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Supporting Information

Introducing neat fullerenes to improve the thermal stability of slot-die coated organic solar cells

Bradley Kirk, Xun Pan, Martyn Jevric, Gunther Andersson, and Mats R. Andersson*

Flinders Institute for Nanoscale Science and Technology, Flinders University, Sturt Road, Bedford Park, Adelaide, SA, 5042, Australia

Active material costs

Table S1: Estimated material cost of active layer materials based on a scale of 100 g. Prices gathered in December 2021, proved by Organic Nano Electronic (ONE=1) materials.

Material	USD/g
PBDB-T-2F (PM6)	650
PTB7-Th	550
PPDT2FBT	500
P3HT	120
Y6	650
ITIC	350
C_{60}	25
C_{70}	75
$PC_{61}BM$	150
$PC_{71}BM$	300

Slot-die theoretical thickness

To determine the theoretical thickness of the specific layers coated via slot-die coating, the following equations were employed:

$$d_{wet} = \frac{f}{S\,w} \qquad \text{(eq. S1)}$$

$$d_{dry} = \frac{f\ \ c}{S\,w\rho} \qquad \text{(eq. S2)}$$

Where d is the thickness (cm), f is the flow rate (cm^3 min^{-1}), S is the drum rotation speed (cm min^{-1}), w is the meniscus guide width (cm), c is the solid content in the ink (g cm^{-3}) and ρ is the density of the dried material[75, 76]. The estimated material density polymer to be 1.0 g mL^{-1}, while the densities for $PC_{61}BM$ & C_{60} are 1.5 g mL^{-1} [77] and 1.68 g mL^{-1} [78] respectively. The material densities of $PC_{71}BM$ & C_{70} are similar to $PC_{61}BM$ & C_{60} respectively.

Square-wave voltammetry

A 0.1M of tetrabutylammonium hexafluorophosphate (Bu_4NPF_6) in anhydrous acetonitrile was used as the electrolyte solution. The recorded square-wave voltammograms were referred to ferrocene/ferrocenium (Fc/Fc^+). The working electrode was coated with sample film by dipping it into the respective solutions. All materials were coated from chlorobenzene solutions. The electrolyte solution was purged with nitrogen gas for a minimum of 10 min prior to each measurement. During the measurements, the electrolyte solution surface was kept under nitrogen atmosphere to ensure inert conditions. Energy levels of the materials were calculated by setting the peak potential of Fc/Fc^+ versus the normal hydrogen electrode (NHE) to 0.630 V and the NHE versus the vacuum level to 4.5 V.[79-81] That is, $E_{HOMO} = -(E_{ox,peak} + 5.13)$ and $E_{LUMO} = -(E_{red,peak} + 5.13)$.

Dynamic mechanical thermal analysis

The blend solutions used were exactly the same as the ones used for solar cell fabrication. The prepared samples were dried under high vacuum of approximately 6×10^{-7} torr overnight. The measurements were performed in a strain-controlled tension mode at a heating rate of 3 °C min^{-1} from −110 to 300 °C under a nitrogen atmosphere at a frequency of 1 Hz. The annealing of samples was done in the DMA instrument under a nitrogen atmosphere for 30 min. The storage modulus (E'), loss modulus (E") and Tan δ were recorded as a function of temperature for each sample.

Solar simulation results

Table S2: **Device characteristics of PPDT2FBT:Fullerene devices under different coating conditions.**

