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Biogenic Fluorescent Protein-Silk Fibroin Phosphors for Highly Performing Light-Emitting Diodes

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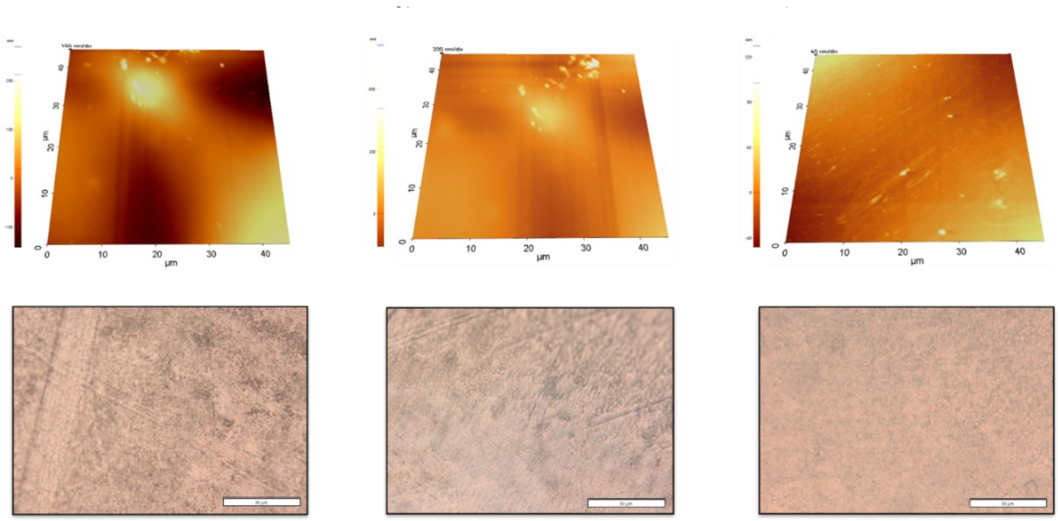

Figure S1. AFM topography images (top) and optical microscopy (x10) pictures (bottom) of films with 3% wt. SF (left), 6% (center) and 9% (right).

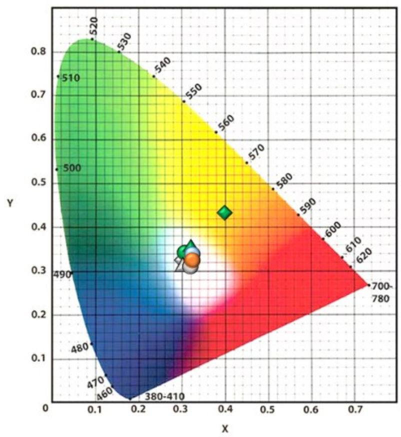

Figure S2. Color diagram representing the x/y CIE color coordinates of: i) fresh SF films (circle) with 3 (grey), 6 (orange), 9 (blue) wt. % of SF, ii) SF-films after thermal- (triangle) and photo-degradation (diamond) stress, and iii) fresh (circle), heated (triangle), and irradiated (diamond) FP-SF films (green).

