

## **Supporting Information**

### **Morphology of OLED film stacks containing solution-processed phosphorescent dendrimers**

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**Table S1** – Room temperature models for films containing a *d*-CBP:IrG1 emissive layer

| <i>Film number</i> | <i>Layer material</i> | <i>Thickness [<math>\text{\AA}</math>]</i> | <i>SLD [<math>10^{-6} \text{\AA}^{-2}</math>]</i> | <i>Roughness [<math>\text{\AA}</math>]</i> |
|--------------------|-----------------------|--------------------------------------------|---------------------------------------------------|--------------------------------------------|
| <b>1</b>           | <i>d</i> -CBP:IrG1    | $461 \pm 1$                                | $4.12 \pm 0.01$                                   | $9 \pm 1$                                  |
| <b>2</b>           | PEDOT:PSS*            | $282 \pm 1$                                | $1.92 \pm 0.01$                                   | $16 \pm 1$                                 |
|                    | <i>d</i> -CBP:IrG1    | $464 \pm 1$                                | $4.13 \pm 0.01$                                   | $9 \pm 1$                                  |
| <b>3</b>           | <i>d</i> -CBP:IrG1    | $430 \pm 1$                                | $4.08 \pm 0.01$                                   | $12 \pm 1$                                 |
|                    | TPBi                  | $288 \pm 1$                                | $2.72 \pm 0.01$                                   | $12 \pm 1$                                 |
| <b>4</b>           | PEDOT:PSS*            | $324 \pm 1$                                | $1.90 \pm 0.02$                                   | $17 \pm 1$                                 |
|                    | <i>d</i> -CBP:IrG1    | $452 \pm 1$                                | $4.11 \pm 0.01$                                   | $9 \pm 1$                                  |
|                    | TPBi                  | $291 \pm 1$                                | $2.75 \pm 0.01$                                   | $8 \pm 1$                                  |

\* The thickness and SLD of the PEDOT:PSS layer was fixed during the temperature measurements as it did not appear to change.

**Table S2** – Room temperature models for films containing a *d*-TCTA:IrG1 emissive layer

| <i>Film number</i> | <i>Layer material</i> | <i>Thickness [<math>\text{\AA}</math>]</i> | <i>SLD [<math>10^{-6} \text{\AA}^{-2}</math>]</i> | <i>Roughness [<math>\text{\AA}</math>]</i> |
|--------------------|-----------------------|--------------------------------------------|---------------------------------------------------|--------------------------------------------|
| <b>5</b>           | <i>d</i> -TCTA:IrG1   | $442 \pm 1$                                | $4.06 \pm 0.01$                                   | $10 \pm 1$                                 |
| <b>6</b>           | PEDOT:PSS*            | $324 \pm 1$                                | $1.92 \pm 0.01$                                   | $17 \pm 1$                                 |
|                    | <i>d</i> -TCTA:IrG1   | $460 \pm 1$                                | $4.09 \pm 0.01$                                   | $9 \pm 1$                                  |
| <b>7</b>           | <i>d</i> -TCTA:IrG1   | $429 \pm 1$                                | $4.09 \pm 0.01$                                   | $9 \pm 1$                                  |
|                    | TPBi                  | $287 \pm 1$                                | $2.73 \pm 0.01$                                   | $7 \pm 1$                                  |
| <b>8</b>           | PEDOT:PSS*            | $322 \pm 1$                                | $1.81 \pm 0.02$                                   | $14 \pm 1$                                 |
|                    | <i>d</i> -TCTA:IrG1   | $446 \pm 1$                                | $4.07 \pm 0.01$                                   | $9 \pm 1$                                  |
|                    | TPBi                  | $288 \pm 1$                                | $2.75 \pm 0.01$                                   | $6 \pm 1$                                  |

\* The thickness and SLD of the PEDOT:PSS layer was fixed during the temperature measurements as it did not appear to change.

**Table S3** – Model parameters for Film **2** on thermal annealing

| <b>Temperature range (°C)</b> | <b>Layer (numbered from substrate upwards)</b> | <b>Thickness (Å)</b> | <b>SLD (<math>10^{-6} \text{ Å}^{-2}</math>)</b> | <b>Roughness (Å)</b> |
|-------------------------------|------------------------------------------------|----------------------|--------------------------------------------------|----------------------|
| 139 – 140                     | 1 (PEDOT:PSS)                                  | 282                  | 1.92                                             | 5                    |
|                               | 2 ( <i>d</i> -CBP:IrG1)                        | 470                  | 4.02                                             | 4                    |
| 149 – 150                     | 1                                              | 282                  | 1.92                                             | 19                   |
|                               | 2                                              | 469                  | 4.03                                             | 4                    |