Fullerene	Drum speed (m min^{-1})	Flow-rate (mL min^{-1})	Ink Conc (mg mL^{-1})	Theoretical thickness[1] (nm)	J_{SC} (mA cm^{-2})	V_{OC} (V)	FF	PCE (%)
$PC_{71}BM$	1.0	0.100	25.0	150	11.5 *±0.5*	0.77 *±0.01*	0.61 *±0.01*	5.40 *±0.27*
	1.0	0.200	25.0	300	14.4 *±0.5*	0.76 *±0.02*	0.54 *±0.02*	5.99 *±0.18*
	0.2	0.050	14.0	150	14.6 *±0.6*	0.76 *±0.01*	0.61 *±0.02*	6.71 *±0.16*
	0.2	0.050	14.0	200	17.8 *±0.2*	0.77 *±0.00*	0.59 *±0.00*	8.13 *±0.08*
	0.2	0.028	25.0	200	17.9 *±0.3*	0.77 *±0.01*	0.62 *±0.01*	8.49 *±0.07*
$PC_{61}BM$	1.0	0.100	14.0	150	10.5 *±0.5*	0.77 *±0.01*	0.57 *±0.04*	4.65 *±0.37*
	0.2	0.050	14.0	200	14.8 *±0.3*	0.80 *±0.00*	0.62 *±0.01*	7.27 *±0.09*
	0.2	0.035	25.0	200	15.9 *±0.6*	0.78 *±0.01*	0.61 *±0.01*	7.63 *±0.20*

[1] Thermal thickness calculated using equation 2

Average Cell No.: 6 cells / Device Area: 0.1 cm^2/ Inverted devices

Table S3: Device characteristics of PPDT2FBT:$PC_{61}BM$ devices aging at room temperature under dark nitrogen conditions.

Aging (hr)	J_{SC} (mA cm^{-2})		V_{OC} (V)		FF		PCE (%)	
0.00	15.5	*± 0.2*	0.792	*± 0.00*	0.62	*± 0.00*	7.56	*± 0.11*
23.50	14.0	*± 0.2*	0.788	*± 0.00*	0.66	*± 0.01*	7.29	*± 0.09*
47.75	14.1	*± 0.3*	0.790	*± 0.00*	0.64	*± 0.01*	7.14	*± 0.12*
72.50	14.6	*± 0.3*	0.790	*± 0.01*	0.62	*± 0.01*	7.17	*± 0.12*
96.00	14.4	*± 0.4*	0.789	*± 0.01*	0.63	*± 0.01*	7.15	*± 0.31*
168.0	14.4	*± 0.1*	0.789	*± 0.00*	0.63	*± 0.01*	7.14	*± 0.14*
194.0	13.9	*± 0.4*	0.787	*± 0.00*	0.63	*± 0.01*	6.87	*± 0.08*
218.0	13.7	*± 0.3*	0.787	*± 0.01*	0.62	*± 0.02*	6.71	*± 0.12*
240.0	14.0	*± 0.3*	0.767	*± 0.04*	0.64	*± 0.04*	6.87	*± 0.16*
264.0	13.1	*± 0.3*	0.784	*± 0.01*	0.61	*± 0.01*	6.23	*± 0.12*
337.0	13.8	*± 0.2*	0.782	*± 0.00*	0.61	*± 0.02*	6.56	*± 0.13*
361.0	14.1	*± 0.5*	0.785	*± 0.01*	0.62	*± 0.01*	6.82	*± 0.29*
385.0	14.1	*± 0.6*	0.787	*± 0.01*	0.60	*± 0.03*	6.66	*± 0.14*

Average Cell No.: 6 cells / Device Area: 0.1 cm^2/ Inverted devices

Table S4: Device characteristics of PPDT2FBT:$PC_{61}BM$ devices aging at 85 °C under dark nitrogen conditions.

