# Supporting Information

## A cleavage-type pyrrolyl enone photoinitiator enables photobleaching-regulated uniform thick-film photopolymerization

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### 1. $^1$H NMR spectrum of DPP

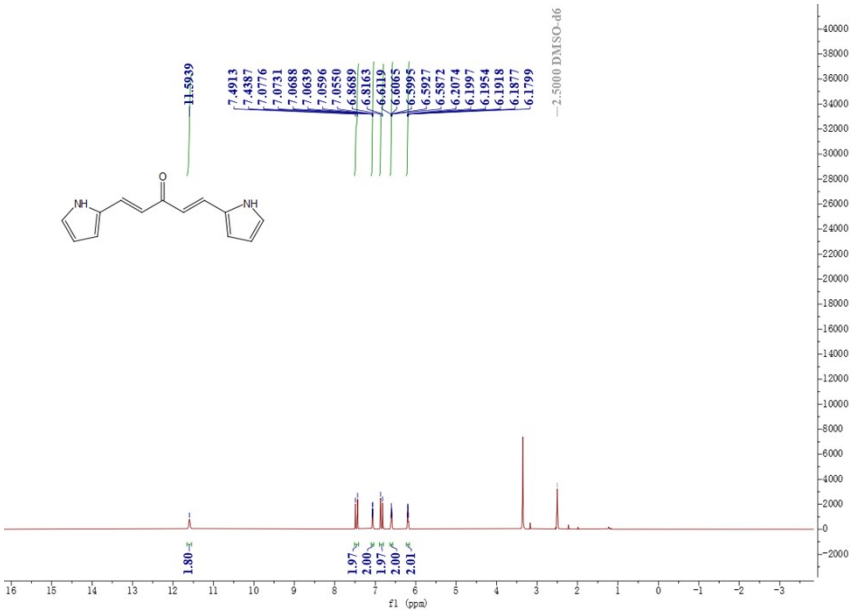


**Fig.S1** $^1$H NMR spectrum of DPP dissolved in DMSO-*d*6.

## 2. $^{1}$H NMR spectrum of C7-DPP-C7

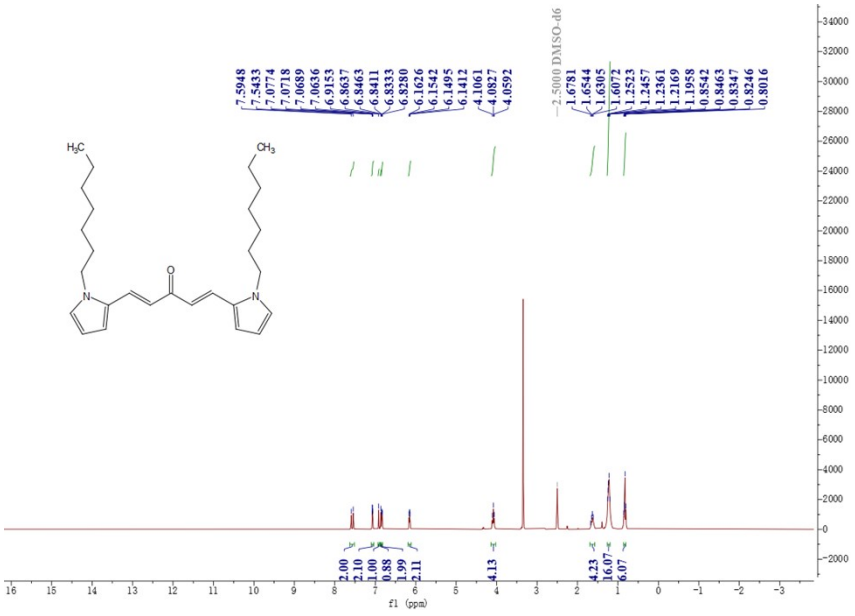


