 4.5 Hz), 7.50 (dd, 4H, ^3J = 6.6 Hz, ^3J = 7.4 Hz), 7.94 (m, 4H), 8.16 (d, 1H, ^3J = 4.5 Hz), 8.70 (d, 2H, ^3J = 7.5 Hz), 8.73 (d, 2H, ^3J = 7.5 Hz), 9.00 (d, 2H, J_p = 13.5 Hz), 9.01 (s, 2H).
^{13}C RMN (CD_3CN, 300 MHz) : δ = 16.51, 64.93, 120, 125,68, 125,95, 126.16, 127.23, 128.51, 128.91, 129.28, 130.48, 139.11, 138.22, 153.04, 153.40, 155.90, 158.39, 158.72
MS-FAB (*m/z*) : calcd for $C_{42}H_{35}B_2F_8N_6O_3PRuS_2$ = 1042.1 found = 1042.1.

4'-diethylphosphonate-2,2':6',2''-terpyridine-(4'-[(3',4'-dioctyl-2,2':5',2''-*tert*hiophen)-5-yl]-2,2':6',2''-terpyridine ruthenium (II) di-tetrafluoroborate (**12**)

A suspension of 4'-diethylphosphonate-2,2':6',2''-terpyridine ruthenium (III) trichloride **9** (89 mg, 0.16 mmol.) and $AgBF_4$ (91 mg, 0.47 mmol.) was heated to 60 °C for 1 h 30 in a degassed mixture of ethanol (20 mL) and dry DMF (5 mL). After cooling to room temperature, the solution was filtered to remove AgCl and the filtrate was placed in a round bottom flask. The solution was degassed and 91 mg (0.14 mmol.) of 4'-[(3',4'-dioctyl-2,2':5',2''-terthiophen)-5-yl]-2,2':6,2''-terpyridine in 8 mL of dry DMF was added dropwise with 93 mg (0.8 mmol.) of hydroquinone. The resulting mixture was refluxed for 5 hours. The DMF was removed on a rotary evaporator and the residue was dissolved in a minimum of dichloromethane. The desired complex was precipitated by addition of petroleum ether, filtered, washed with petroleum ether and dried under vacuum. Purification was performed by column chromatography on silica gel with acetone/H_2O/KNO_3 (90/10/0 to 78/20/2) as eluent to give a red product. The solid was dissolved in methanol and treated with $NaBF_4$ solution to precipitate complex **12** as a red solid (105 mg, 60%).

^{1}H RMN ($CDCl_3$, 300 MHz) : δ = 0.88 (m, 6H), 1.25-1.67 (m, 30H), 2.68 (t, 2H), 2.77 (t, 2H), 4.48 (q, 4H), 6.81 (m, 1H), 7.14 (m, 4H), 7.24 (m, 2H), 7.32 (d, 2H, ^3J = 5.1 Hz), 7.41 (dd, 1H, ^3J = 4.8 Hz, ^4J = 1.5 Hz), 7.49 (d, 2H, ^3J = 5.4 Hz), 7.82 (m, 4H), 8.16 (d, 1H, ^3J = 3.3 Hz), 8.47 (d, 2H, ^3J = 7.8 Hz), 8.60 (d, 2H, ^3J = 7.9 Hz), 8.86 (s, 2H), 8.90 (d, 2H, J_p= 13 Hz).

^{13}C RMN ($CDCl_3$, 300 MHz) : δ = 14.08, 16.52 22.67, 29.26, 29.31, 29.68, 29.91, 29.98, 30.84, 31.90, 64.24, 119.16, 119.50, 124.95, 125.23, 127.51, 127.69, 128.01, 128.15, 128.25, 128.58, 129.80, 130.84, 131.31, 133.67, 137.84, 138.08, 138.36, 140.55, 140.68, 140.88, 141.67, 145.89, 151.91, 152.64, 154.65, 155.39, 157.45, 157.51

MS-FAB (*m/z*) : calcd for $C_{62}H_{69}B_2F_8N_6O_3PRuS_3$ = 1348.3433 found = 1261 (M^+- BF_4), 1174 (M^+- $2BF_4$).