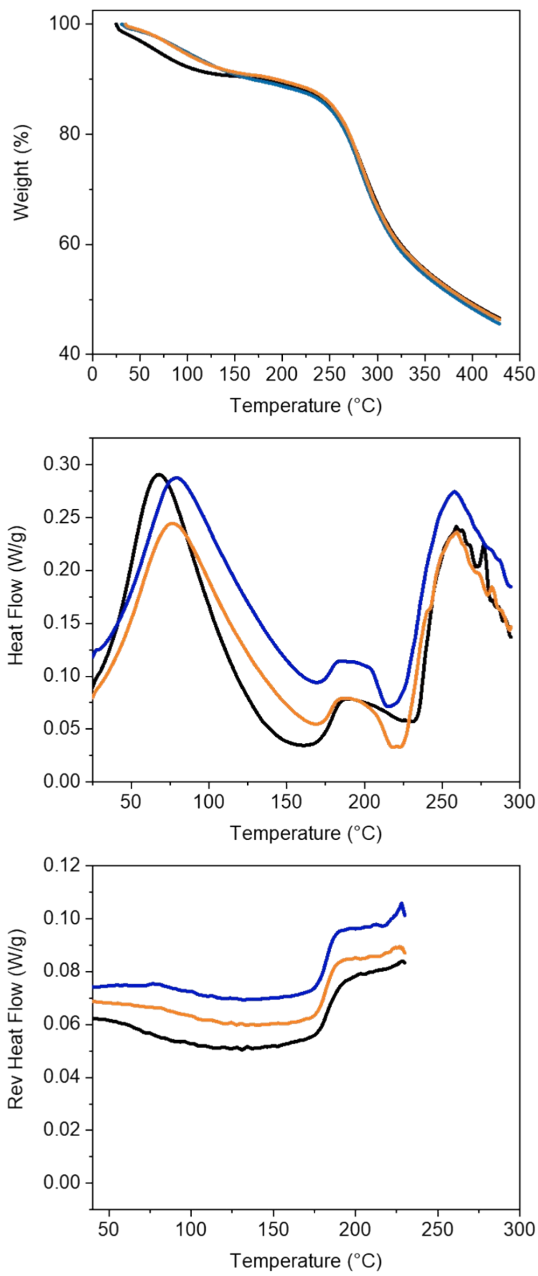

Figure S3. Calorimetry assays of TGA (top) and MDSC for the reversible (middle) and non-reversible (bottom) representations of fresh 3 (black), 6 (orange) and 9 (blue) wt. % SF films.

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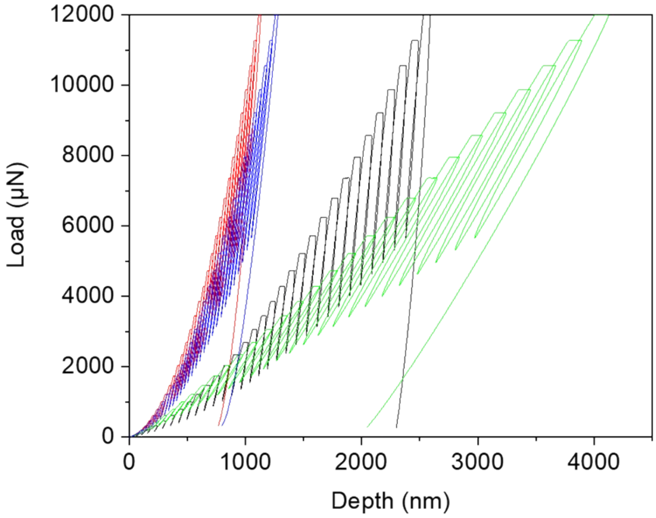

Figure S4. Representative load-displacement curves corresponding to cyclic nanoindentation tests performed using a maximum load of 12 mN on fresh 3 wt.% SF-film (red), FP-SF fresh (blue), heated SF-films (black), and irradiated SF-films (green).