Aging (hr)	J_{SC} (mA cm^{-2})		V_{OC} (V)		FF		PCE (%)	
0	14.6	*± 0.4*	0.775	*± 0.01*	0.57	*± 0.02*	6.39	*± 0.13*
1	14.7	*± 0.4*	0.783	*± 0.01*	0.60	*± 0.01*	6.83	*± 0.19*
3	15.3	*± 0.9*	0.784	*± 0.00*	0.60	*± 0.01*	7.15	*± 0.50*
21	16.1	*± 0.8*	0.777	*± 0.00*	0.58	*± 0.02*	7.18	*± 0.41*
44	16.1	*± 0.3*	0.781	*± 0.00*	0.59	*± 0.01*	7.37	*± 0.22*
67	15.4	*± 0.3*	0.775	*± 0.00*	0.56	*± 0.01*	6.64	*± 0.05*
131	15.5	*± 0.9*	0.775	*± 0.00*	0.56	*± 0.02*	6.69	*± 0.27*
154	14.1	*± 0.7*	0.776	*± 0.01*	0.55	*± 0.02*	5.96	*± 0.30*
174	14.2	*± 0.6*	0.753	*± 0.06*	0.57	*± 0.06*	6.05	*± 0.28*
222	14.1	*± 0.5*	0.780	*± 0.00*	0.56	*± 0.01*	6.15	*± 0.30*
293	12.9	*± 0.3*	0.778	*± 0.00*	0.54	*± 0.01*	5.44	*± 0.20*
323	12.4	*± 0.6*	0.780	*± 0.00*	0.56	*± 0.01*	5.39	*± 0.23*
344	13.0	*± 0.7*	0.756	*± 0.05*	0.54	*± 0.01*	5.28	*± 0.50*
366	13.4	*± 0.5*	0.781	*± 0.00*	0.53	*± 0.01*	5.52	*± 0.22*
388	12.5	*± 0.7*	0.773	*± 0.01*	0.53	*± 0.02*	5.10	*± 0.15*

Average Cell No.: 6 cells / Device Area: 0.1 cm^2/ Inverted devices

Table S5: Device characteristics of PPDT2FBT:Fullerene devices aging at 120 °C under dark nitrogen conditions.

PPDT2FBT:$PC_{61}BM$ (1:2)									
Aging (hr)	J_{SC} (mA cm^{-2})		V_{OC} (V)		FF		PCE (%)	
0.00	13.8	*± 0.4*	0.78	*± 0.00*	0.61	*± 0.02*	6.52	*± 0.26*
1.00	14.9	*± 0.3*	0.78	*± 0.00*	0.57	*± 0.00*	6.56	*± 0.14*
4.00	13.7	*± 0.3*	0.78	*± 0.01*	0.52	*± 0.01*	5.49	*± 0.12*
24.00	6.3	*± 0.4*	0.84	*± 0.02*	0.42	*± 0.01*	2.22	*± 0.08*
42.00	5.7	*± 0.6*	0.84	*± 0.02*	0.41	*± 0.02*	1.98	*± 0.21*
66.50	5.5	*± 0.1*	0.84	*± 0.00*	0.40	*± 0.01*	1.83	*± 0.11*
137.00	4.5	*± 0.3*	0.84	*± 0.07*	0.37	*± 0.03*	1.38	*± 0.04*
155.50	5.0	*± 0.4*	0.83	*± 0.01*	0.37	*± 0.00*	1.54	*± 0.11*
180.50	4.7	*± 0.4*	0.83	*± 0.01*	0.37	*± 0.02*	1.46	*± 0.13*
PPDT2FBT:$PC_{61}BM$:C_{60} (1:1.9:0.1)								
Aging (hr)	J_{SC} (mA cm^{-2})		V_{OC} (V)		FF		PCE (%)	
0.00	15.6	*± 1.1*	0.76	*± 0.01*	0.57	*± 0.01*	6.72	*± 0.24*
1.00	14.0	*± 0.9*	0.74	*± 0.02*	0.49	*± 0.02*	5.02	*± 0.22*
4.00	10.7	*± 0.4*	0.80	*± 0.01*	0.47	*± 0.01*	4.00	*± 0.08*
24.00	7.9	*± 0.2*	0.80	*± 0.02*	0.45	*± 0.01*	2.84	*± 0.11*
45.25	7.1	*± 0.3*	0.81	*± 0.02*	0.43	*± 0.01*	2.51	*± 0.03*
PPDT2FBT:$PC_{61}BM$:C_{70} (1:1.9:0.1)								
Aging (hr)	J_{SC} (mA cm^{-2})		V_{OC} (V)		FF		PCE (%)	
0.00	14.7	*± 0.6*	0.74	*± 0.00*	0.54	*± 0.02*	5.82	*± 0.07*
1.00	14.9	*± 0.3*	0.74	*± 0.01*	0.54	*± 0.01*	5.92	*± 0.06*
4.00	13.7	*± 0.6*	0.73	*± 0.01*	0.52	*± 0.01*	5.20	*± 0.19*
24.00	9.3	*± 0.7*	0.74	*± 0.03*	0.47	*± 0.01*	3.22	*± 0.25*
42.00	7.3	*± 0.6*	0.78	*± 0.02*	0.44	*± 0.01*	2.50	*± 0.18*
66.50	6.8	*± 0.3*	0.79	*± 0.01*	0.41	*± 0.01*	2.21	*± 0.10*
137.00	6.0	*± 0.3*	0.81	*± 0.01*	0.39	*± 0.02*	1.90	*± 0.14*
155.50	6.0	*± 0.1*	0.80	*± 0.01*	0.40	*± 0.01*	1.92	*± 0.07*
180.50	6.0	*± 0.3*	0.80	*± 0.01*	0.40	*± 0.02*	1.90	*± 0.13*

