Electronic Supplementary Material (ESI) for:

Interface engineering of the moisture-induced ionic dielectric layer for low voltage, high-performance, organic field-effect transistors

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Characterization of Chicken Albumen:

Fourier-transform infrared spectroscopy (FTIR) was carried out (Figure S1) on albumen to study the presence hydrocarbon (–CH) and hydroxyl (–OH) stretching vibration. We have observed FTIR signals at 2950 cm^{-1} and 3350 cm^{-1} confirming the presence of –OH and –CH groups, respectively. The stretching vibration corresponding to the carboxyl (C=O) group has been observed at 1650 cm^{-1}. However, the other peaks as observed at 1545 cm^{-1} and 1440 cm^{-1} are due to the bending vibration of the amide (N–H) and carboxylate (C–O) group.[1] The FTIR spectrum confirms the presence of polar groups such as amide, carboxyl, and hydroxyl.

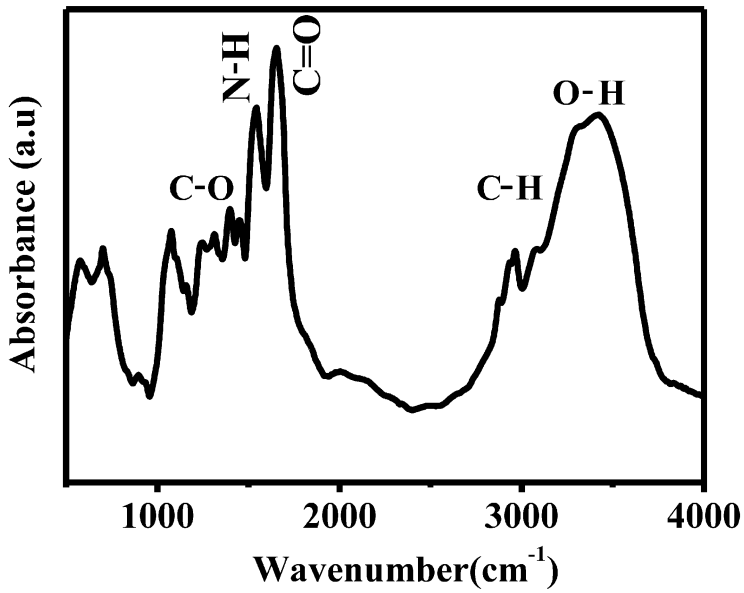

Fig. S1. FTIR spectrum of chicken albumen film showing the presence of the polar groups, like N–H, C=O, and –OH.

Chicken albumen consists of different proteins such as ovalbumin, ovotransferrin, and ovomucoid, etc. The molecular weight of chicken albumen is characterized by sodium dodecyl sulphate–polyacrylamide gel electrophoresis (SDS-PAGE). The corresponding SDS-PAGE

image for as collected chicken albumen is shown in Figure S2. The different bands corresponding to the presence of different proteins in chicken albumen are given in table 1 and its matches with the literature results[2]. Ovomucoid appeared at 35-40 kDa instated of 28 kDa in SDS-PAGE. It happened as the ovomucoid was may not completely be separated from ovalbumin.

Protein	Molecular weight (kDa)
Ovomucoid	30-40
Ovalumin	45
Ovotransferrin	76

Table S1: Molecular weight of measured proteins that present in chicken albumen.

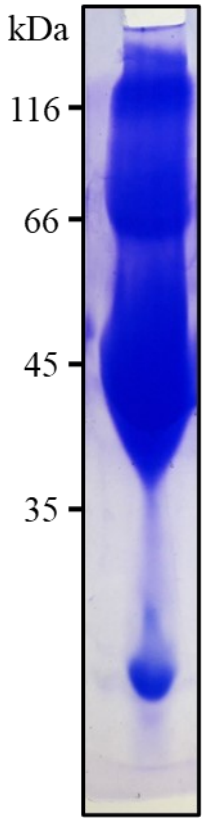

Fig. S2. SDS-PAGE image of as collected chicken albumen.

The x-ray photoelectron spectra (XPS) for carbon, nitrogen, and oxygen have been carried out on albumen film annealed at different temperatures (40ºC to 140ºC). The corresponding XPS spectra of C1s, N1s, and O1s are shown in Figure S3(a–c), respectively. These spectra reveal that variation of annealing temperature does not affect the binding energy of these atoms.