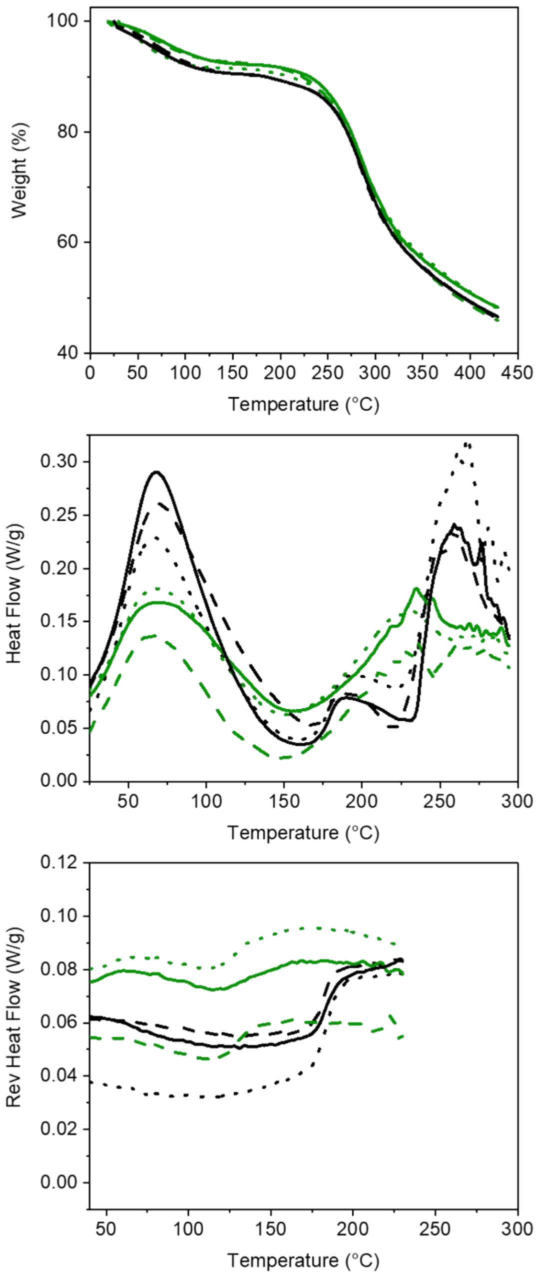

Figure S5. Calorimetry assays of TGA (top) and MDSC for the reversible (center) and non-reversible (bottom) representations of pristine SF films (black) and FP-doped ones (green) for fresh (solid line), photo- (dashed line), and thermal-degraded (dotted line).

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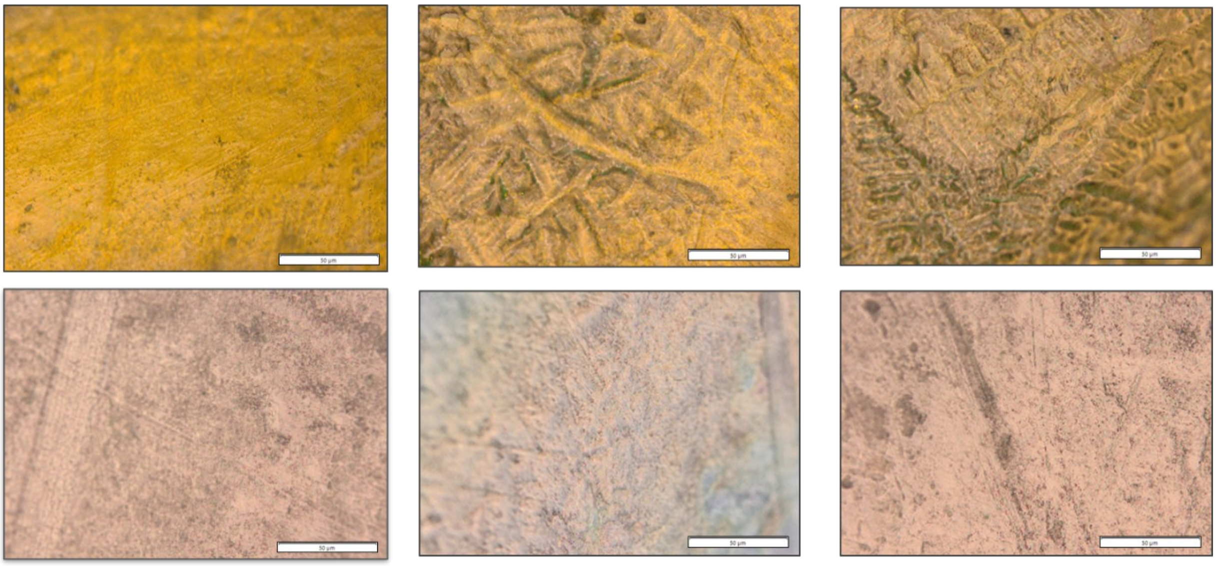

Figure S6. Top: Optical microscope (x10) images of fresh (left), heated (center), and irradiated (right) FP-SF (top) and SF (bottom) films.