**Table S4** – Model parameters for Film **3** on thermal annealing

| <b>Temperature range (°C)</b> | <b>Layer (numbered from substrate upwards)</b> | <b>Thickness (Å)</b> | <b>SLD (<math>10^{-6} \text{ Å}^{-2}</math>)</b> | <b>Roughness (Å)</b> |
|-------------------------------|------------------------------------------------|----------------------|--------------------------------------------------|----------------------|
| 90 – 91                       | 1 ( <i>d</i> -CBP:IrG1)                        | 433                  | 4.02                                             | 4                    |
|                               | 2 (TPBi)                                       | 293                  | 2.68                                             | 5                    |
| 94 – 95                       | 1                                              | 443                  | 3.98                                             | 27                   |
|                               | 2                                              | 288                  | 2.71                                             | 4                    |
| 100 – 101                     | 1                                              | 494                  | 3.84                                             | 5                    |
|                               | 2                                              | 237                  | 2.71                                             | 5                    |
| 105 – 106                     | 1                                              | 544                  | 3.78                                             | 30                   |
|                               | 2                                              | 197                  | 2.70                                             | 10                   |
| 110 – 111                     | 1                                              | 563                  | 3.67                                             | 33                   |
|                               | 2                                              | 171                  | 2.83                                             | 5                    |
| 113 – 114                     | 1                                              | 589                  | 3.58                                             | 36                   |
|                               | 2                                              | 144                  | 3.02                                             | 4                    |
| 117 – 118                     | 1<br>(Completely mixed)                        | 729                  | 3.46                                             | 4                    |
| 118 – 119                     | 1                                              | 543                  | 3.39                                             | 88                   |
|                               | 2                                              | 182                  | 3.65                                             | 4                    |
| 159 – 160                     | 1                                              | 612                  | 3.40                                             | 63                   |
|                               | 2                                              | 131                  | 3.51                                             | 4                    |

**Table S5** – Model parameters for Film 4 on thermal annealing

| <b>Temperature range (°C)</b> | <b>Layer (numbered from substrate upwards)</b> | <b>Thickness (Å)</b> | <b>SLD (<math>10^{-6} \text{ Å}^{-2}</math>)</b> | <b>Roughness (Å)</b> |
|-------------------------------|------------------------------------------------|----------------------|--------------------------------------------------|----------------------|
| 90 – 91                       | 1 (PEDOT:PSS)                                  | 324                  | 1.90                                             | 24                   |
|                               | 2 ( <i>d</i> -CBP:IrG1)                        | 459                  | 3.96                                             | 5                    |
|                               | 3 (TPBi)                                       | 293                  | 2.76                                             | 4                    |
| 94 – 95                       | 1                                              | 324                  | 1.90                                             | 7                    |
|                               | 2                                              | 469                  | 3.92                                             | 30                   |
|                               | 3                                              | 282                  | 2.74                                             | 4                    |
| 100 – 101                     | 1                                              | 324                  | 1.90                                             | 5                    |
|                               | 2                                              | 522                  | 3.77                                             | 4                    |
|                               | 3                                              | 231                  | 2.70                                             | 4                    |
| 105 – 106                     | 1                                              | 324                  | 1.90                                             | 15                   |
|                               | 2                                              | 559                  | 3.73                                             | 5                    |
|                               | 3                                              | 198                  | 2.72                                             | 10                   |
| 110 – 111                     | 1                                              | 324                  | 1.90                                             | 32                   |
|                               | 2                                              | 590                  | 3.60                                             | 41                   |
|                               | 3                                              | 161                  | 2.79                                             | 5                    |
| 113 – 114                     | 1                                              | 324                  | 1.90                                             | 32                   |
|                               | 2                                              | 598                  | 3.58                                             | 29                   |
|                               | 3                                              | 155                  | 3.04                                             | 4                    |
| 117 – 118                     | 1                                              | 324                  | 1.90                                             | 5                    |
|                               | 2                                              | 753                  | 3.46                                             | 4                    |
| 118 – 119                     | 1                                              | 324                  | 1.90                                             | 15                   |
|                               | 2                                              | 527                  | 3.38                                             | 98                   |
|                               | 3                                              | 225                  | 3.62                                             | 4                    |
| 159 – 160                     | 1                                              | 324                  | 1.90                                             | 8                    |

|  |   |     |      |     |
|--|---|-----|------|-----|
|  | 2 | 410 | 3.27 | 245 |
|  | 3 | 343 | 3.56 | 4   |

**Table S6** – Model parameters for Film 6 on thermal annealing

| <b>Temperature range (°C)</b> | <b>Layer (numbered from substrate upwards)</b> | <b>Thickness (Å)</b> | <b>SLD (<math>10^{-6} \text{ Å}^{-2}</math>)</b> | <b>Roughness (Å)</b> |
|-------------------------------|------------------------------------------------|----------------------|--------------------------------------------------|----------------------|
| 139 – 140                     | 1 (PEDOT:PSS)                                  | 324                  | 1.92                                             | 17                   |
|                               | 2 ( <i>d</i> -TCTA:IrG1)                       | 464                  | 3.98                                             | 4                    |
| 149 – 150                     | 1                                              | 324                  | 1.92                                             | 19                   |
|                               | 2                                              | 459                  | 3.93                                             | 4                    |
| 159 – 160                     | 1                                              | 324                  | 1.92                                             | 25                   |
|                               | 2                                              | 461                  | 3.92                                             | 4                    |

