Electronic Supplementary Material (ESI) for Materials Horizons.

Supporting information

1. Preparation and characterization of the gel- and rubber-like materials

All the luminescent compounds, such as small-molecules, polymers, and coordination complexes were purchased from Merck and Sigma Aldrich and used as received. The carbon quantum dots were prepared according to literature.[1] The gels were prepared as follows. As a first step, the branched and linear poly(ethylene oxide) compounds – *i.e.*, trimethylolpropane ethoxylate (TMPE) with Mn. of 450 mol. wt. and linear poly(ethylene oxide) (l-PEO) with Mn. of $5x10^6$ mol. wt. and 1 mg of the luminescent compounds were mixed with different amounts of solvent – *i.e.*, water or acetonitrile. Upon strong stirring (750 or 1500 rpm) under ambient conditions over night, this mixture becomes a gel. The mass ratio is optimized for the formation of a gel-like material that allows the further film forming *via* doctor-blading onto any kind of substrate like, for example, glass slides. The doctor-blading was performed using a rectangular stamp of a thickness of 50 µm that was placed onto the support. They can also be applied onto 3D substrates by introducing them into the gels. Subsequently, the films or coated materials were introduced into a vacuum station under 1-10 mbar for less than 1 h. The final materials are best described as rubbers, which are easily peeled off from the substrate with a tweezer and can be transferred to another substrate. The thickness of the rubbers can be controlled either by the thickness of the stamp or by the subsequent deposition of one layer on top of another with an excellent adhesion. The thickness and roughness were measured using a profilometer Dektak XT from Bruker. The presence of the luminescent materials was corroborated by spectroscopic techniques – steady-state absorption and photoluminescence characterizations, as well as excited-state lifetimes and photoluminescence quantum yields that were performed by using Perkin Elmer Lambda, Fluoromax-P-spectrometer (Horiba-JobinYvon), and SPEX Fluorolog-3 (Horiba-JobinYvon) supplied with an integrated TCSPC software. The refraction index was measured by using Krüss refractometer equipment from A Kross Optronic.

The rheological measurements of the gels and of the bare rubbers were carried out with an MCR 301 rheometer from Anton Paar at a temperature of 295.16 K. The gels were studied using a cone-and-plate geometry with a diameter of 25 mm and a cone angle of 1°. The oscillatory measurements of the rubbers were performed with a parallel-disk configuration with a plate diameter of 25 mm and a gap width of 1 mm. Amplitude sweeps were carried out at an angular frequency of 1 rad/s in a deformation ranged between 0.1% and 2% to determine the linear viscoelastic regime of the materials studied. Frequency sweeps were carried out in the linear viscoelastic regime at angular frequencies ranging from 0.1 to 100 rad/s. The study of the impact of luminescent materials on the rheological properties of the rubbers was performed with a narrow-gap rheometer in the parallel-disk configuration at a temperature of 297.76 K. It is based on a UDS 200 rotational rheometer from Physica. As disks, it uses glass plates of 75 mm and 50 mm diameter with an evenness of $\lambda/4$ and $\lambda/10$, respectively. The gap width is set up and measured independently from the rheometer with a confocal interferometric sensor resulting in a gap width with a precision of up to ±0.7 µm. Further details about this setup and its alignment are provided in reference 2. The samples were squeezed at normal forces of about 5-9 N to a gap width of 200 µm.

2. Fabrication and characterization of the WHLEDs

The blue-LEDs were purchased from Luxeon (LXHL-PR03) and Winger (WEPRB3-S1). The preparation of the WHLEDs concerns a two-step procedure. Firstly, the gels are deposited onto the LED wetting the complete surface. Secondly, the coated LED is transferred to the vacuum chamber under 1-10 mbar for less than 1 h. This procedure is repeated to enhance the light down-conversion efficiency of the WHLED. As well, the design of a cascade coating is easily performed by repeating the above-described steps depositing subsequently high- and low-energy emitting gels. Independently of the thickness of the coating, it can be easily peeled off from the LED surface for a further analysis. The optimized thickness of the coatings is mentioned in the main text. The WHLEDs were characterized by using a Keithley 2400 as a current source, while the luminous efficiency and changes of the electroluminescence spectrum were monitored by using Avantes spectrophotometer (Avaspec-ULS2048L-USB2) in conjunction with a sphere Avasphere 30-Irrad.

Table S1. Test of the formation of the gel and rubber materials by changing different parameters like the nature of the solvents, the amount of the solvents, the b-PEO:l-PEO mass ratio, and the stirring conditions.

	Solvent	Volume [µL]	b-PEO/l-PEO [wt.]	Stirring [rpm]	Gel formation	Rubber formation
polar protic	Water	50	6:1/12:1	750/1500	Highly viscous	Yes
		150	6:1/12:1	750/1500	Good viscosity	Yes
	Ethanol	50	6:1/12:1	750/1500	Immiscible	No
		150	6:1/12:1	750/1500	Immiscible	No
	Isopropanol	50	6:1/12:1	750/1500	Immiscible	No
		150	6:1/12:1	750/1500	Immiscible	No
polar aprotic	Acetonitrile	50	6:1/12:1	750/1500	Low viscosity	No
		150	6:1/12:1	750/1500	Good viscosity	Yes
	Cyclohexanone	50	6:1/12:1	750/1500	Immiscible	No
		150	6:1/12:1	750/1500	Immiscible	No
	THF	50	6:1/12:1	750/1500	Immiscible	No
		150	6:1/12:1	750/1500	Immiscible	No
a	Toluene	50	6:1/12:1	750/1500	Immiscible	No