4'-phosphonic acid-2,2':6',2''-terpyridine-(4'-[(2,2'-bisthiophen)-5-yl]-2,2':6',2''-terpyridine) ruthenium (II) di-tetrafluoroborate (**14**)

To a solution of the previous complex (96 mg, 0.09 mmol.) in freshly distilled DMF (5 mL) was carefully added an excess of dry trimethylsilyl bromide (460 mg, 3 mmol.) under argon. The mixture was heated at 50 °C for 36 hours under argon. After cooling to room temperature, the desired complex was precipitated by addition of dichloromethane. The suspension was filtered, washed with dichloromethane and dried under vacuum. The solid was dissolved in methanol and treated with water to precipitate complex **15** as a red solid (40 mg, 45%).

^{1}H RMN (MeOD, 300 MHz) : δ = 7.20 to 7.65 (m, 12H), 8.1 (m, 4H), 8.33 (d, 1H, ^3J = 3.6 Hz), 8.75 (d, 2H, ^3J = 8.2 Hz), 8.94 (d, 2H, ^3J = 8.2 Hz), 9.17 (d, 2H, ^{3}J = 13.8 Hz), 9.2 (s, 2H).

^{13}C RMN (MeOD, 300 MHz) : δ = 119.87, 119.95, 125.47, 125.60, 125.90, 126.02, 128.70, 130.73, 132.02, 139.26, 152.69, 153.01, 155.52, 155.95, 156.12, 158.86

MS- MALDI-TOF (*m/z*) : calcd for $C_{38}H_{27}B_2F_8N_6O_3PRuS_2$ = 986.0425 found = 811.0 (M^+- 2 BF_4).

4'-phosphonic acid-2,2':6',2''-terpyridine-(4'-[(3',4'-dioctyl-2,2':5',2''-*tert*thiophen)-5-yl]-2,2':6',2''-terpyridine) ruthenium (II) di-tetrafluoroborate (**15**)

To a solution of complex **12** (70 mg, 0.05 mmol.) in freshly distilled DMF (10 mL), was carefully added an excess of dry trimethylsilyl bromide (400 mg, 2.5 mmol.) under argon. The mixture was heated at 50 °C for 36 hours under argon. After cooling to room temperature, the desired complex was precipitated by addition of dichloromethane. The suspension was filtered, washed with dichloromethane and dried under vacuum. The solid was dissolved in methanol and treated with water to precipitate complex **15** as a red solid (65 mg, 90%).

^{1}H RMN ($CDCl_3$/MeOD (90/10), 300 MHz) : δ = 0.83 (m, 6H), 1.28-1.62 (m, 24H), 2.71 (t, 2H), 2.81 (t, 2H), 7.0-7.2 (m, 6H), 7.32 (m, 6H), 7.90 (m, 4H), 8.21 (d, 1H, ^3J = 3.6 Hz), 8.62 (d, 4H, ^3J = 8.4 Hz), 8.85 (s, 2H), 9.14 (m, 2H).

^{13}C RMN ($CDCl_3$/MeOD (90/10), 300 MHz) : δ = 13.51, 22.24, 27.70, 28.80, 28.89, 29.21, 29.43, 29.45, 30.30, 30.35, 31.47, 118.52, 124.39, 124.43, 125.59, 125.92, 127.15, 127.48, 127.66, 128.22, 129.29, 131.10, 131.19, 135.13, 137.49, 138.07, 140.05, 140.72, 141.35, 141.90, 144.95, 151.27, 151.68, 154.69, 157.40, 157.78

MS- MALDI-TOF (*m/z*) : calcd for $C_{58}H_{61}B_2F_8N_6O_3PRuS_3$ = 1292.2 found =1117.2 (M^+-$2BF_4$).

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