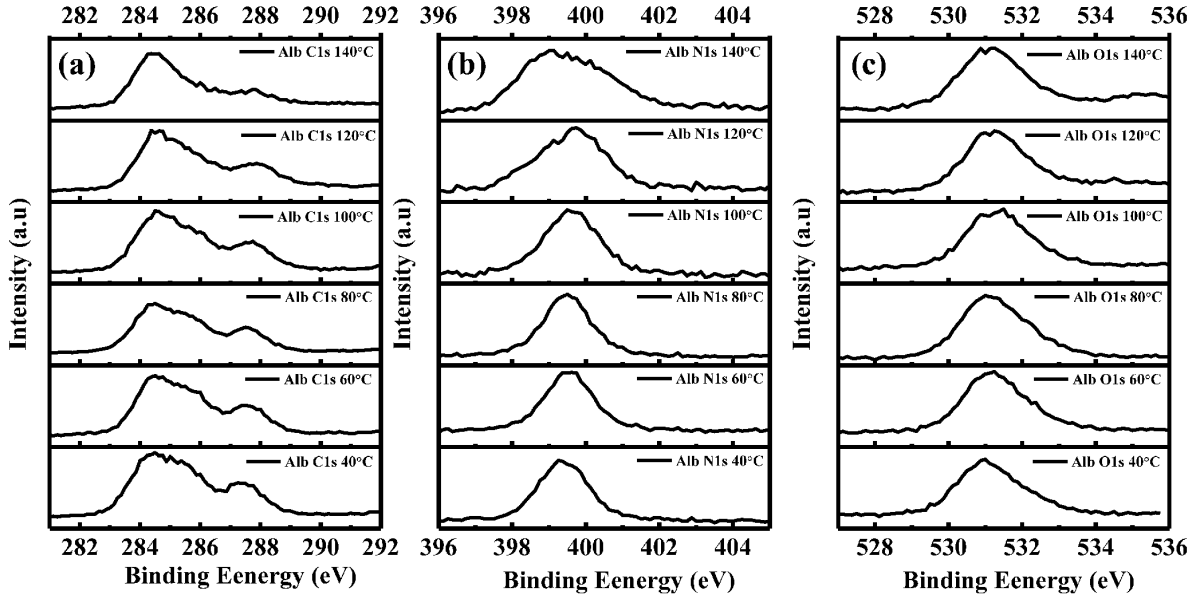

Fig. S3. High-resolution XPS spectra of (a) carbon (b) nitrogen (c) oxygen 1s spectra taken from chicken albumen film annealed at a temperature from 40 ºC to 140 ºC.

We have examined the percentage of weight loss of chicken albumen during annealing at three different temperatures (40 ºC, 100 ºC, and 140 ºC) upto a time duration from 0 to 90 min. It is presented in Figure S4. The loss of water molecules due to this heating process relatively lower at an annealing temperature of 40 ºC compear to the 100 ºC, and 140 ºC annealing temperature. It indicates that water molecules in chicken albumen not evaporated completely at 40 ºC and also crosslinking of sulfur atoms has not occurred at that temperature. However,

at annealing temperature 100 ºC and 140 ºC percentage of water molecules loss saturated quickly (within 20 min of annealing) and get enough time to crosslink the albumen film.

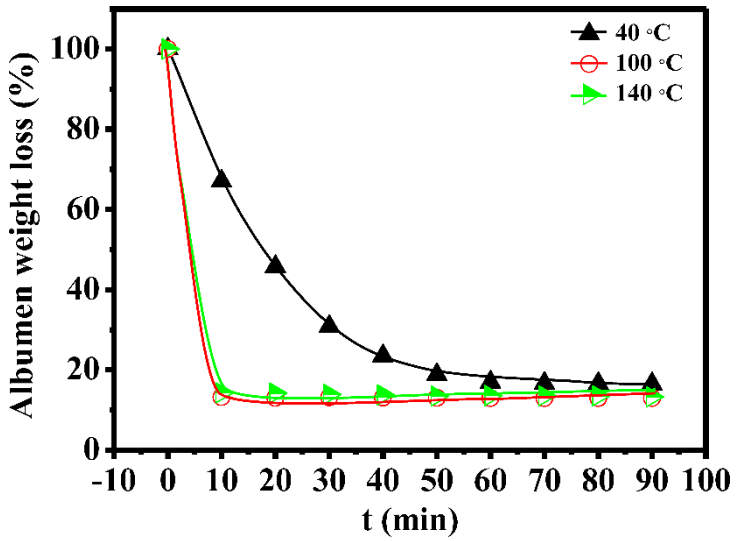

Fig. S4: Percentage of water loss from chicken albumen with time at annealing temperatures of 40 ºC, 100 ºC, and 140 ºC.