	150	6:1/12:1	750/1500	Immiscible	No
Hexane	50	6:1/12:1	750/1500	Immiscible	No
	150	6:1/12:1	750/1500	Immiscible	No
Chloroform	50	6:1/12:1	750/1500	Immiscible	No
	150	6:1/12:1	750/1500	Immiscible	No

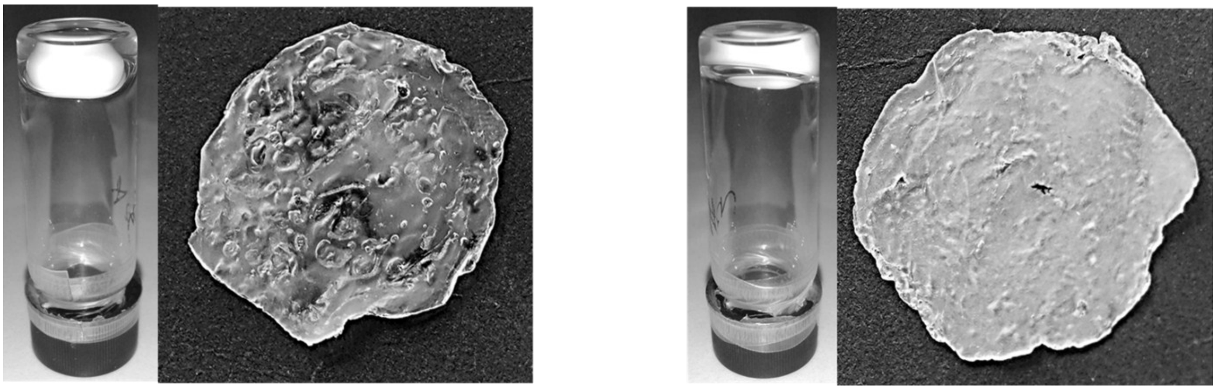


Figure S1. Pictures of the gels (notice the magnetic stirrer) and rubbers (diameter ~2.5 cm) prepared with water (left) and acetonitrile (right) with a mixture of b-PEO:l-PEO of 12:1 wt.

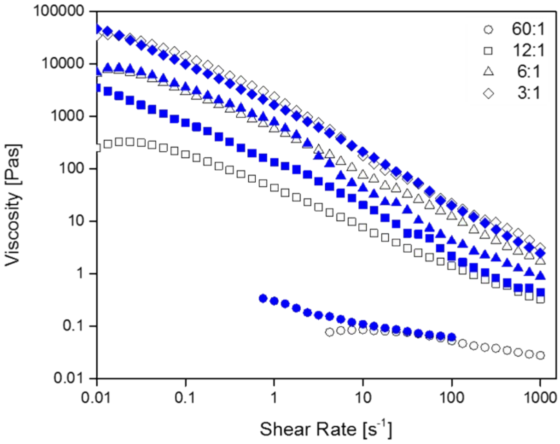


Figure S2. Viscosity functions of water-based (open symbols) and acetonitrile-based (solid symbols) gels with different mass ratios of b-PEO:l-PEO.

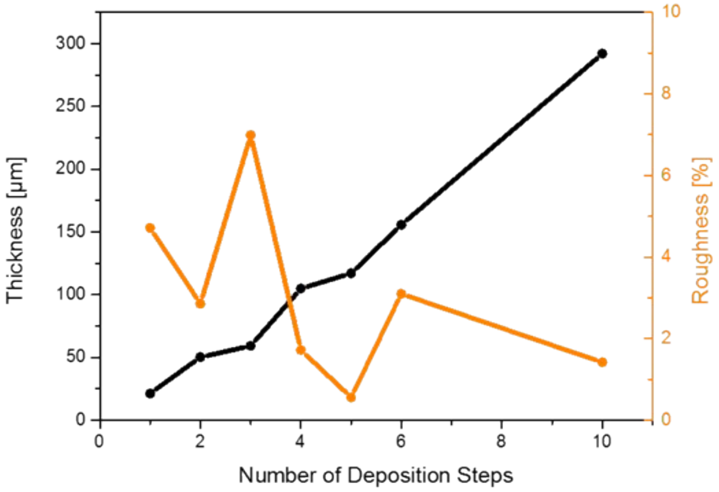


Figure S3. Changes of the thickness and roughness values of the acetonitrile-based rubbers upon repetitive deposition steps.

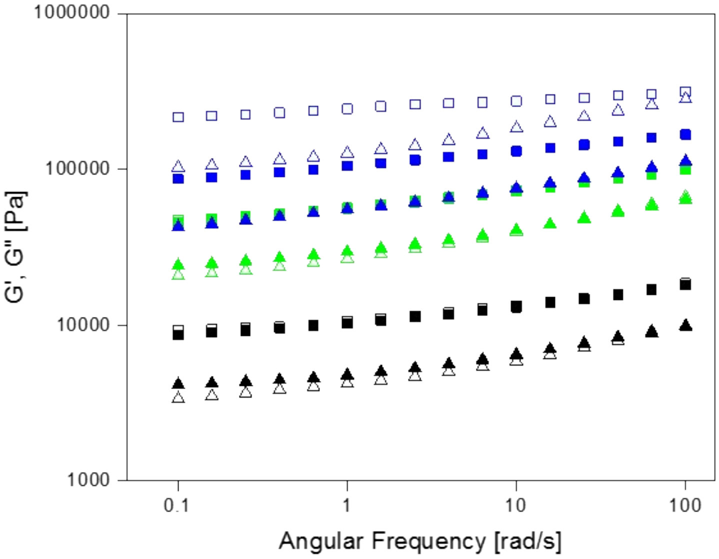
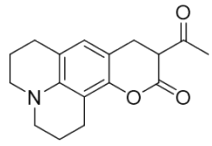


Figure S4. Storage G’ (square) and loss G” (triangles) moduli as function of angular frequency for water-based (open symbols) and acetonitrile-based (solid symbols) rubbers at different mass ratios of b-PEO:l-PEO: 12:1 (black symbols), 6:1 (green symbols), and 3:1 (blue symbols).