**Fig.S2** $^{1}$H NMR spectrum of C7-DPP-C7 dissolved in DMSO-*d*6.

## 3. $^{13}$C NMR spectrum of C7-DPP-C7

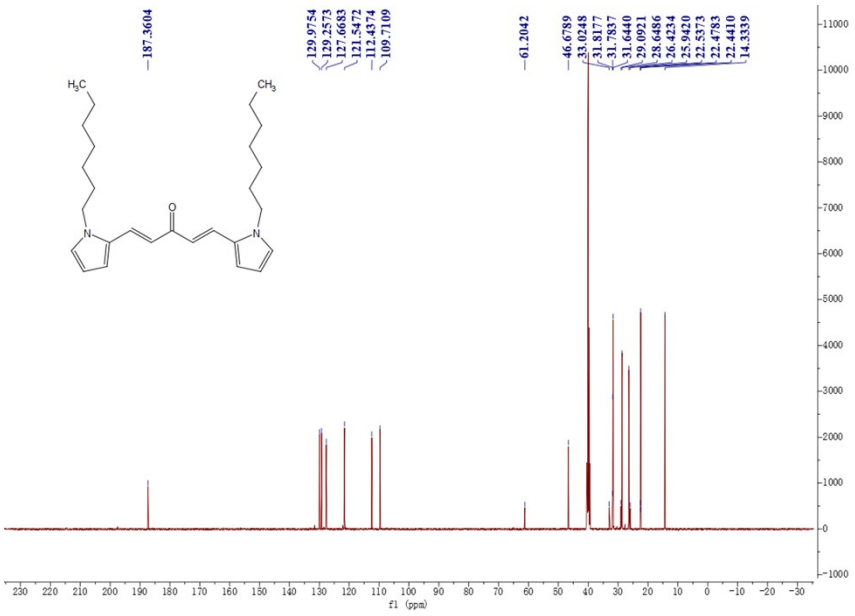


**Fig.S3** $^{13}$C NMR spectrum of **C7-DPP-C7** dissolved in DMSO-*d*6.

## 4. HRMS spectrum

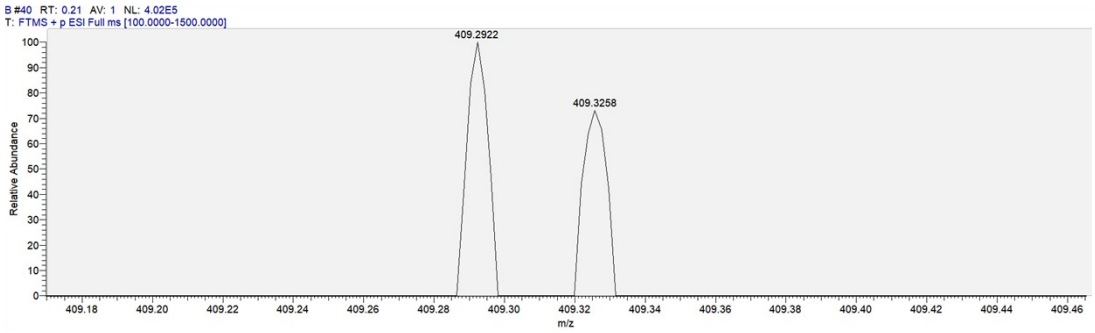


**Fig.S4** HRMS spectrum of **C7-DPP-C7**.

## 5. HPLC-MS spectrum

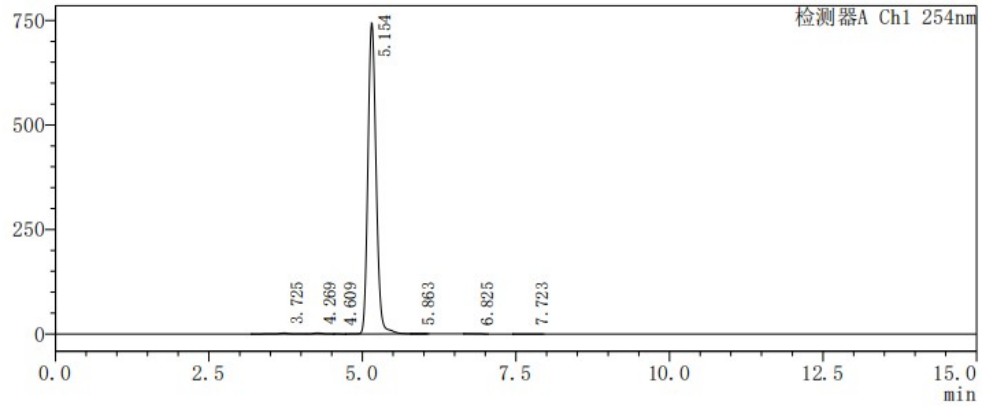


