## Supplemental Information for "DLC-Engineered Flat Gravure Surface: Enabling Sustainable Fabrication to Replace chrome for Printing Conductive Line Electrodes in Flexible Electronics"

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Trial.	Experiment Name		Print Cond	Defenence Figure	
No.	Experiment Name	Blade Type	Ink Type	Substrate Type	Reference rigure
T1	Trial One Print Experiment	Metal	Black60	White PET	Figure S-17 and S-18
T2	Trial Two Print Experiment	Metal	Black60	Clear PET	Figure S-19 and S-20
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T4	Trial Four Print Experiment	Metal	ECI7007	Clear PET	Figure S-23 and S-24
T5	Trial Five Print Experiment	Plastic	Black60	White PET	Figure S-25 and S-26
T6	Trial Six Print Experiment	Plastic	Black60	Clear PET	Figure S-27 and S-28
T7	Trial Seven Print Experiment	Plastic	ECI7007	White PET	Figure S-29 and S-30
T8	Trial Eight Print Experiment	Plastic	ECI7007	Clear PET	Figure S-31 and S-32

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3D	Parameter symbol and its	Description:		
parameter filter tools:	parameter name given in parenthesis:			
S Parameters– Height	$S_a$ (average roughness in nm) $S_q$ (average root mean square roughness in nm) $S_z$ (maximum height of the surface in nm).	The $S_a$ and $S_q$ parameters represent an overall measure of the texture comprising the surface, while $S_z$ represents the the maximum height of the surface.		
S Parameter– Hybrid tool	S <sub>dq</sub> (average root mean square slope in degree) S <sub>dr</sub> (developed interfacial area ratio)	$S_{dq}$ is the root mean square (RMS) surface slope comprising the surface, evaluated over all directions. $S_{dr}$ is the developed interfacial area ratio expressed as the percentage of the additional surface area contributed by the texture compared to an ideal plane that is the size of the measurement region		
Rz Analysis (Legacy parameter & not a 3D parameter)	Rz (average roughness)	Used for capturing the average of the 10 highest to 10 lowest points perpendicular to the surface		

**Table S-1**. Names of 3D parameter filter tools used as Analyzer algorithm in Vision 64 software to evaluate the OPDx file that has 3D surface data of the plain portion of chrome and standard DLC layers



Figure S-1. Bruker Images of chrome Surface. (A) Gray Scale Image and (B) 3D Contour Color Plot.



*Figure S-2*. Bruker Images of standard DLC Surface. (A) Gray Scale Image and (B) 3D Contour Color Plot.

Surface Free Energy (SFE) and Contact Angle (CA) on chrome Surface:									
Live view	🔲 🖬 💽 📓 🔟 🗃 🔍 🖂	Live view 🔀 🚺 🕸 🔍 🗔							
10.5 mm,	· 83 86 20	. <u>0.5 mm</u> , 							
76.0° Measurement: ThinkLab-Chrome Step: 4 Temperatu	76.4° re: 20.0 °C Substance: water (Air) 10 FPS	53.4° 53.3° Measurement: ThinkLab-Chrome Temperature: 20.0 °C diiodo-methane (Air) 10 FPS							
(A	) Contact Angle of test l	liquids on chrome surface.							
SFE calculation		Wetting envelope							
Model OWRK •	Substance   Mean CA [º]	SFT total [mN/m] ▼ Target contact angle 0  °							
Use error weighting 🔽	o 🔽 water 80.32 (±2.55)								
	x 🔽 diiodo-methane 54.29 (±1.50)								
Correlation coefficient : 1.00 $(1+\cos\theta)\sigma_1$	Calculation was successful.	20 -							
$\frac{2\sqrt{\sigma_1^D}}{2\sqrt{\sigma_1^D}}$	SFE total 36.98 ±1.90 mN/m								
12 - 8 - <b>0</b>	SFE disperse 31.85 ±0.86 mN/m								
	SFE polar 5.13 ±1.04 mN/m								
0 0.4 0.8 1.2 1.	6	0 5 10 15							
$\sqrt{\sigma_1^r}/\sigma_1^D$		SFT polar [mN/m]							
(B) Surface Free Energy of chrome surface and its wetting envelop.									

**Figure S-3**. Contact Angle and Surface Free Energy measured on chrome Surface using the KRÜSS MSA (Mobile Surface Analyzer). (A)Contact Angle of test liquids, water and diiodo-methane on chrome surface. (B) Surface Free Energy calculation (SFE total, SFE disperse and SFE polar) using Owens, Wendt, Rabel and Kaelble (OWRK) method along with wetting envelop plot.