Small Molecules



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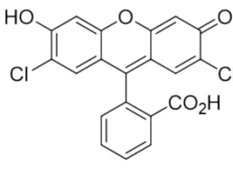

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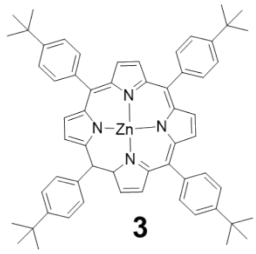

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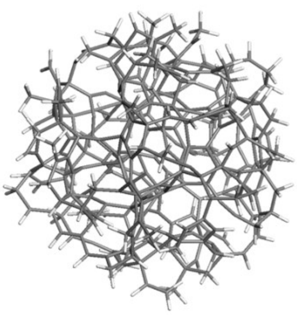
Carbon Nanodots



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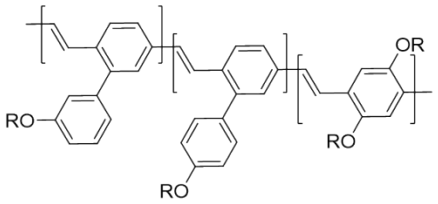
Polymers



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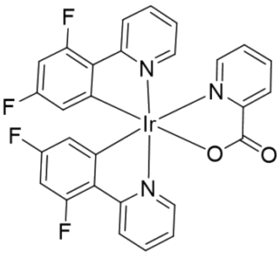
Complexes



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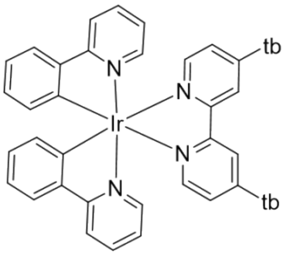

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Figure S5. Chemical structures of the luminescent materials, such as small-molecules (**1**-**3**), graphitic quantum dots (**4**), polymers (**5**), and coordination complexes (**6** and **7**).

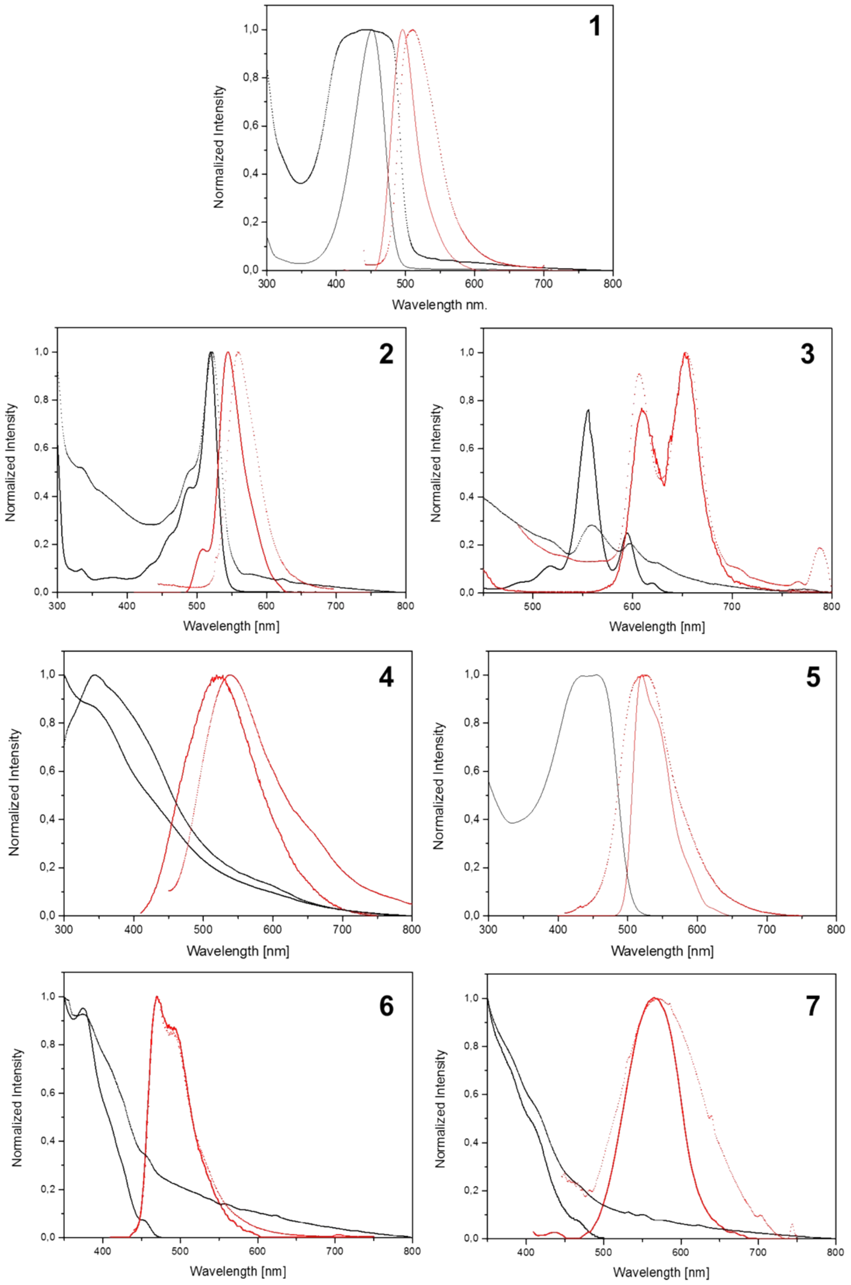

Figure S6. Absorption (black) and emission (red) spectra of the luminescent compounds in solution (solid line) and rubbers (close symbols).