Average Cell No.: 6 cells / Device Area: 0.1 cm^2/ Inverted devices

Square-wave results

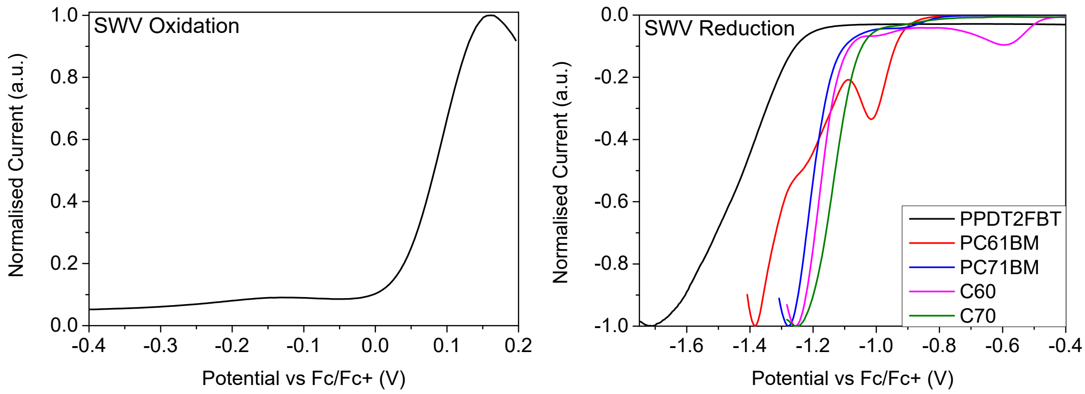

Figure S1: Oxidation square-wave curve of PPDT2FBT (LEFT), as well as Reduction square-wave curves of PPDT2FBT, $PC_{61}BM$, $PC_{71}BM$, C_{60}, and C_{70} (RIGHT)

Solar simulation light curves

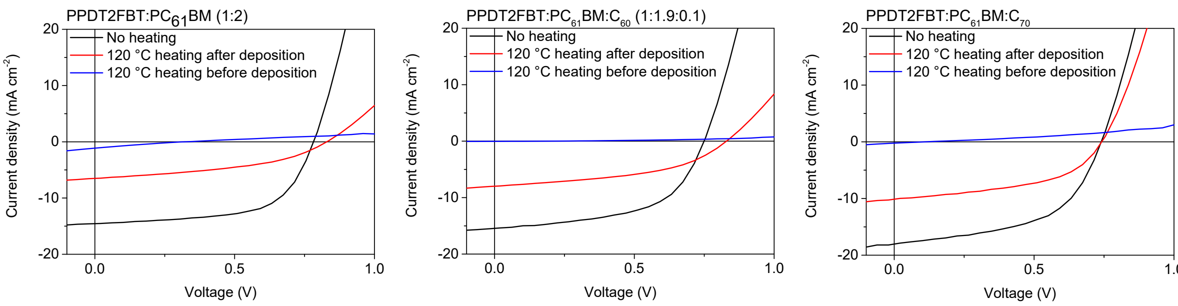

Figure S2: Light current-density / Voltage (JV) curve of PPDT2FBT:$PC_{61}BM$ (1:2) (LEFT), PPDT2FBT:$PC_{61}BM$:C_{60} (1:1.9:0.1) (MIDDLE) & PPDT2FBT:$PC_{61}BM$:C_{70} (1:1.9:0.1) (RIGHT) after experiencing different heating conditions, prior/post to MoO_X/Al deposition