**Fig.S5** HPLC-MS spectrum of **C7-DPP-C7**.

## 6. Photolysis experiments

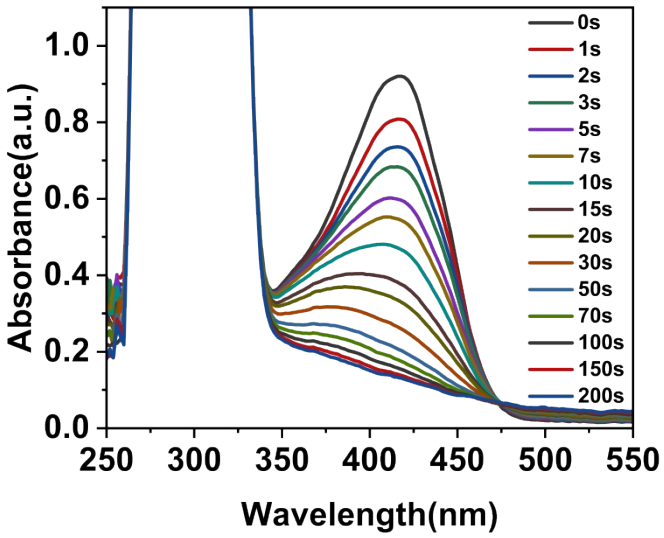


**Fig.S6** Photolysis spectra of C7-DPP-C7 with coinitiator EDB under LED@405nm irradiation.

## 7. Theoretically Calculated Cartesian Coordinates of C7-DPP-C7

**Table.1** Cartesian coordinates of the molecule **C7-DPP-C7** (optimized at the B3LYP/6-31G(d,p) level in acetonitrile using the PCM solvent model).