Table S2. Photophysical properties of **1-7** in solution and rubbers.

Compound	PLQY$_{sol/rubber}$ [%]	Lifetimes$_{sol/rubber}$ [ns]	
		τ_1	τ_2
1	68/30	0.70/0.48	3.11/1.11
2	87/36	0.97/0.24	-
3	3.3/1.1	1.28/0.32	1.95/1.05
4	20/5	2.60/1.64	10.30/8.27
5	69/74	0.70/1.0	0.22/0.27
6	3.9/15.7	103/651	-
7	7/35	65/383	-

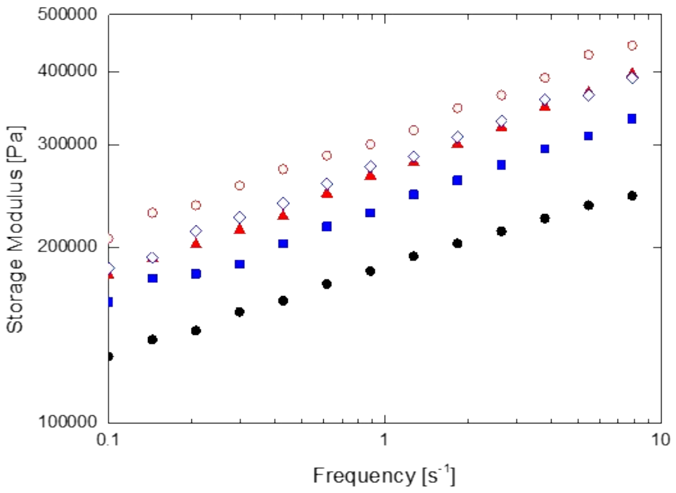

Figure S7. Frequency sweeps of the storage modulus for different rubbers prepared with b-PEO:l-PEO 6:1 wt. and **1** (blue diamond), **5** (red triangle), and **7** (red circle), compared to the references based on water (black circle) and acetonitrile (blue square).[3]

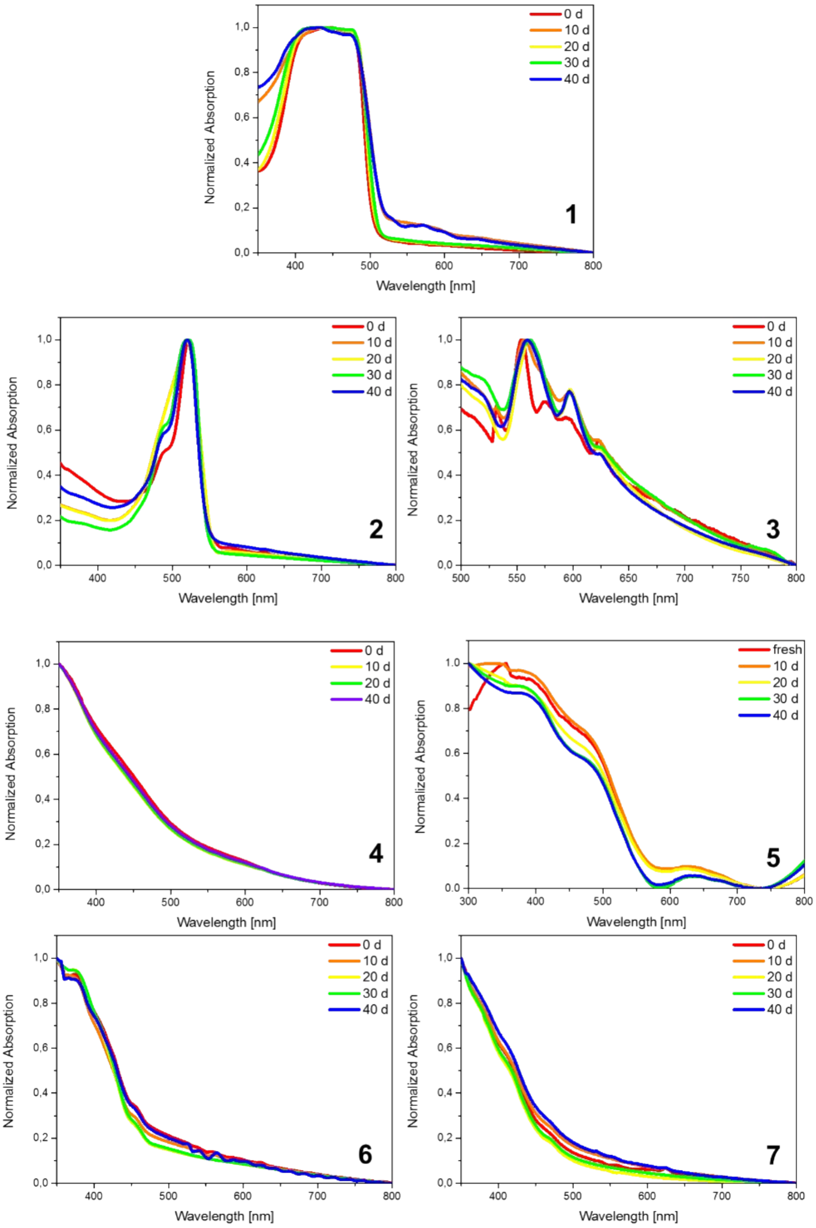

Figure S8. Changes in the absorption spectra of rubbers based on **1-7** over time under ambient storage conditions.