**Table S7** – Model parameters for Film 7 on thermal annealing

| <b>Temperature range (°C)</b> | <b>Layer (numbered from substrate upwards)</b> | <b>Thickness (Å)</b> | <b>SLD (<math>10^{-6} \text{ Å}^{-2}</math>)</b> | <b>Roughness (Å)</b> |
|-------------------------------|------------------------------------------------|----------------------|--------------------------------------------------|----------------------|
| 110 – 111                     | 1 (PEDOT:PSS)                                  | 437                  | 3.98                                             | 5                    |
|                               | 2 ( <i>d</i> -TCTA:IrG1)                       | 293                  | 2.70                                             | 6                    |
| 115 – 116                     | 1                                              | 437                  | 3.99                                             | 13                   |
|                               | 2                                              | 292                  | 2.71                                             | 5                    |
| 116 – 117                     | 1                                              | 385                  | 3.97                                             | 22                   |
|                               | 2 (mixed interlayer)                           | 70                   | 3.56                                             | 18                   |
|                               | 3                                              | 272                  | 2.70                                             | 5                    |
| 120 – 121                     | 1                                              | 273                  | 3.97                                             | 9                    |
|                               | 2                                              | 235                  | 3.58                                             | 20                   |
|                               | 3                                              | 231                  | 2.70                                             | 6                    |
| 130 – 131                     | 1                                              | 226                  | 3.91                                             | 90                   |
|                               | 2                                              | 346                  | 3.41                                             | 14                   |
|                               | 3                                              | 159                  | 2.70                                             | 5                    |
| 131 – 132                     | 1                                              | 303                  | 3.58                                             | 9                    |
|                               | 2                                              | 301                  | 3.49                                             | 15                   |
|                               | 3                                              | 133                  | 3.08                                             | 5                    |
| 134 – 135                     | 1<br>(completely mixed)                        | 727                  | 3.42                                             | 3                    |
| 135 – 136                     | 1                                              | 150                  | 3.35                                             | 60                   |
|                               | 2                                              | 418                  | 3.39                                             | 95                   |
|                               | 3                                              | 161                  | 3.65                                             | 5                    |
| 159 – 160                     | 1                                              | 677                  | 3.43                                             | 45                   |
|                               | 2                                              | 59                   | 3.65                                             | 6                    |

**Table S8** – Model parameters for Film **8** on thermal annealing

| <b>Temperature range (°C)</b> | <b>Layer (numbered from substrate upwards)</b> | <b>Thickness (Å)</b> | <b>SLD (<math>10^{-6} \text{ Å}^{-2}</math>)</b> | <b>Roughness (Å)</b> |
|-------------------------------|------------------------------------------------|----------------------|--------------------------------------------------|----------------------|
| 110 – 111                     | 1 (PEDOT:PSS)                                  | 322                  | 1.81                                             | 16                   |
|                               | 2 ( <i>d</i> -TCTA:IrG1)                       | 446                  | 3.92                                             | 5                    |
|                               | 3 (TPBi)                                       | 287                  | 2.74                                             | 4                    |
| 116 – 117                     | 1                                              | 322                  | 1.81                                             | 17                   |
|                               | 2                                              | 445                  | 3.91                                             | 20                   |
|                               | 3                                              | 289                  | 2.73                                             | 5                    |
| 117 – 118                     | 1                                              | 322                  | 1.81                                             | 18                   |
|                               | 2                                              | 430                  | 3.90                                             | 17                   |
|                               | 3 (mixed interlayer)                           | 65                   | 3.14                                             | 52                   |
|                               | 4                                              | 240                  | 2.70                                             | 6                    |
| 120 – 121                     | 1                                              | 322                  | 1.81                                             | 17                   |
|                               | 2                                              | 333                  | 3.90                                             | 83                   |
|                               | 3                                              | 187                  | 3.45                                             | 17                   |
|                               | 4                                              | 218                  | 2.71                                             | 6                    |
| 125 – 126                     | 1                                              | 322                  | 1.81                                             | 20                   |
|                               | 2                                              | 102                  | 4.06                                             | 113                  |
|                               | 3                                              | 457                  | 3.54                                             | 6                    |
|                               | 4                                              | 181                  | 2.70                                             | 10                   |
| 129 – 130                     | 1                                              | 322                  | 1.81                                             | 23                   |
|                               | 2                                              | 174                  | 3.50                                             | 32                   |
|                               | 3                                              | 417                  | 3.53                                             | 6                    |
|                               | 4                                              | 147                  | 3.03                                             | 7                    |
| 134 – 135                     | 1                                              | 322                  | 1.81                                             | 7                    |
|                               | 2                                              | 122                  | 3.11                                             | 63                   |

|           |   |     |      |     |
|-----------|---|-----|------|-----|
|           | 3 | 471 | 3.40 | 89  |
|           | 4 | 145 | 3.42 | 6   |
| 135 – 136 | 1 | 322 | 1.81 | 5   |
|           | 2 | 144 | 3.19 | 87  |
|           | 3 | 478 | 3.36 | 118 |
|           | 4 | 113 | 3.62 | 7   |
| 159 – 160 | 1 | 322 | 1.81 | 5   |
|           | 2 | 229 | 3.21 | 57  |
|           | 3 | 497 | 3.39 | 79  |
|           | 4 | 10  | 3.65 | 10  |