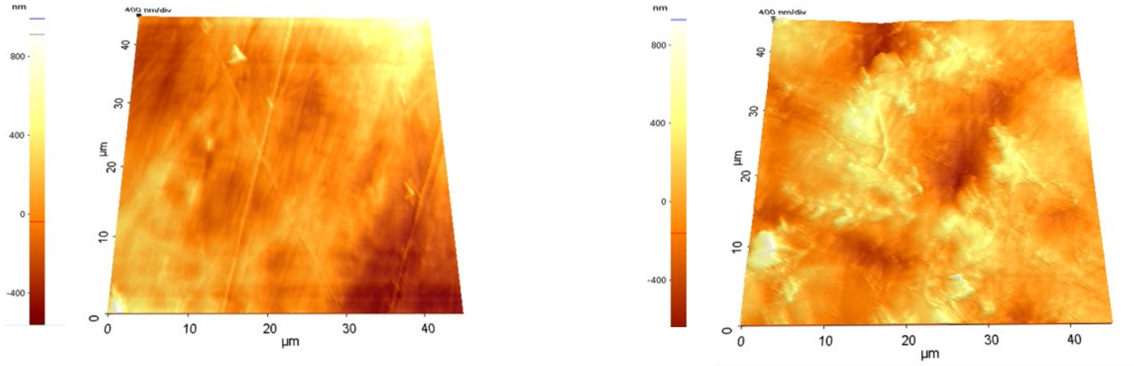

Figure S7. AFM topography pictures of heated (left) and irradiated (right) FP-SF filters.

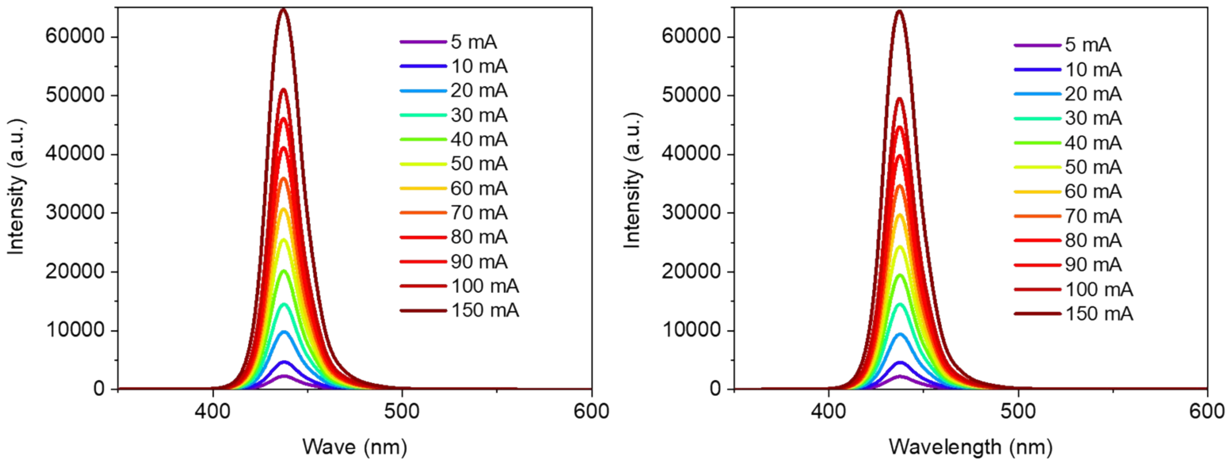

Figure S8. Electroluminescence spectra of a bare LED chip (left) and that covered with a SF-film (right) upon increasing the applied current.