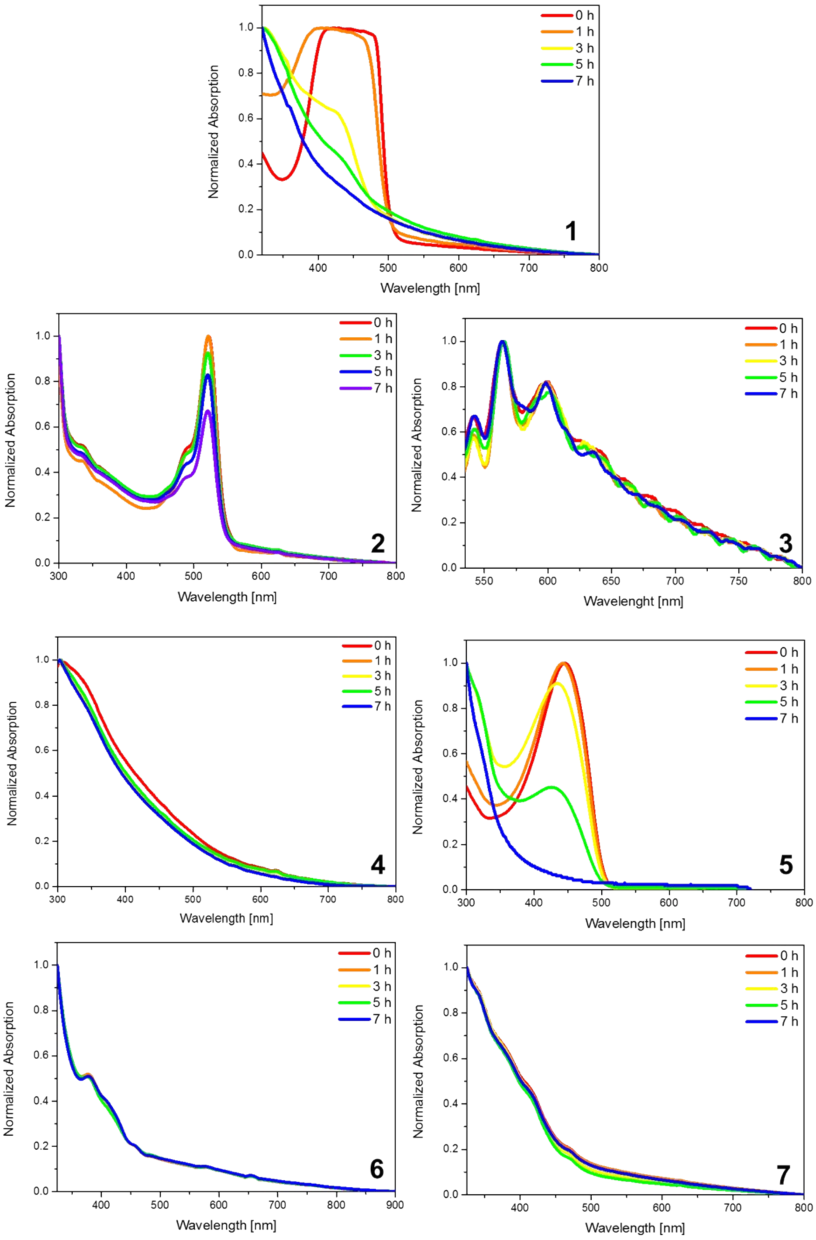

Figure S9. Changes in the absorption spectra of rubbers based on **1**-**7** over time upon UV irradiation (310 nm; 8 W) in ambient conditions.