| Center Number | Atomic Number | Atomic Type | Coordinates (Angstroms) X | Y | Z |
|---|---|---|---|---|---|
| 1 | 6 | 0 | 6.218164 | -2.648049 | 0.257115 |
| 2 | 6 | 0 | 3.975133 | -2.654640 | 0.113973 |
| 3 | 6 | 0 | 5.784746 | -3.867462 | 0.761659 |
| 4 | 6 | 0 | 2.666393 | -2.159546 | -0.195898 |
| 5 | 6 | 0 | 1.484329 | -2.786852 | 0.031108 |
| 6 | 6 | 0 | 0.208584 | -2.159085 | -0.347338 |
| 7 | 6 | 0 | -1.003631 | -2.938265 | -0.050291 |
| 8 | 8 | 0 | 0.157806 | -1.036692 | -0.885751 |
| 9 | 6 | 0 | -2.235838 | -2.457806 | -0.355543 |
| 10 | 6 | 0 | -3.491655 | -3.110404 | -0.129378 |
| 11 | 6 | 0 | -5.722934 | -3.380717 | -0.133129 |
| 12 | 6 | 0 | -5.176643 | -4.538742 | 0.405309 |
| 13 | 1 | 0 | 7.216770 | -2.249900 | 0.148106 |
| 14 | 1 | 0 | 3.716713 | -4.674808 | 0.965724 |
| 15 | 1 | 0 | 6.420167 | -4.656381 | 1.138541 |
| 16 | 1 | 0 | 2.593092 | -1.180183 | -0.664058 |
| 17 | 1 | 0 | 1.448635 | -3.767176 | 0.498513 |
| 18 | 1 | 0 | -0.879913 | -3.909333 | 0.421480 |
| 19 | 1 | 0 | -2.251679 | -1.474451 | -0.821007 |
| 20 | 1 | 0 | -6.753677 | -3.108743 | -0.309784 |
| 21 | 1 | 0 | -5.733394 | -5.400927 | 0.744216 |
| 22 | 1 | 0 | -3.042745 | -5.085358 | 0.748632 |
| 23 | 7 | 0 | -4.714159 | -2.523156 | -0.460585 |
| 24 | 7 | 0 | 5.134989 | -1.919522 | -0.138010 |
| 25 | 6 | 0 | -3.780236 | -4.371863 | 0.407985 |
| 26 | 6 | 0 | 4.381210 | -3.873899 | 0.672406 |
| 27 | 6 | 0 | -4.927550 | -1.173373 | -0.988188 |
| 28 | 1 | 0 | -4.168353 | -0.970585 | -1.748793 |
| 29 | 1 | 0 | -5.892806 | -1.181784 | -1.501963 |
| 30 | 6 | 0 | -4.914311 | -0.086643 | 0.094374 |
| 31 | 1 | 0 | -3.947730 | -0.102171 | 0.612799 |
| 32 | 1 | 0 | -5.677633 | -0.327514 | 0.844765 |
| 33 | 6 | 0 | -5.171255 | 1.309674 | -0.484628 |
| 34 | 1 | 0 | -6.136425 | 1.314010 | -1.010179 |
| 35 | 1 | 0 | -4.408705 | 1.535748 | -1.242953 |
| 36 | 6 | 0 | -5.169246 | 2.412865 | 0.580913 |
| 37 | 1 | 0 | -4.204054 | 2.406605 | 1.106609 |
| 38 | 1 | 0 | -5.931072 | 2.184551 | 1.339579 |
| 39 | 6 | 0 | -5.426292 | 3.812453 | 0.009664 |
| 40 | 1 | 0 | -6.391006 | 3.818146 | -0.517752 |
| 41 | 1 | 0 | -4.664064 | 4.040892 | -0.749022 |

| | | | | | |
|---|---|---|---|---|---|
| 42 | 6 | 0 | -5.426417 | 4.917377 | 1.073286 |
| 43 | 1 | 0 | -4.462484 | 4.911441 | 1.600113 |
| 44 | 1 | 0 | -6.188016 | 4.688767 | 1.831244 |
| 45 | 6 | 0 | -5.683812 | 6.312457 | 0.495659 |
| 46 | 1 | 0 | -6.656343 | 6.358976 | -0.007796 |
| 47 | 1 | 0 | -5.677775 | 7.077826 | 1.278743 |
| 48 | 1 | 0 | -4.918168 | 6.583312 | -0.240441 |
| 49 | 6 | 0 | 5.214351 | -0.550803 | -0.653892 |
| 50 | 1 | 0 | 6.205002 | -0.437563 | -1.102878 |
| 51 | 1 | 0 | 4.487464 | -0.438894 | -1.463361 |
| 52 | 6 | 0 | 4.995848 | 0.519964 | 0.422590 |
| 53 | 1 | 0 | 4.006800 | 0.381522 | 0.876487 |
| 54 | 1 | 0 | 5.732245 | 0.371630 | 1.222348 |
| 55 | 6 | 0 | 5.114058 | 1.940905 | -0.141986 |
| 56 | 1 | 0 | 6.106853 | 2.071506 | -0.595538 |
| 57 | 1 | 0 | 4.386562 | 2.065863 | -0.954147 |
| 58 | 6 | 0 | 4.900815 | 3.023449 | 0.925499 |
| 59 | 1 | 0 | 3.895162 | 2.913921 | 1.355664 |
| 60 | 1 | 0 | 5.604592 | 2.846170 | 1.749759 |
| 61 | 6 | 0 | 5.082832 | 4.464514 | 0.422417 |
| 62 | 1 | 0 | 5.053234 | 5.142840 | 1.286002 |
| 63 | 1 | 0 | 6.086998 | 4.572608 | -0.012577 |
| 64 | 6 | 0 | 4.036436 | 4.929288 | -0.600019 |
| 65 | 1 | 0 | 4.078758 | 4.294975 | -1.494173 |
| 66 | 1 | 0 | 3.032825 | 4.790257 | -0.174791 |
| 67 | 6 | 0 | 4.217711 | 6.393136 | -1.014019 |
| 68 | 1 | 0 | 5.201052 | 6.556214 | -1.470094 |
| 69 | 1 | 0 | 3.459097 | 6.701136 | -1.741117 |
| 70 | 1 | 0 | 4.141023 | 7.061540 | -0.148634 |