The FESEM images of pentacene film grown on albumen/Al/glass substrates have been taken at various annealing temperatures of albumen films (40ºC to 140 ºC). The corresponding images are presented in Fig. S5(a-f), respectively. Formation of the dendrite structure of pentacene films are visible for the samples up to 100 ºC anneal temperature of the albumen layers. However, the morphology of pentacene film changes for the samples annealed at 120 and 140 ºC. These morphologies are consistent with AFM results.

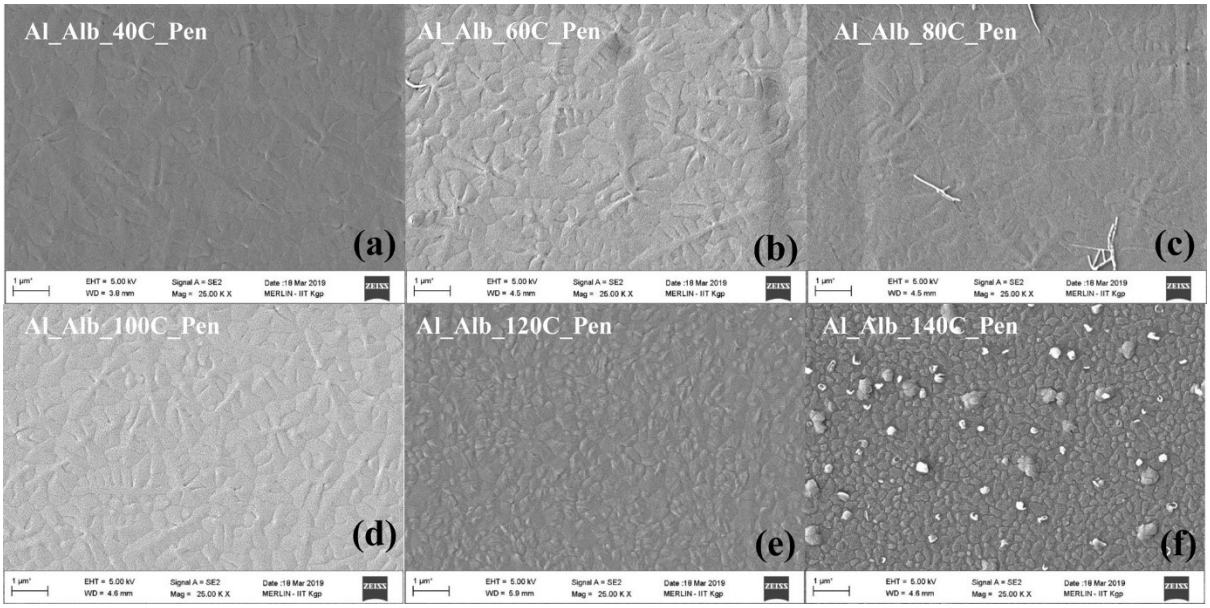

Fig. S5. FESEM images of pentacene films grown on albumen/Al surfaces annealed at (a) 40 ºC (b) 60 ºC, (c) 80 ºC, (d) 100 ºC, (e) 120 ºC and (f) 140 ºC for 90 minutes.

Surface roughness exponent (α) of albumen and pentacene film extracted from the linear portion of $G^{1/2}(r)$ vs r plots. In both of these films, we have measured the variation of α with annealing temperature. It is shown in Figure S6. The average value of α for both of these films is 0.85 (±0.06). A smaller value of α implies a rougher local surface whereas a larger value of it implies a smother local surface[3]. So, the α value for both of these films (Albumen and pentacene) is simply indicated the smoother local roughness of them.