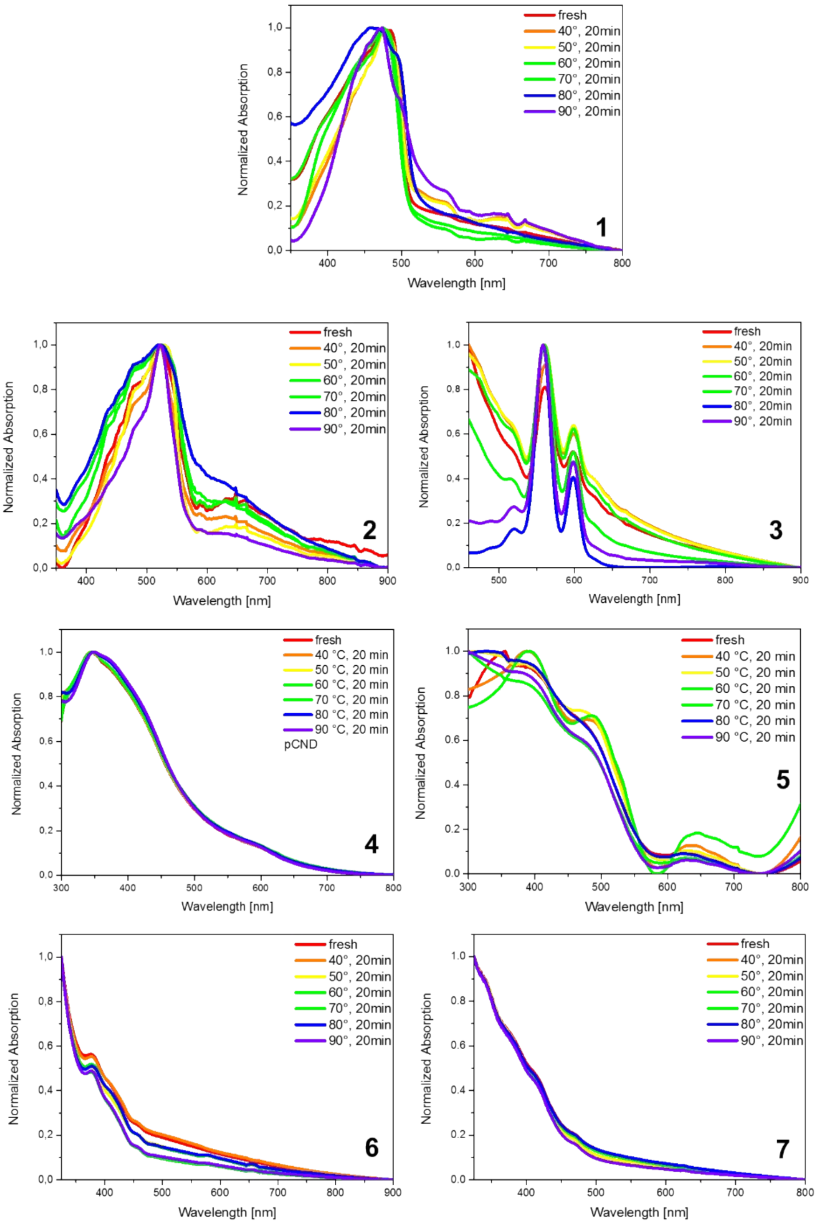

Figure S10. Changes in the absorption spectra of rubbers based on **1**- **7** upon heating in ambient conditions.

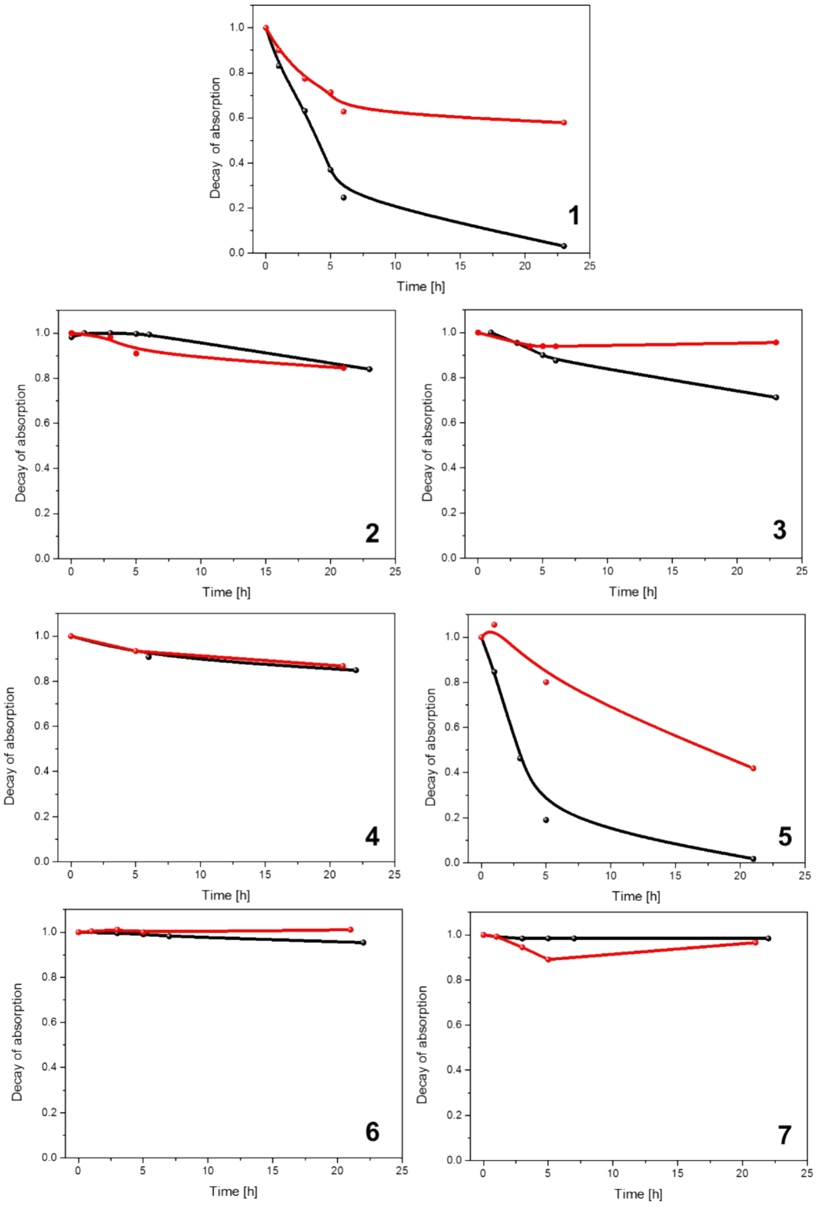

Figure S11. Comparison of the change in absorption of compounds 1-7 in solution (black) and in the rubber (red).