**Figure S-4**. Contact Angle and Surface Free Energy measured on standard DLC surface using the KRÜSS MSA (Mobile Surface Analyzer). (A)Contact Angle of test liquids, water and diiodo-methane on standard DLC surface. (B) Surface Free Energy calculation (SFE total, SFE disperse and SFE polar) using Owens, Wendt, Rabel and Kaelble (OWRK) method along with wetting envelop plot.



**Figure S-5**. Contact Angle and Surface Free Energy measured on Clear PET plastic surface using the KRÜSS MSA (Mobile Surface Analyzer). (A)Contact Angle of test liquids, water and diiodo-methane on Clear PET plastic surface. (B) Surface Free Energy calculation (SFE total, SFE disperse and SFE polar) using Owens, Wendt, Rabel and Kaelble (OWRK) method along with wetting envelop plot.



**Figure S-6**. Contact Angle and Surface Free Energy measured on White PET plastic surface using the KRÜSS MSA (Mobile Surface Analyzer). (A)Contact Angle of test liquids, water and diiodo-methane on White PET plastic surface. (B) Surface Free Energy calculation (SFE total, SFE disperse and SFE polar) using Owens, Wendt, Rabel and Kaelble (OWRK) method along with wetting envelop plot.



*Figure S-7.* Surface Tension (ST) of ECI 7007 ink measured with the instrument First Ten Angstroms FTA200 using Pendant Drop Method.



*Figure S-8*. Surface Tension (ST) of Black 60 ink measured with the instrument First Ten Angstroms FTA200 using Pendant Drop Method.



*Figure S-9.* Contact Angle of Black 60 ink on White PET plastic surface. Measured with the instrument First Ten Angstroms FTA200 using Sessile Drop Method.



*Figure S-10*. Contact Angle of Black 60 ink on Clear PET plastic surface. Measured with the instrument First Ten Angstroms FTA200 using Sessile Drop Method.



*Figure S-11*. Contact Angle of ECI 7007 ink on White PET plastic surface. Measured with the instrument First Ten Angstroms FTA200 using Sessile Drop Method.



*Figure S-12.* Contact Angle of ECI 7007 ink on Clear PET plastic surface. Measured with the instrument First Ten Angstroms FTA200 using Sessile Drop Method.



*Figure S-13*. Contact Angle of Black 60 ink on chrome surface. Measured with the instrument First Ten Angstroms FTA200 using Sessile Drop Method.



*Figure S-14.* Contact Angle of Black 60 ink on standard DLC surface. Measured with the instrument First Ten Angstroms FTA200 using Sessile Drop Method.



*Figure S-15.* Contact Angle of ECI 7007 ink on chrome surface. Measured with the instrument First Ten Angstroms FTA200 using Sessile Drop Method.



*Figure S-16*. Contact Angle of ECI 7007 ink on standard DLC surface. Measured with the instrument First Ten Angstroms FTA200 using Sessile Drop Method.



**Figure S-17**. Trial One Print Experiment- 20 µm Grid (Blade Type: Metal, Ink Type: Black60, Substrate Type: White PET).



**Figure S-18**. Trial One Print Experiment- 30 µm Grid (Blade Type: Metal, Ink Type: Black60, Substrate Type: White PET).



*Figure S-19*. Trial Two Print Experiment - 20 μm Grid (Blade Type: Metal, Ink Type: Black60, Substrate Type: Clear PET).



**Figure S-20**. Trial Two Print Experiment - 30 µm Grid (Blade Type: Metal, Ink Type: Black60, Substrate Type: Clear PET).



*Figure S-21*. Trial Three Print Experiment - 20 μm Grid (Blade Type: Metal, Ink Type: ECI7007, Substrate Type: White PET).



*Figure S-22*. Trial Three Print Experiment - 30 μm Grid (Blade Type: Metal, Ink Type: ECI7007, Substrate Type: White PET).



**Figure S-23**. Trial Four Print Experiment - 20 µm Grid (Blade Type: Metal, Ink Type: ECI7007, Substrate Type: Clear PET).



*Figure S-24*. Trial Four Print Experiment - 30 μm Grid (Blade Type: Metal, Ink Type: ECI7007, Substrate Type: Clear PET).



*Figure S-25*. Trial Five Print Experiment - 20 μm Grid (Blade Type: Plastic, Ink Type: Black60, Substrate Type: White PET).



*Figure S-26*. Trial Five Print Experiment - 30 μm Grid (Blade Type: Plastic, Ink Type: Black60, Substrate Type: White PET).