Scanning electron microscopy images

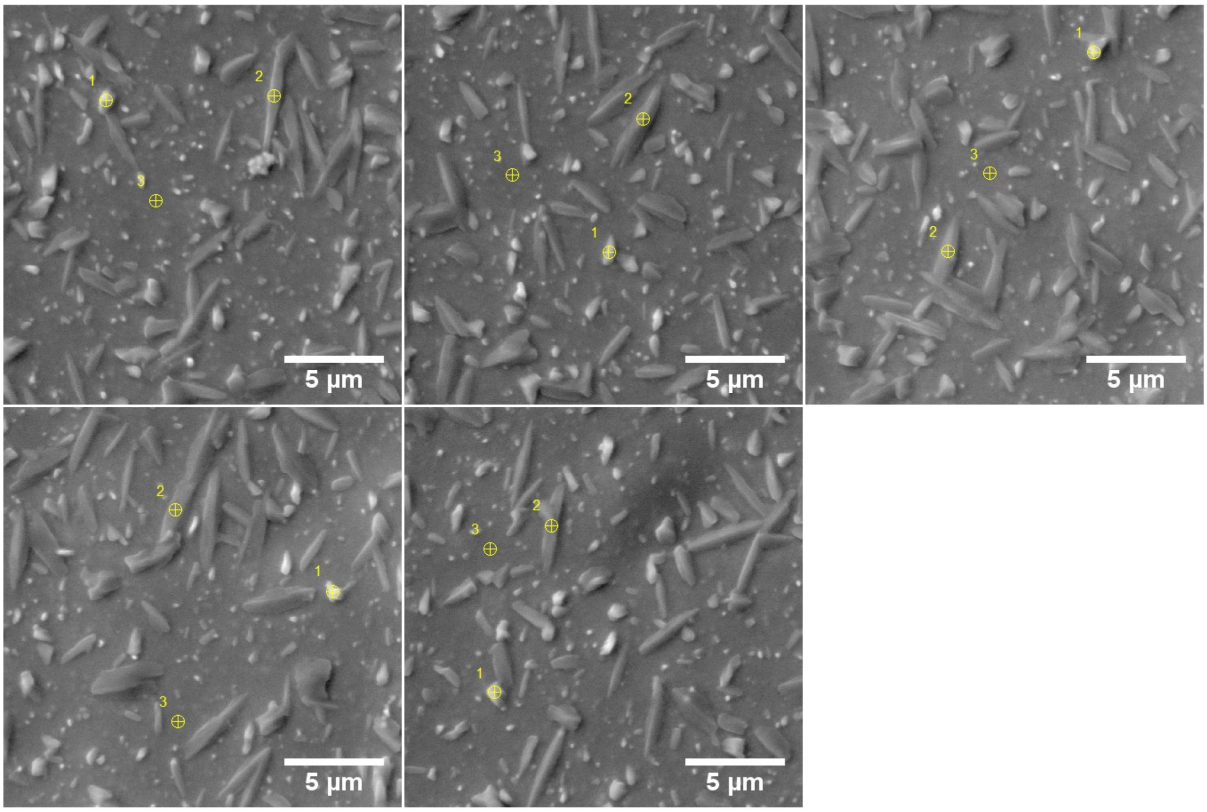

Figure S3: Scanning Electron Microscopy Image of the PPDT2FBT:$PC_{61}BM$ surface after 24 hours at 120 °C with marked locations of Auger Spectroscopy was measured.