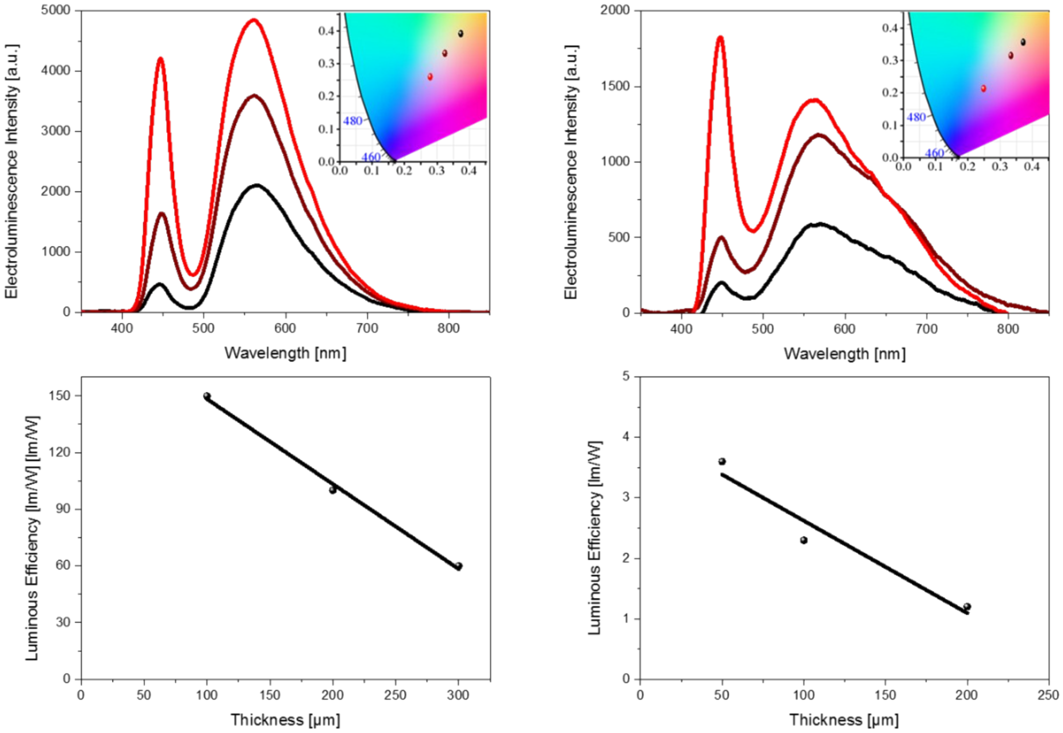

Figure S12. Upper part - Exemplary electroluminescence spectra of CC- (left) and QD-WHLEDs (right) with three different coating thicknesses – *i.e.*, thicker (black), optimum (brown), and thinner (red) that are related to values of 300/200/100 μm and 200/100/50 μm for CC- and QD-WHLEDs, respectively. The insets show the corresponding CIE coordinates. Bottom part - Changes of the luminous efficiency upon increasing the coating thickness.

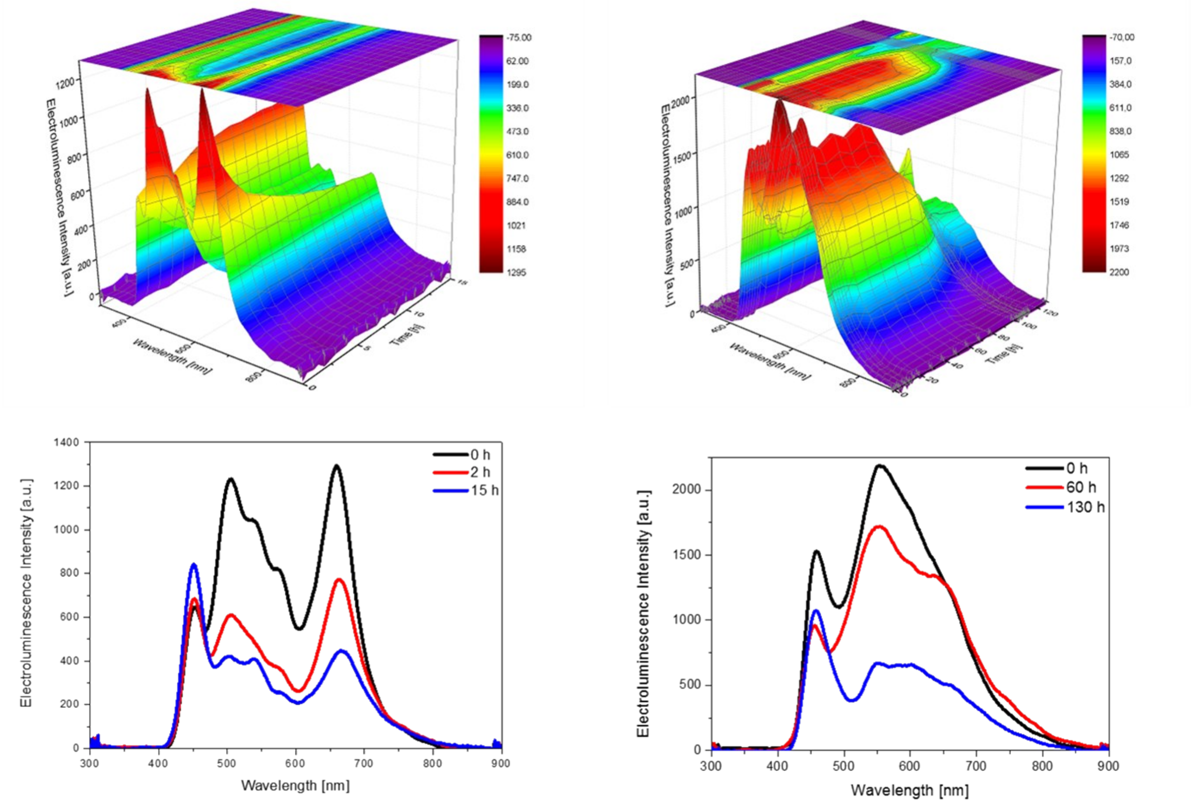

Figure S13. Changes in the electroluminescence spectrum of SM-WHLED blue-LED/**1**/**2**/**3** (left) and QD-WHLED blue-LED/**4** (right) over time.