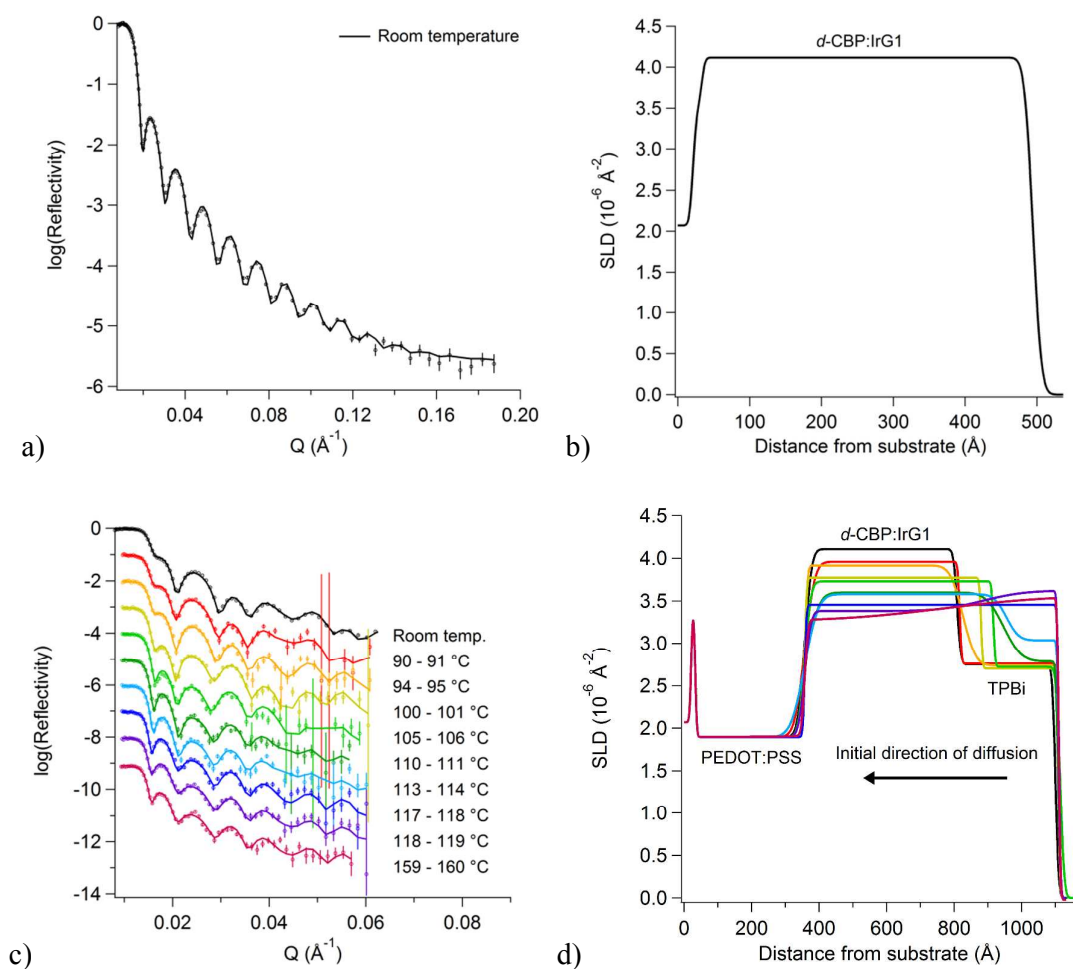


Figure S1 – a) Neutron reflectivity profiles and b) SLD *versus* distance from substrate plots for: a), b) Film 1 (*d*-CBP:IrG1); and c), d) Film 4 (PEDOT:PSS/*d*-CBP:IrG1/TPBi). NR profiles have been offset for clarity, with individual points indicating experimental data, and solid lines indicating model fits. In Figure S1 d) the arrow indicates the initial direction of material diffusion leading to the fully intermixed layer. Colours of thickness plots match the legend for the NR profiles.