**Table.2** Cartesian coordinates of the photolysis product ethylene derivatives (optimized at the B3LYP/6-31G(d,p) level in acetonitrile using the PCM solvent model).

| Center Number | Atomic Number | Atomic Type | Coordinates (Angstroms) X | Y | Z |
|---|---|---|---|---|---|
| 1 | 6 | 0 | 2.393873 | -1.730630 | 0.000004 |
| 2 | 6 | 0 | 3.736019 | -2.064032 | 0.000001 |
| 3 | 6 | 0 | 4.458622 | -0.851740 | -0.000003 |
| 4 | 6 | 0 | 3.544241 | 0.202009 | -0.000002 |
| 5 | 7 | 0 | 2.270416 | -0.362588 | 0.000002 |
| 6 | 6 | 0 | 3.900690 | 1.608488 | -0.000005 |
| 7 | 6 | 0 | 3.162763 | 2.734037 | 0.000001 |
| 8 | 6 | 0 | 1.006383 | 0.384240 | 0.000004 |
| 9 | 6 | 0 | -0.245568 | -0.491903 | 0.000001 |
| 10 | 6 | 0 | -1.522694 | 0.359341 | 0.000002 |
| 11 | 6 | 0 | -2.804095 | -0.483119 | -0.000003 |
| 12 | 6 | 0 | -4.085828 | 0.358523 | 0.000004 |
| 13 | 6 | 0 | -5.369388 | -0.480857 | -0.000008 |
| 14 | 6 | 0 | -6.645623 | 0.366109 | 0.000002 |
| 15 | 1 | 0 | 1.525440 | -2.368896 | 0.000007 |
| 16 | 1 | 0 | 4.135166 | -3.068755 | 0.000001 |
| 17 | 1 | 0 | 5.533739 | -0.729111 | -0.000006 |
| 18 | 1 | 0 | 4.982899 | 1.736097 | -0.000012 |
| 19 | 1 | 0 | 3.660768 | 3.698177 | -0.000002 |
| 20 | 1 | 0 | 2.081132 | 2.756622 | 0.000009 |
| 21 | 1 | 0 | 0.992078 | 1.034481 | 0.881988 |
| 22 | 1 | 0 | 0.992079 | 1.034484 | -0.881978 |
| 23 | 1 | 0 | -0.243819 | -1.143929 | -0.881975 |
| 24 | 1 | 0 | -0.243820 | -1.143933 | 0.881974 |
| 25 | 1 | 0 | -1.521274 | 1.018659 | 0.878831 |
| 26 | 1 | 0 | -1.521272 | 1.018667 | -0.878820 |
| 27 | 1 | 0 | -2.803336 | -1.143755 | -0.878412 |
| 28 | 1 | 0 | -2.803336 | -1.143768 | 0.878394 |
| 29 | 1 | 0 | -4.086256 | 1.019927 | 0.878267 |
| 30 | 1 | 0 | -4.086254 | 1.019947 | -0.878245 |
| 31 | 1 | 0 | -5.368615 | -1.141646 | -0.877580 |
| 32 | 1 | 0 | -5.368617 | -1.141669 | 0.877547 |
| 33 | 1 | 0 | -7.542978 | -0.261392 | -0.000010 |
| 34 | 1 | 0 | -6.691499 | 1.012535 | 0.883993 |
| 35 | 1 | 0 | -6.691495 | 1.012563 | -0.883969 |