**Figure S-27**. Trial Six Print Experiment - 20 µm Grid (Blade Type: Plastic, Ink Type: Black60, Substrate Type: Clear PET).



**Figure S-28**. Trial Six Print Experiment - 30 µm Grid (Blade Type: Plastic, Ink Type: Black60, Substrate Type: Clear PET).



*Figure S-29*. Trial Seven Print Experiment - 20 μm Grid (Blade Type: Plastic, Ink Type: ECI7007, Substrate Type: White PET).



**Figure S-30**. Trial Seven Print Experiment - 30 µm Grid (Blade Type: Plastic, Ink Type: ECI7007, Substrate Type: White PET).



*Figure S-31*. Trial Eight Print Experiment - 20 μm Grid (Blade Type: Plastic, Ink Type: ECI7007, Substrate Type: Clear PET)



**Figure S-32**. Trial Eight Print Experiment - 30 µm Grid (Blade Type: Plastic, Ink Type: ECI7007, Substrate Type: Clear PET)

## Statistical Analysis of Line Width

There were three print samples obtained for each of the trial experiments and associated print conditions. All of the three samples were used to collect 25 data point/replicates of line width data in one observation for each of the printed grids of 20  $\mu$ m and 30  $\mu$ m.

The statistical analysis of the line width data collected can be divided into two parts:

- 1. Comparing the DLC and chrome surfaces, using the same blade type, ink, and substrate for the eight trial experiments, for each individual line spacing under  $20\mu m$  and  $30\mu m$  grids, as well as for the combined grids spacing for the  $20\mu m$  and  $30\mu m$  line widths.
- 2. Performing a factorial analysis to identify which factors (surface, blade, substrate, ink, and grid) are significant to the line width.

Part 1 Results: -

Statistical analysis of data from eight experimental trials was conducted using Minitab software v21.2. A two-sample test for variance was applied to each comparison to determine whether to assume equal or unequal variance, followed by a two-sample t-test at a 5% significance level. The hypothesis that Standard DLC is better to chrome was tested. The results of the tests are tabulated in Tables S-1 and S-2. There were 25 observations for each grid spacing (1000  $\mu$ m, 500  $\mu$ m, and 200  $\mu$ m) for 20  $\mu$ m and 30  $\mu$ m line widths, resulting in a total of 75 observations. In Table S-2, results are shown for 20  $\mu$ m line widths.

uvi											
		$20 \mu\text{m}$ Grid 2-sample t-test p value ( $\alpha = 0.05$ : Reject Null hypothesis) *									
		20.000	-sample	1-test p val	$\frac{1}{20}$ um Crid $\frac{20}{20}$ um Crid $\frac{20}{20}$ um Crid				nid		
al No.		1000 μm Spacing		500 μm Spacing		20 µm Grid 200 µm Spacing		All Spacing			
	<b>C C</b>										
	Surface	(Sample)	5 5120	(samples size)		(Samples size		(Samples Size			
Ξ	Type		5 <u>)</u>		) D		)	Arra Mi dela	D		
<b>`</b>		Avg width	P	Avg width	P	Avg width	P	Avg width	P .		
		±Std Dev	value	±Std Dev	value	±Std Dev	value	±Std Dev	value		
1	chrome	46.9 ±1.3	0.000	53.4 ±0.6	0 000	58.5 ±1.1	0 000	53.0 ±4.9	0 000		
	Std DLC	51.4 ±0.7	0.000	57.2 ±1.7	0.000	59.9 ± <i>0.6</i>	0.000	56.2 ± <i>3.7</i>	0.000		
2	chrome	36.9 ±1.3	0.000	41.0 ±0.6	0.000	45.2 ± <i>0.5</i>	0.000	41.0 ± <i>3.6</i>	0.000		
2	Std DLC	41.7 ±0.4	0.000	44.9 ±0.6		51.7 ± <i>1.0</i>		46.1 ± <i>4.3</i>			
2	chrome	32.3 ±1.0	0.000	35.4 ±1.4	0.000	37.7 ±1.8	0.000	35.1 ±2.7	0.000		
5	Std DLC	36.3 ±1.2	0.000	39.4 ±1.8		40.3 ±2.5		38.7 ±2.5			
4	chrome	24.2 ±2.0	0.000	27.0 ±1.2	0.000	33.1 ±2.2	0 000	28.1 ±4.2	0.002		
4	Std DLC	26.7 ±1.6	0.000	28.6 ±1.2	0.000	34.4 ±1.4	0.000	29.9 ± <i>3.6</i>	0.005		
	chrome	39.6 ± <i>0.7</i>	0 0 6 0	42.0 ±0.3	0.000	48.6 ± <i>0.3</i>	0.000	43.4 ± <i>3.</i> 9	0.001		
5	Std DLC	39.4 ± <i>0.8</i>	0.000	45.0 ±0.3	0.000	53.9 ±1.9		46.1 ±6.1			
6	chrome	27.6 ±2.1	0.000	34.9 ±0.5	0 000	42.7 ±2.3	0.000	35.0 ±6.5	0.000		
b	Std DLC	31.7 ±0.6	0.000	38.1 ±0.6	0.000	50.7 ±1.6		40.1 ± <i>8.1</i>			