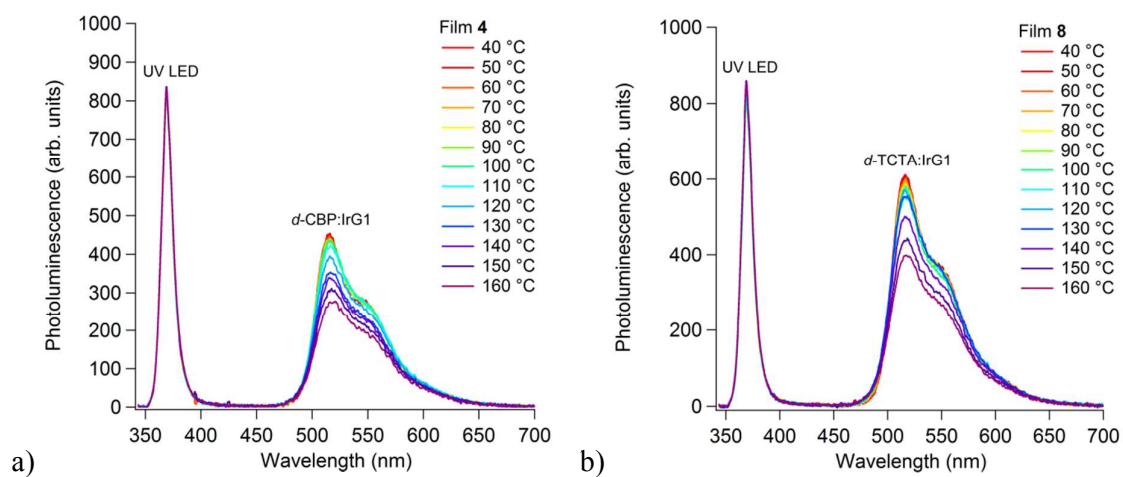


Figure S2 – *In situ* PL spectra for a) Film 4 (PEDOT:PSS/*d*-CBP:IrG1/TPBi); b) Film 8 (PEDOT:PSS/*d*-TCTA:IrG1/TPBi).