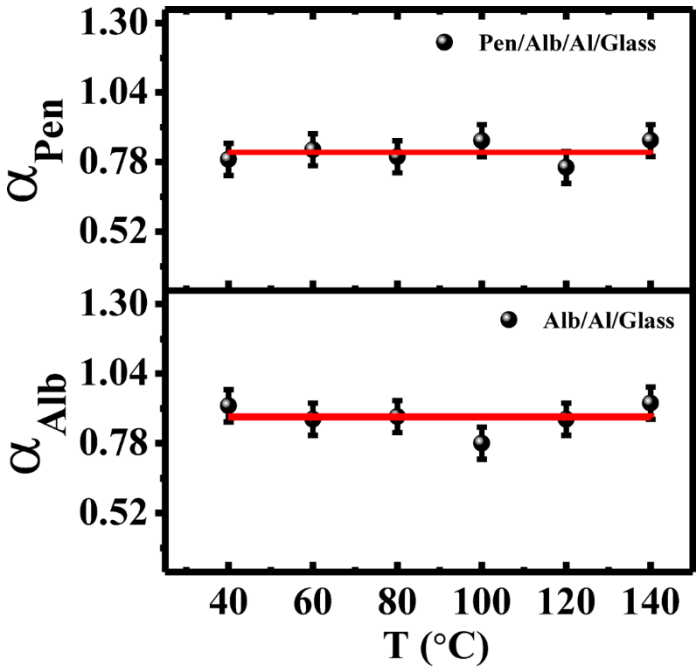

Fig. S6. Shows the variations of surface roughness exponent (α) of albumen and pentacene film for various annealing temperatures of the albumen layer.

In order to characterize the dielectric property of pristine and crosslinked chicken albumen, we have prepared two films by spin coating them on the aluminium surface. After spin-coating Sample 1 is kept at room temperature for 48 hours and Sample 2 is annealed at 100 ºC for 90 min. The variation of dielectric constant at various frequencies for pristine (Sample 1) and crosslinking (Sample 2) chicken albumen is shown in Figure S7. We have observed that at a lower frequency (<450 Hz) the dielectric constant of Sample 1 is higher than Sample 2. It is mainly due to the presence of water molecules (Dielectric constant of water 80.4) inside the Sample 1 is relatively higher than Sample 2. The dielectric constant of pristine albumen and crosslinking chicken albumen at 100 Hz and 1 kHz are given in below Table S2.

	100 Hz	1 kHz
Pristine albumen (25 ºC)	8.1	3.5
Crosslinked albumen (100 ºC)	4.4	4.2

Table S2. Dielectric constant of pristin and crosslinked chicken albumen.

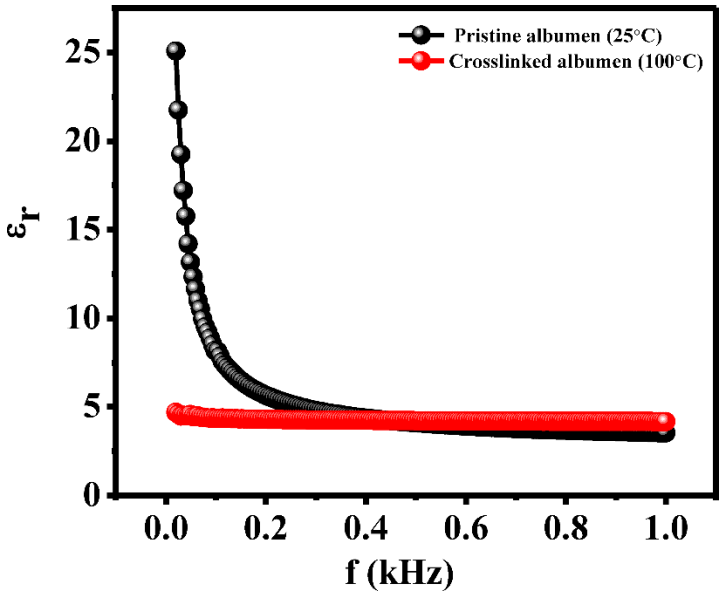

Fig. S7. Frequency-dependent dielectric constant variation for pristine and crosslinked chicken albumen film.