Auger electron spectroscopy spectra

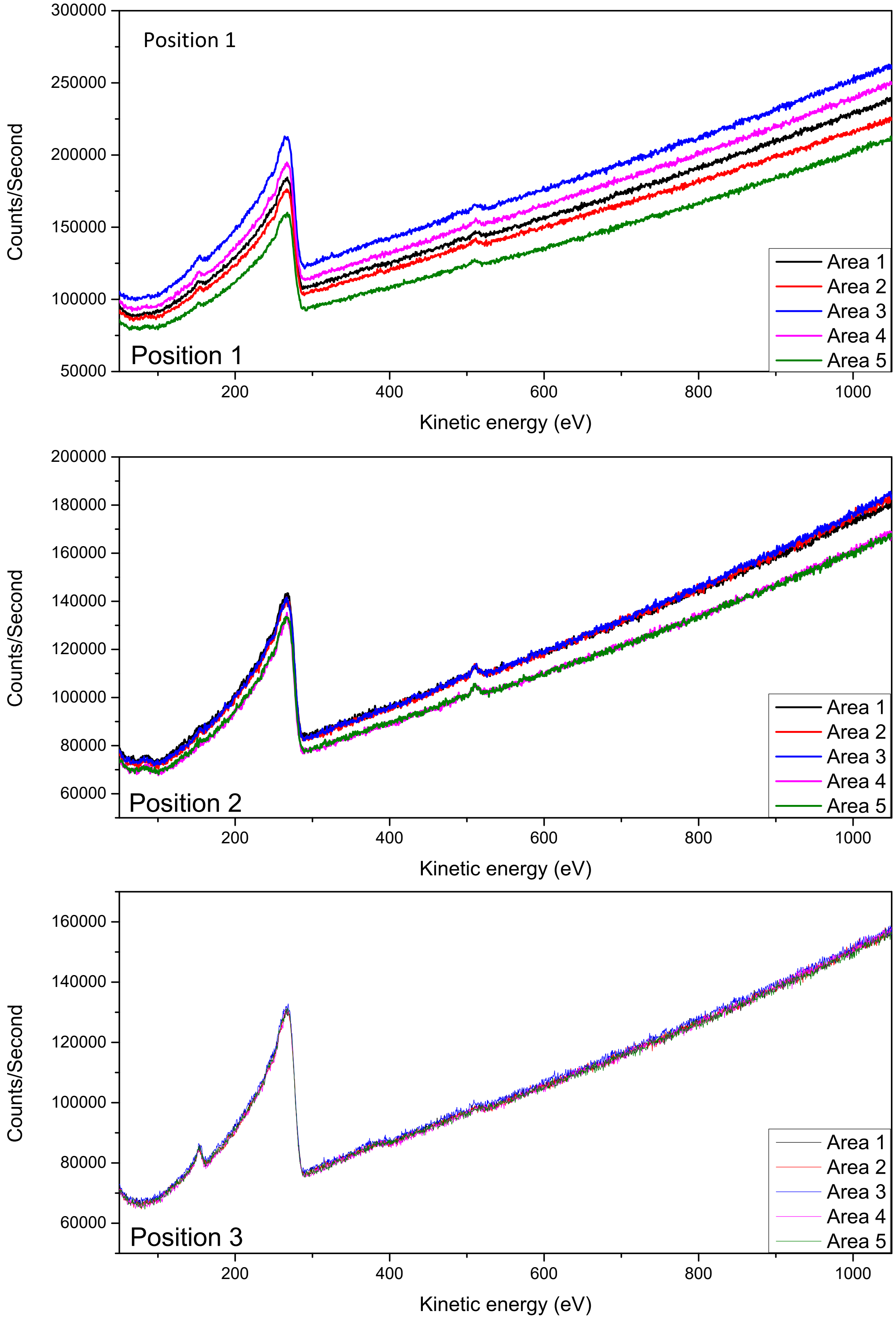

Figure S4: Raw auger electron spectra of 3 positions (five different point scans per position).