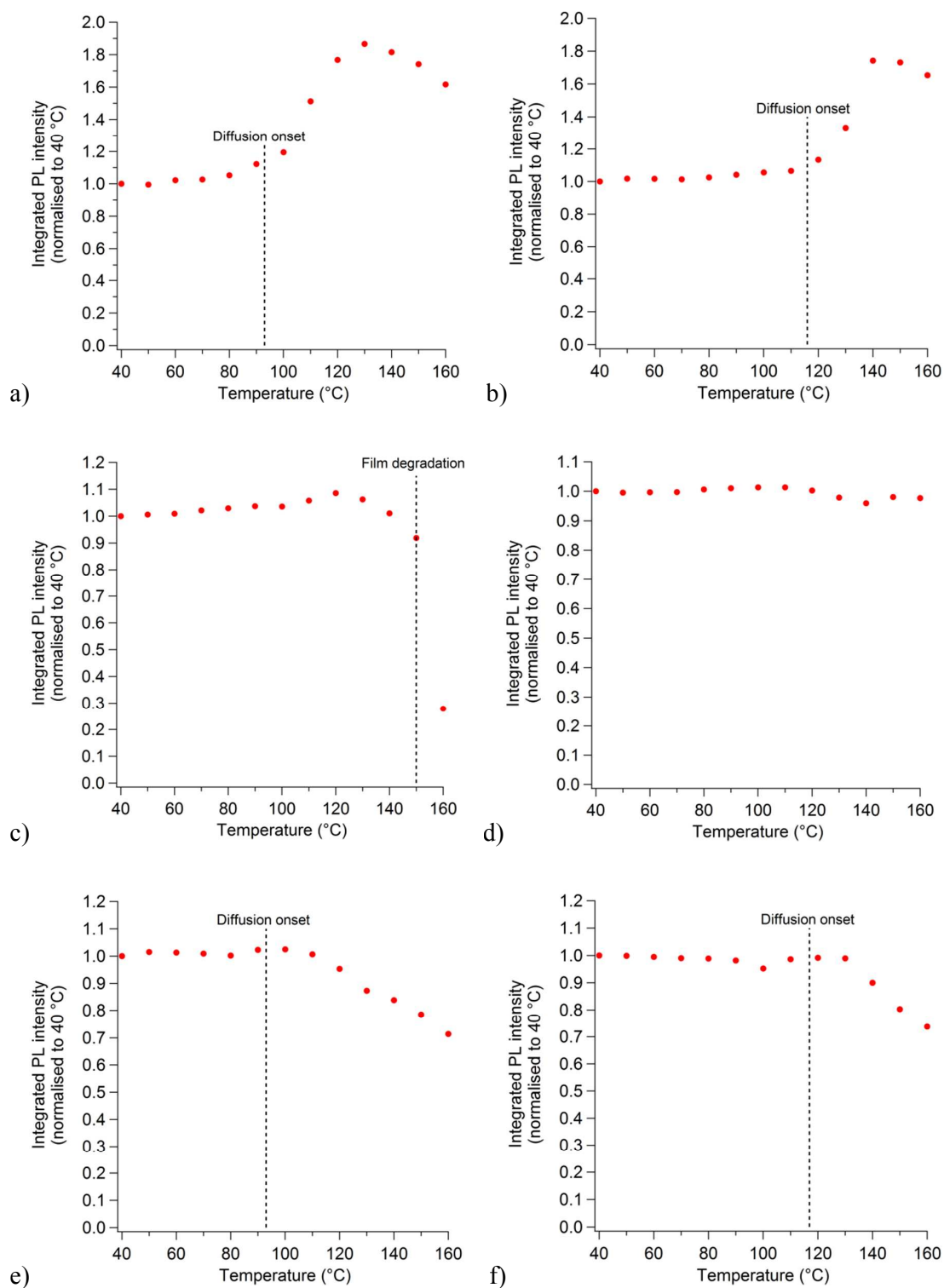


Figure S3 – Integrated PL intensity (normalised to the emission area at 40 °C) for a) Film 3 (*d*-CBP:IrG1/TPBi); b) Film 7 (*d*-TCTA:IrG1/TPBi) [between 475 and 650 nm]; c) Film 2 (PEDOT:PSS/ *d*-CBP:IrG1); d) Film 6 (PEDOT:PSS/*d*-TCTA:IrG1) [between 475 and 650 nm]; e) Film 4 (PEDOT:PSS/*d*-CBP:IrG1/TPBi); f) Film 8 (PEDOT:PSS/*d*-TCTA:IrG1/TPBi) [between 475 and 650 nm].

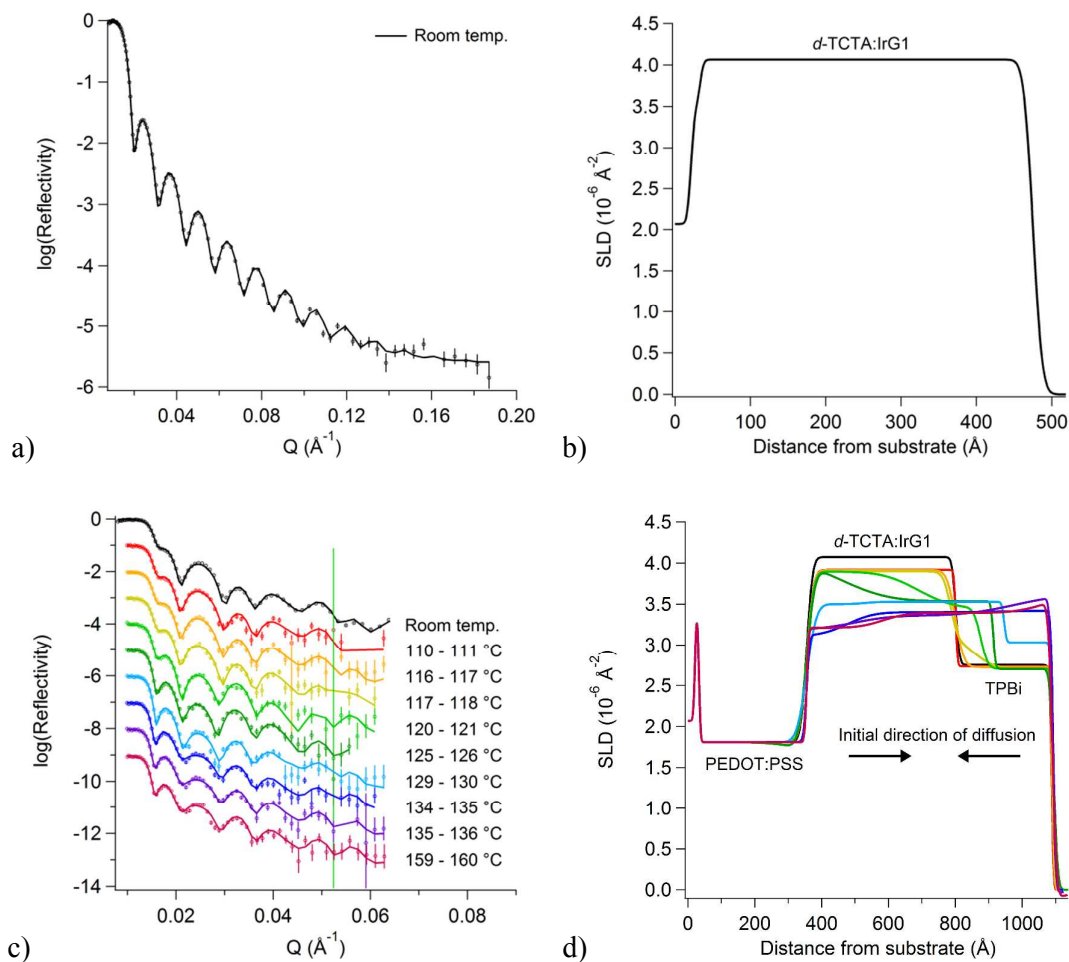


Figure S4 – Neutron reflectivity profiles and SLD *versus* distance from substrate plots for: a, b) Film 5 (*d*-TCTA:IrG1); and c, d) Film 8 (PEDOT:PSS/*d*-TCTA:IrG1/TPBi). NR profiles have been offset for clarity, with individual points indicating experimental data, and solid lines indicating model fits. In Figure S4 d) the arrows indicate the initial direction of material diffusion leading to the intermixed layer. Colours of thickness plots match the legend for the NR profiles.