We have extracted the effective saturation field-effect mobility[2] of the device considering the capacitance of albumen film in the lower frequency region (2.5 to 10 Hz) annealed at a temperature of 100 ºC. The effective saturation mobility (μ_{eff}) of the device considering the capacitance at 2.5 Hz and 10 Hz are found to be 0.55 (± 0.08) cm^2/ Vs and 0.72 (± 0.08) cm^2/Vs, respectively. The corresponding frequency-dependent capacitance and effective mobility variation are presented in Figure S8.

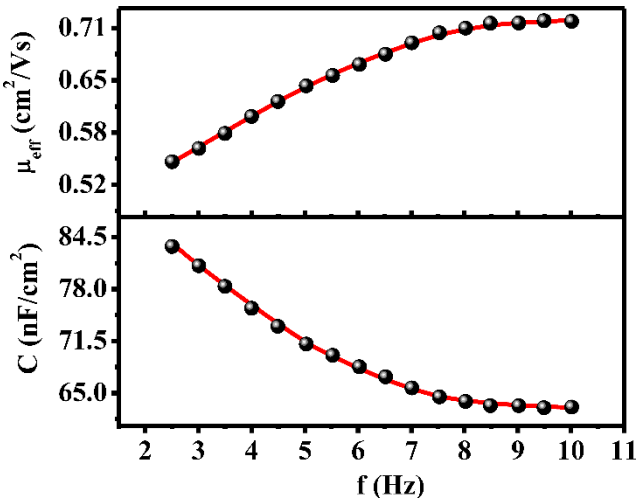

Fig. S8. Frequency-dependent capacitance and effective saturation mobility variation of chicken albumen-based OFETs.

The effective saturation device mobility[4] has also been measured for 119 devices in total to check the reproducibility of chicken albumen-based OFETs prepared in ambient conditions. The average device mobility considering the capacitance at 2.5 Hz is around 0.3 cm^2/Vs. The corresponding histogram distribution is presented in Figure S9.

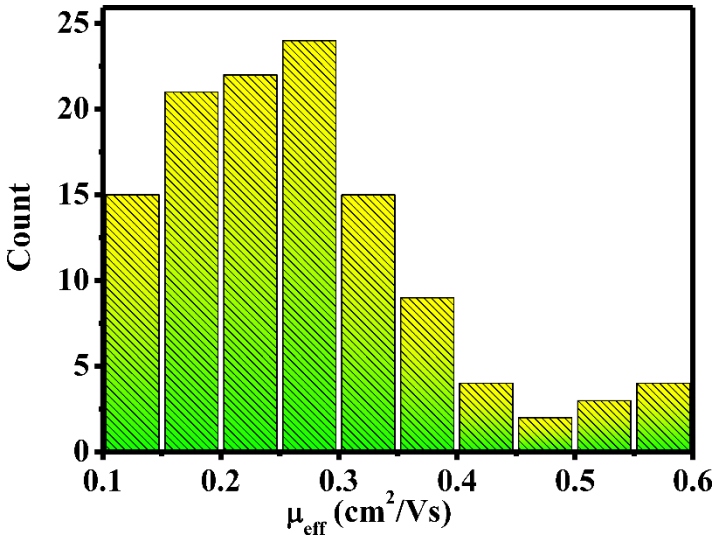

Fig. S9. Histogram distribution of effective device mobility of the chicken albumen-based OFETs.

It has been found that the device shows relatively poor performance when the correlation length (ξ) of the albumen surface increases beyond 205 nm. The performance of the device in terms of transconductance, threshold voltage, and coercive voltage with ξ is shown in Fig. S10 (a-c), respectively. The transconductance of the device increases with increasing ξ upto 205 nm (maximum transconductance 11.68 μS) but above that correlation length, it again decreases. However, the threshold voltage of the device exponentially changes, and coercive voltage increase linearly upto a correlation length of 205 nm.