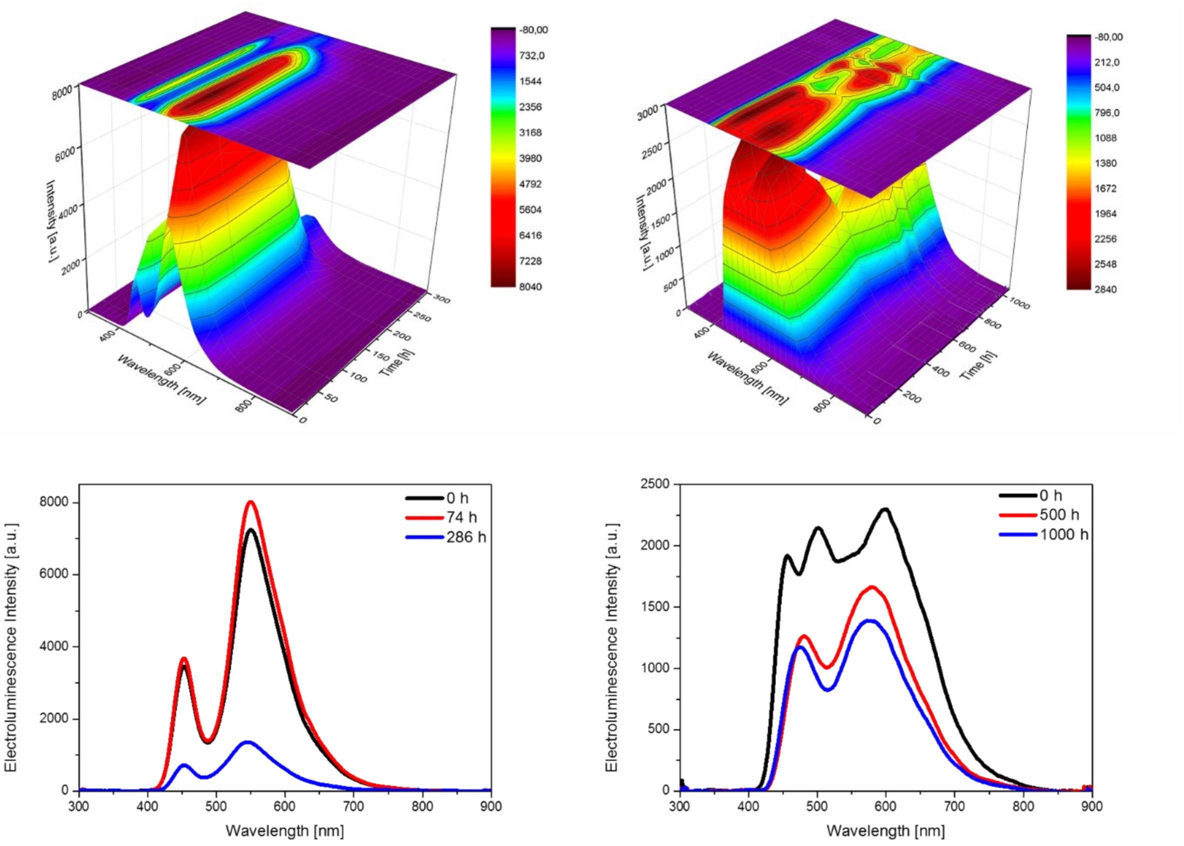

Figure S14. Changes in the electroluminescence spectrum of P-WHLED blue-LED/**5** (left) and CC-WHLED blue-LED/**6**/**7** (right) over time.

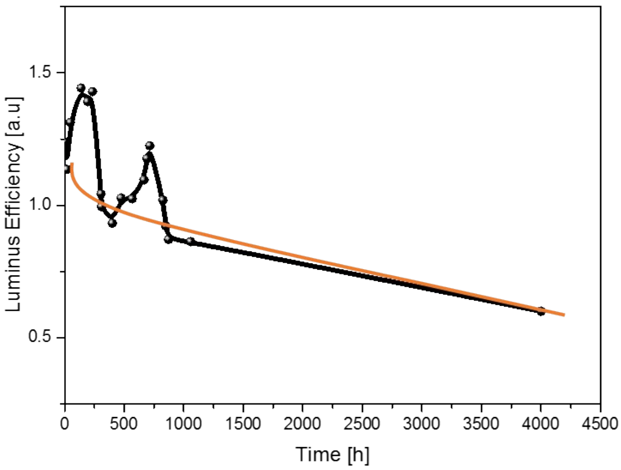

Figure S15. Extrapolated lifespan of CC-WHLEDs.

References

1 V. Strauss, J. T. Margraf, C. Dolle, B. Butz, T. J. Nacken, J. Walter, W. Bauer, W. Peukert, E. Spiecker, T. Clark, D. M. Guldi, *J. Am. Chem. Soc.* 2014, **136**, 17308.

2 H. Dakhil, A. Wierschem, *Appl. Rheol.* 2014, **24**, 63795.

3 Please notice that the differences are caused by variation between samples rather than by the presence of the dopants.