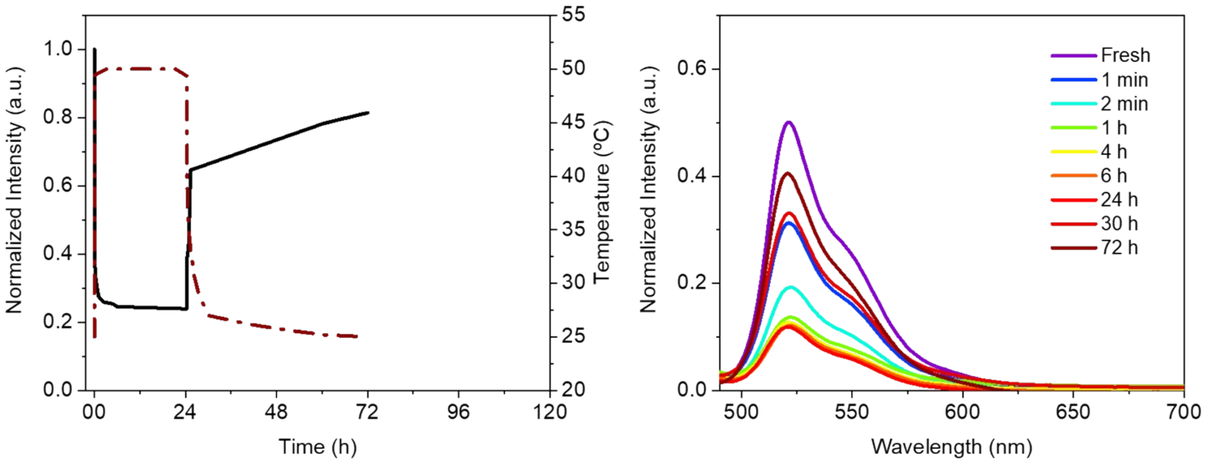

Figure S9. Thermal stability of FP-polymer bio-phosphor at a constant temperature (50 ºC) for 24 h and after cooling, monitoring the emission intensity (left), and emission band shape (right) upon excitation with a blue LED (450 nm) for 5 s every 10 min.

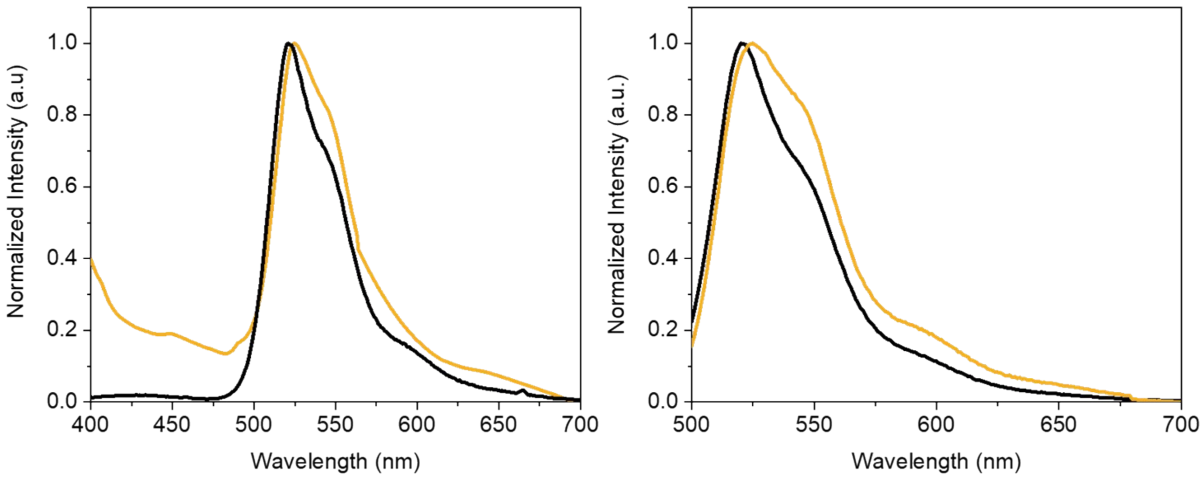

Figure S10. Emission spectra at excitation wavelengths of 375 nm (left) and 490 nm (right) of fresh (black) and after isothermal treatment (orange) FP-SF films. Please notice that the tail of the excitation lamp is noted (400 nm) in the left graph.

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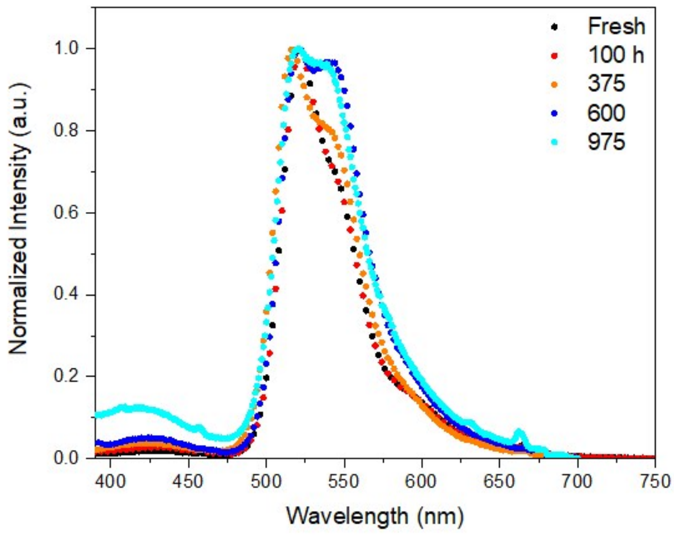

Figure S11. Emission spectra at excitation wavelengths of 375 nm of FP-SF films used in remote Bio-HLEDs.