**Table S-2.** Statistical Comparisons of 20 µm Grids

7	chrome	22.2 ±0.8	0.000	26.1 ±0.5	0.000	33.7 ±1.1	0.000	27.3 ±4.9	0.000
	Std DLC	26.4 ±1.0		33.2 ±0.8		39.7 ±1.1		33.1 ±5.5	
0	chrome	20.4 ±1.0	0 2 2 2	22.7 ±1.0	0.000	29.8 ±1.8	0.000	24.3 ±4.2	0.010
0	Std DLC	20.5 ±1.1	0.333	24.6 ±1.1	0.000	32.5 ±0.9	0.000	25.9 ± <i>5.1</i>	0.019
	*Null Hansethesis share $(-)$ , $(-)$ DLC								

\*Null Hypothesis: chrome( $\sigma_1$ ) >= ( $\sigma_2$ ) DLC, Alternate Hypothesis chrome ( $\sigma_1$ ) < ( $\sigma_2$ ) DLC, Significance level: < $\alpha$  = 0.05

Note that for the 20  $\mu$ m line widths, all conditions show a significant higher line width when using standard DLC surface than when using chrome surface except for two conditions. In these conditions, the p value is higher than 0.05 for 20  $\mu$ m line width and 1000  $\mu$ m spacing in trial T5 and T8 as shown. In Table S-3, results are shown for 30  $\mu$ m line widths.

*Table S-3*. *Statistical Comparisons of 30 µm Grids* 

		30 μm Grid 2-sample t-test p value (<α=0.05: Reject Null hypothesis)*							
ial No.	Surface	30 μm Grid 1000 μm Spacing (Samples size N = 25)		30 μm Grid 500 μm Spacing (Samples size N = 25)		30 μm Grid 200 μm Spacing (Samples size N = 25)		30 μm Grid All Spacing (Samples Size in each Trial: N=75	
۲	1900	Avg Width	P	Avg Width	P	Avg Width	P	Avg Width	Р
		±Std Dev	value	±Std Dev	value	±Std Dev	value	±Std Dev	value
т1	chrome	67.1 ±1.4	0.000	71.3 ±2.0	0.000	85.2 ±0.7	0.000	74.5 ±7.9	0.000
	Std DLC	73.5 ±1.5	0.000	76.0 ±1.2	0.000	90.7 ±1.2	0.000	80.1 ± <i>7.8</i>	
т2	chrome	54.6 ±2.7	0.000	58.0 ±1.0	0.000	73.8 ±2.3	0.000	62.1 ±8.7	0.000
12	Std DLC	62.1 ±1.6		64.6 ±0.7		82.9 ±1.8		69.9 ±9.4	
т2	chrome	46.2 ±1.2	0.000	47.5 ±1.2	0.000	53.7 ±1.0	0.000	49.2 ± <i>3.5</i>	0.000
15	Std DLC	50.1 ±1.8		52.5 ±1.7		56.3 ±1.9		53.0 ± <i>3.1</i>	
ти	chrome	36.3 ±2.1	0.000	43.5 ±1.9	0.008	50.7 ±2.2	0.000	43.5 ±6.3	0.002
14	Std DLC	40.7 ±1.9		44.8 ±1.8		53.8 ± <i>2.9</i>		46.4 ±5.9	
т5	chrome	58.7 ±1.0	0.023	59.9 ±0.7	0.000	72.4 ±0.7	0.000	63.7 ±6.3	0.001
15	Std DLC	59.3 ±1.3	0.025	63.5 ±0.6	0.000	80.1 ±0.6		67.7 ±9.1	
та	chrome	47.9 ±1.3	0.406	51.8 ±1.0	0.000	67.8 ±0.8	0.000	55.8 ± <i>8.7</i>	0.012
10	Std DLC	48.0 ±1.2	0.400	56.1 ±0.8	0.000	74.6 ±0.7	0.000	59.6 ±11.2	
T7	chrome	38.2 ±2.4	0.000	40.7 ±1.1	0.000	49.5 ±0.7	0 000	42.8 ±5.1	0.000
<u> </u>	Std DLC	41.9 ±1.5	0.000	45.9 ±1.1	0.000	55.3 ±0.9	0.000	47.7 ±5.8	
тя	chrome	34.3 ±1.2	0.005	37.4 ±0.7	0 000	44.8 ±1.2	0.001	38.8 ±4.5	0.029
10	Std DLC	35.6 ±2.1	0.005	39.2 ±1.2	0.000	46.0 ±1.4	0.001	40.3 ±4.6	0.029