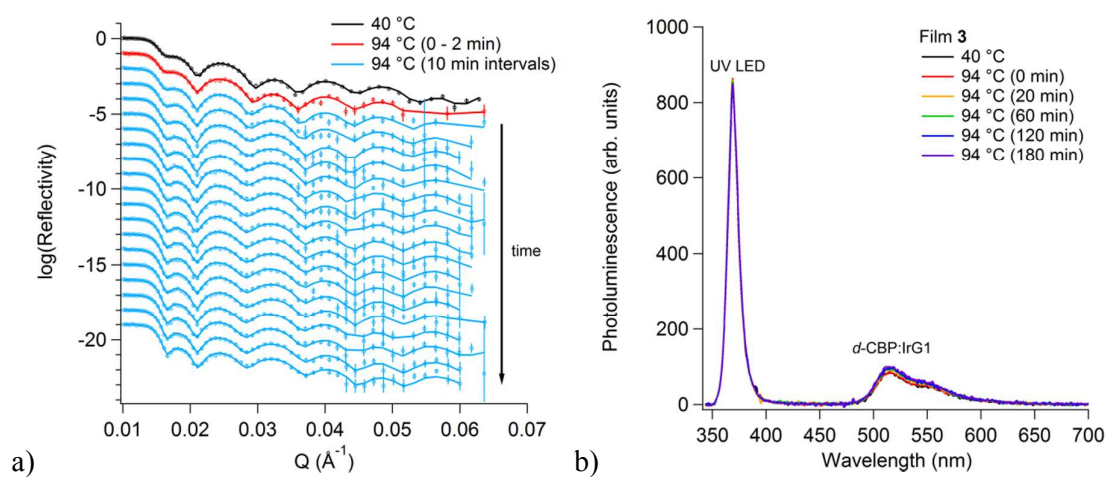


Figure S5 – a) Time resolved neutron reflectivity profiles, b) PL spectra for Film 3 (*d*-CBP:IrG1/TPBi) held at 94 °C.