Table S1. Summary of the state-of-the-art in Bio-HLEDs

Biogenic component	LED structure	Driving mode	x/y CIE color coordinates	CRI	CCT (K)	Lifetime (h)	Efficiency (lm/W)	Ref
			Biogenic matrix					
BSM	411 LED/BMS/RGB dyes	3.0 V	0.32/0.32	-	-	-	-	1
BSA	365 LED /BSA/C460/F/ROX/EDC	-	0.28/0.31	-	5300	106	-	2
Cellulose	450 LED/QDs-cellulose	3.0 V	0.32/0.32	-	-	-	-	3
Starch	450 LED/starch/g-CDs	2.8 V	0.26/0.33	-	-	-	-	4
Starch	450 LED/starch/g-CDs	2.8 V	0.23/0.27	-	-	-	-	4
Cellulose	390 LED/afGQDs	4.1 V	0.33/0.37	-	-	100	-	5
Cellulose nanofibre	410 LED/fGQDs@CNF–clay/yellow/orange	20 mA	0.33/0.37	-	-	-	36.1	5
Cellulose nanofiber	410 LED/fGQDs@CNF–clay/green/orange	20 mA	0.30/0.42	-	-	-	35.4	5
Nanocellulose	UV-LED/crystalline nanocellulose/porcine gastric mucin/RG dyes	-	0.40/0.42	84.4	3543 - 4150	-	-	6
DNA	UV-LED/DNA-curcumin	10 mA	0.39/0.56	-	-	-	1.6	7
Bio-inspired Jellyfish-like	SMD LED Lamp/PAN-NFs/PDMS	2.5 V	0.34/0.35	-	-	-	29.7	8
Cassava	UV-LED/Coumarin/curcumin/sulforhodamine/cassava-based biopolymerfilm	10 mA	0.33/0.32	-	-	-	-	9
			Biogenic emitter					

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FP	440 LED/ PEO/eGFP/mCherry	10 mA	0.32/0.33	80	4500-6000	>100	55	10
FP	395 LED/ PEO/BFP/GFP/mCherry	65 mA	0.35/0.35	70-60	4500-6000	-	3.4	10
FP	450 LED/microstructured PEO/eGFP/mCherry	20 mA	0.33/0.33	-	5500	>60	6	11
FP	450 LED/eGFP/mCherry	-	-		8440	-	248	12
FP	Blue LED/eGFP/mCherry	-	-	-	-	-	-	13
FP	450 LED/eGFP (on chip)	200 mA	-	-	-	1 min	-	14
FP	450 LED/eGFP (on chip)	30 mA	-	-	-	100	-	14
FP	450 LED chip/eGFP (remote)	200 mA	-	-	-	>300	-	14
Fused FP	395 LED/trimer BFP@GFP@mCherry	10 mA	0.37/0.38	91	4300	>400	15	15
FP	440 LED chip/eGFP-AA	200 mA	0.30/0.65	-	-	2	130	16
Protein-Au NCs	380 LED blue/redAuNCs (prepared/measured in oxygen conditions)	30 mA	0.31/0.29	-	6840	10	3	17
Protein-Au NCs	380 LED/blue/redAuNCs (prepared in oxygen/measured in inert conditions)	30 mA	0.34/0.31	-	4803	221	3	17
Protein-Au NCs	380 LED/blue/redAuNCs (prepared/measured in inert conditions)	30 mA	0.32/0.33	-	6377	800	3	17
R-PE	405 LED/R-PE proteins/QD@ZIF-8 film	-	0.34/0.34	85	4955	-	-	18
R-PE	UV-LED/R-PE@HSBW1	14 mA/cm^2	0.33/0.34	85	5740	-	81	19

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