\*Null Hypothesis: chrome( $\sigma_1$ ) >= ( $\sigma_2$ ) DLC, Alternate Hypothesis chrome ( $\sigma_1$ ) < ( $\sigma_2$ ) DLC, Significance level: < $\alpha = 0.05$ 

Note that for the 30  $\mu$ m line widths, all conditions show a significant higher line width when using standard DLC surface than when using chrome surface except for one condition. In these conditions, the p value is higher than 0.05 for 30  $\mu$ m line width and 1000  $\mu$ m spacing

in trial T6 as shown. Although the standard DLC surface still indicates a higher line width, the p value is greater than 0.05, indicating less than 95% confidence.

The two-sample t-test results of the grid-to-grid comparison indicate that the line width measured on printed samples of standard DLC is higher than that of chrome, and is statistically significant with a p-value less than 0.005 in all cases, except for a few where the line width of the print samples obtained from both the standard DLC and chrome were the same. Moreover, the t-test results of all grids comparison were found to be statistically significant.

Part 2 Results: -



a. Residual plots –

*Figure S-33.* Residual plots for Line Width from Minitab. Results suggest acceptable normality, equity of variance and randomness of data.

The residual plot helps to assess whether all the assumptions of ANOVA are met.

- i. Normality: The normal probability plot and histogram suggest that normality is acceptable.
- ii. Equality of variance: The versus fit plot shows some clustering between the 40 and 60  $\mu m$  areas, but the variance appears to be equal.
- iii. Independence and randomness: The versus order plot doesn't have any discernible pattern, showing acceptable randomness, independence of data, and minimal bias.

## Conclusion – ANOVA appears to be valid.

b. Minitab output for the ANOVA –

Table S-4. Analysis of variance in Minitab

Source	DF	Adj SS	Adj MS	<b>F-Value</b>	<b>P-Value</b>
Surface	1	382.9	382.87	140.29	0.000
Blade	1	1257.1	1257.07	460.59	0.000
Substrate	1	1454.4	1454.38	532.89	0.000
Ink	1	7923.9	7923.92	2903.33	0.000
Grid	5	10858.6	2171.72	795.72	0.000
Surface*Blade	1	2.0	2.03	0.74	0.392
Surface*Substrate	1	2.0	2.00	0.73	0.396
Surface*Ink	1	12.3	12.27	4.50	0.038
Surface*Grid	5	11.6	2.33	0.85	0.519
Blade*Substrate	1	44.1	44.05	16.14	0.000
Blade*Ink	1	70.1	70.12	25.69	0.000
Blade*Grid	5	66.3	13.26	4.86	0.001
Substrate*Ink	1	64.1	64.08	23.48	0.000
Substrate*Grid	5	32.2	6.45	2.36	0.051
Ink*Grid	5	477.2	95.44	34.97	0.000
Error	60	163.8	2.73		
Total	95	22822.5			

The ANOVA results in Table S-4, indicate that the surface, blade, substrate, ink and grid factors all have a significant impact on line width, as the p-value is less than 0.005 for these factors. Additionally, the surface does have some impact on ink with respect to line width as shown in this p value. However, the surface does not have an impact on blade, ink or grid, as expected. There is significant interaction between the blade and the substrate, the ink, and the grid. This would indicate that the blade type effect and the effect on line width from the substrate, the ink, and the grid are interrelated. This is to be expected as the blade interaction with the blade is likely dependent on the chemical composition and properties (i.e. viscosity, mixing) of the ink likely affect interaction with the blade leading this this interaction in dependence. There is significant interaction between dependence of line width with substrate and ink with near-significant interrelation of dependence between the substrate and grid. This is to be expected as the ink interacts with the substrate differently based on the composition of the ink and the substrate, while the substrate can have some impact on the grid due to the substrate surface properties and chemical composition. Finally, the line width dependence on ink and grid are interrelated as the ink must have a relationship with grid. This is to be expected as the inks have much different viscosity, thereby affecting the grid line width that can be printed.