**Table.3** Cartesian coordinates of the photolysis product aldehyde derivatives (optimized at the B3LYP/6-31G(d,p) level in acetonitrile using the PCM solvent model).

| Center Number | Atomic Number | Atomic Type | Coordinates (Angstroms) X | Y | Z |
|---|---|---|---|---|---|
| 1 | 6 | 0 | 1.564141 | -2.559724 | 0.000003 |
| 2 | 6 | 0 | 2.835584 | -3.121273 | 0.000000 |
| 3 | 6 | 0 | 3.747584 | -2.057127 | -0.000002 |
| 4 | 6 | 0 | 3.020921 | -0.856346 | -0.000001 |
| 5 | 7 | 0 | 1.666120 | -1.199636 | 0.000002 |
| 6 | 6 | 0 | 3.605577 | 0.446189 | -0.000003 |
| 7 | 6 | 0 | 3.077007 | 1.703568 | 0.000000 |
| 8 | 6 | 0 | 3.966729 | 2.847199 | -0.000002 |
| 9 | 8 | 0 | 3.608120 | 4.023465 | 0.000002 |
| 10 | 6 | 0 | 0.541897 | -0.251353 | 0.000002 |
| 11 | 6 | 0 | -0.837029 | -0.908761 | 0.000001 |
| 12 | 6 | 0 | -1.953760 | 0.144506 | 0.000001 |
| 13 | 6 | 0 | -3.357562 | -0.472711 | -0.000001 |
| 14 | 6 | 0 | -4.480097 | 0.571877 | 0.000001 |
| 15 | 6 | 0 | -5.886106 | -0.040610 | -0.000003 |
| 16 | 6 | 0 | -7.002171 | 1.008458 | 0.000000 |
| 17 | 1 | 0 | 0.602742 | -3.047361 | 0.000005 |
| 18 | 1 | 0 | 3.054557 | -4.179346 | 0.000001 |
| 19 | 1 | 0 | 4.827765 | -2.115941 | -0.000004 |
| 20 | 1 | 0 | 4.695145 | 0.395451 | -0.000007 |
| 21 | 1 | 0 | 2.015011 | 1.914849 | 0.000005 |
| 22 | 1 | 0 | 5.050186 | 2.591928 | -0.000005 |
| 23 | 1 | 0 | 0.638811 | 0.389096 | 0.883123 |
| 24 | 1 | 0 | 0.638812 | 0.389098 | -0.883117 |
| 25 | 1 | 0 | -0.943704 | -1.551020 | -0.882543 |
| 26 | 1 | 0 | -0.943705 | -1.551020 | 0.882544 |
| 27 | 1 | 0 | -1.842016 | 0.793933 | 0.878908 |
| 28 | 1 | 0 | -1.842015 | 0.793935 | -0.878905 |
| 29 | 1 | 0 | -3.467130 | -1.124016 | -0.878476 |
| 30 | 1 | 0 | -3.467131 | -1.124020 | 0.878470 |
| 31 | 1 | 0 | -4.369621 | 1.223950 | 0.878248 |
| 32 | 1 | 0 | -4.369619 | 1.223956 | -0.878242 |
| 33 | 1 | 0 | -5.996138 | -0.692150 | -0.877582 |
| 34 | 1 | 0 | -5.996140 | -0.692157 | 0.877571 |
| 35 | 1 | 0 | -7.992053 | 0.540345 | -0.000003 |
| 36 | 1 | 0 | -6.938927 | 1.653430 | 0.883973 |
| 37 | 1 | 0 | -6.938925 | 1.653438 | -0.883967 |