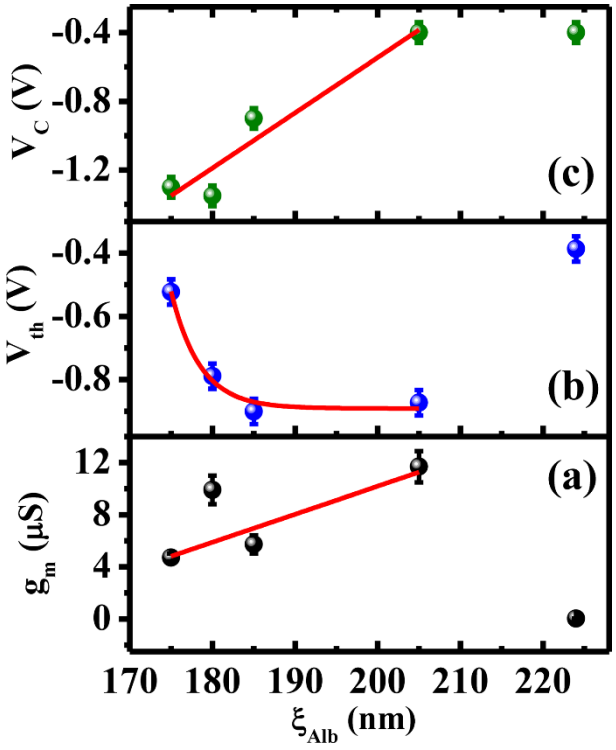

Fig. S10. Variation of device parameters (a) transconductance (b) threshold voltage and (c) coercive voltage with the correlation length of the albumen surface.

We have characterized the bias stress effect of the device by keeping a constant bias to the device. Fig S11a shows the variation of normalized device current with time at a constant V_{GS} and V_{DS} of –1.20 V. It shows an anomalous bias stress effect due to polarization of the polar group presence in chicken albumen (-NH2, -COOH). In order to characterize the stress

time effect on the device, we have taken transfer characteristics of the device after just applying bias stress for 10 min, 20 min, 30 min, 40 min, and 60 min. The corresponding transfer characteristics of the device are shown in Fig S11b. It has been observed that the threshold voltage of the device decreases slowly with the increment of the stress time. It is presented in Fig S11c. The traps presented in the dielectric-semiconducting interface fill up with charge carriers due to the polarization of the polar groups in presence of applied constant bias to the device.

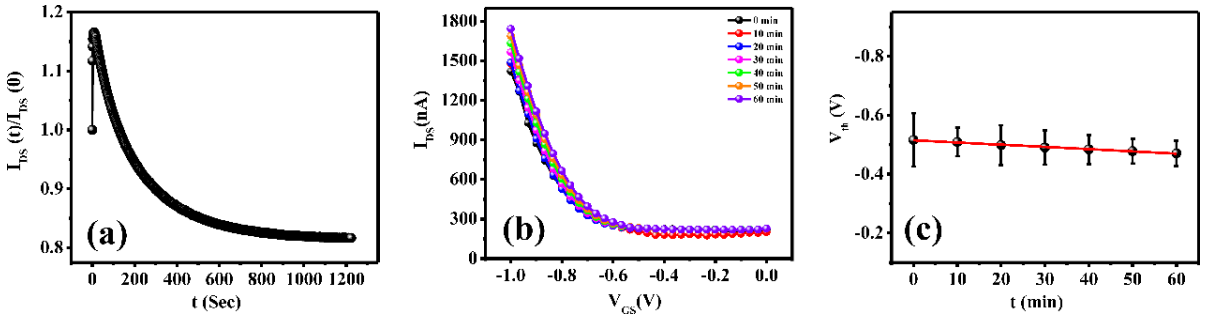

Fig S11. (a) Variation of normalized device current with time during a constant bias of V_{GS} and V_{DS} of –1.20 V. (b) Transfer characteristics of the device at different stress times. (c) Change in threshold voltage with stress time in albumen-based OFETs.

Reference

1. Y. C. Chen, H. C. Yu, C. Y. Huang, W. L. Chung, S. L. Wu and Y. K. Su, *Sci. Rep.*, 2015, **5**, 10022.

2. W. Chaiyasit, R. Brannan, D. Chareonsuk, W. Chanasattru, Comparison of Physicochemical and Functional Properties of Chicken and Duck Egg Albumens. Brazilian Journal of Poultry Science 2019, 21.

3. M. Pelliccione and T.-M. Lu, Evolution of thin film morphology: modeling and simulation, *Springer-Verlag*, New York, 2008.

4. H. H. Choi, K. Cho, C. D. Frisbie, H. Sirringhaus, V. Podzorov, Critical assessment of charge mobility extraction in FETs. *Nat Mater* **2017,** *17* (1), 2-7.