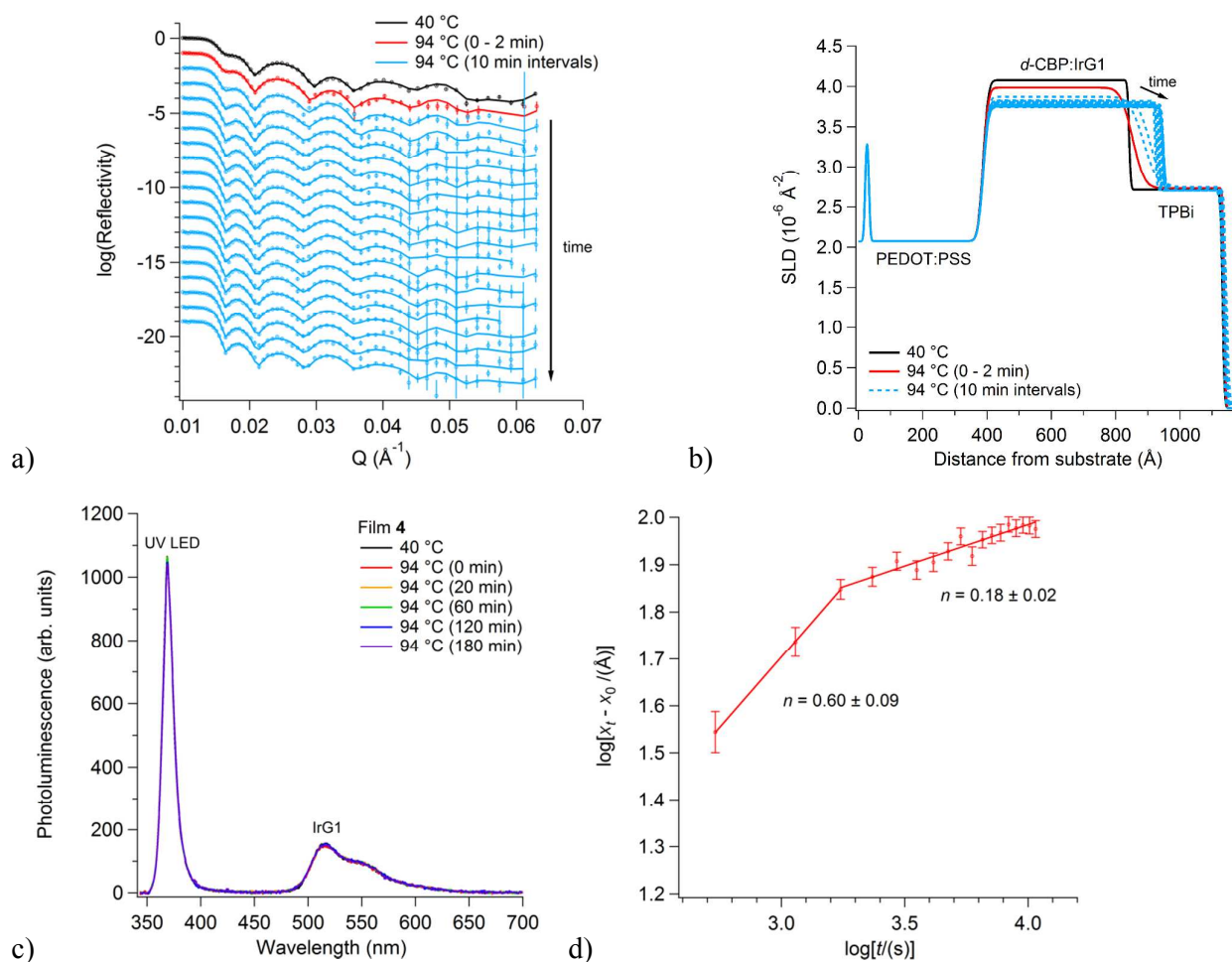


Figure S6 – a) Neutron reflectivity profiles, b) SLD *versus* distance from substrate plots, with the arrow representing the movement direction of the interface from which d) is determined, c) PL spectra, and d) plot relating the interface position to time (log-log plot) for Film 4 (PEDOT:PSS/*d*-CBP:IrG1/TPBi).

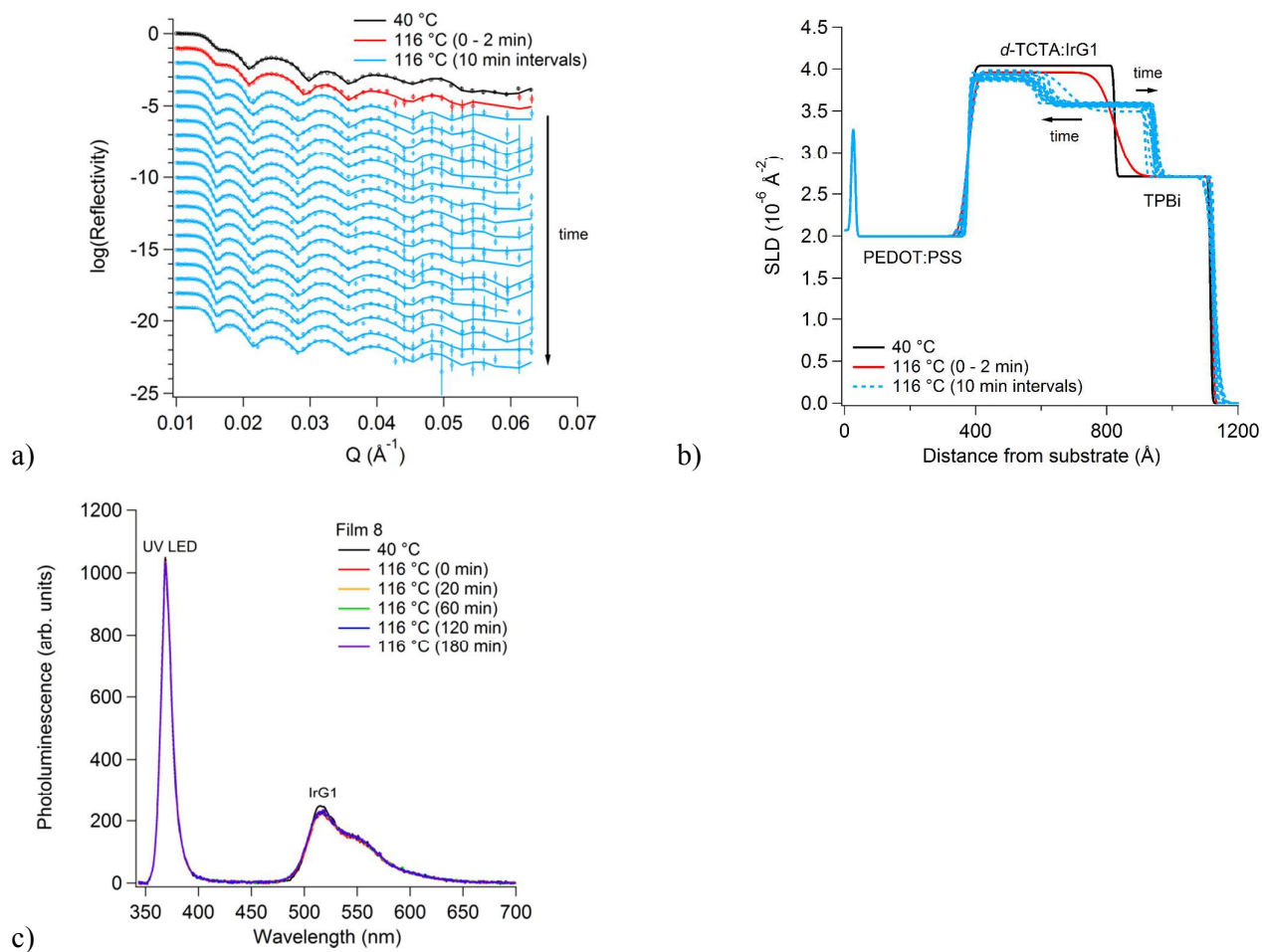


Figure S7 – a) Neutron reflectivity profiles, b) SLD *versus* distance from substrate plots (the arrows represent the movement direction of the interfaces), and c) PL spectra for Film 8 (PEDOT:PSS/*d*-TCTA:IrG1/TPBi).