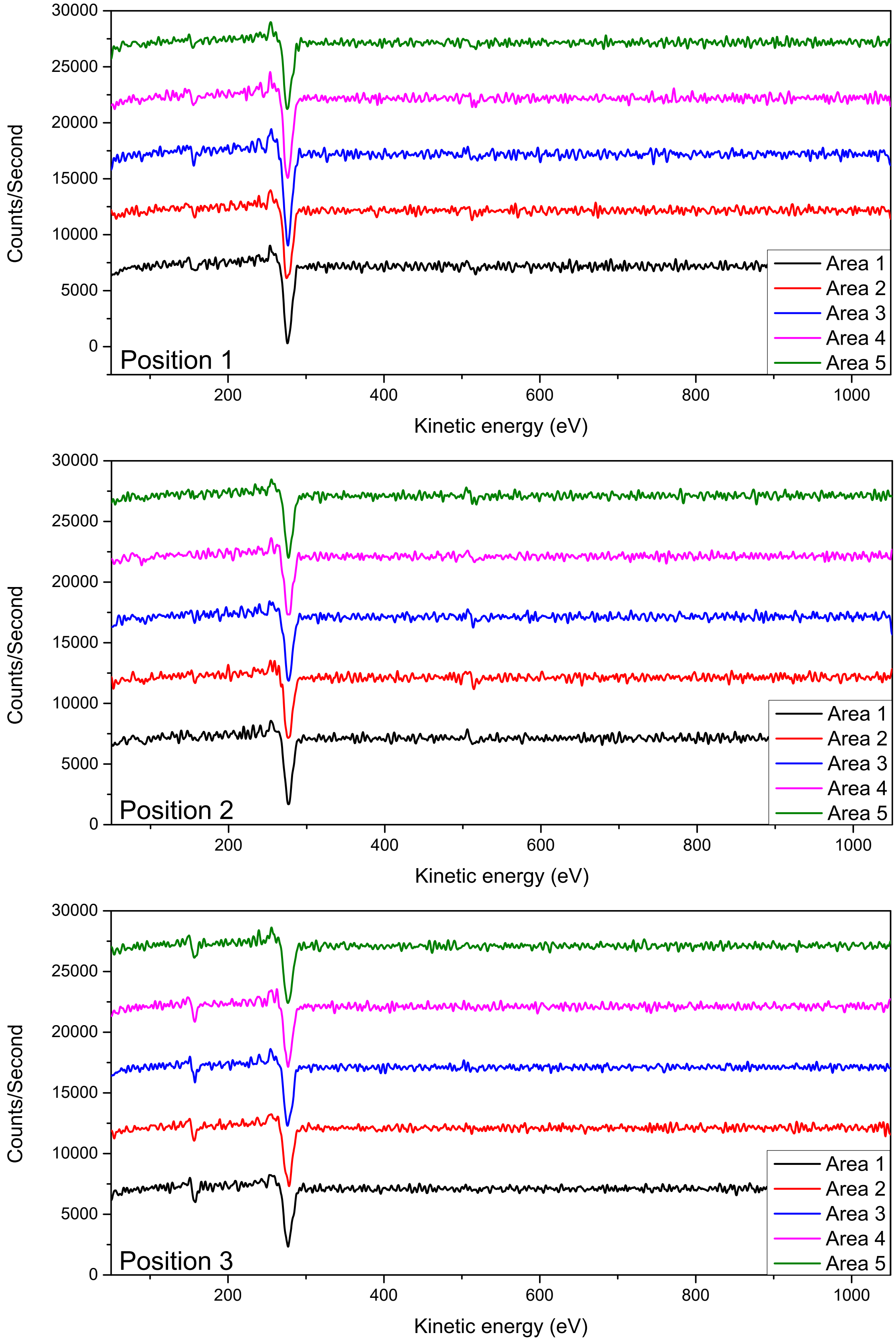

Figure S5: Auger electron spectroscopy scans of 3 positions (five-point scan areas per position). Spectra has been smoothed (9-point Savitzky-Golay) and derived (5-point)