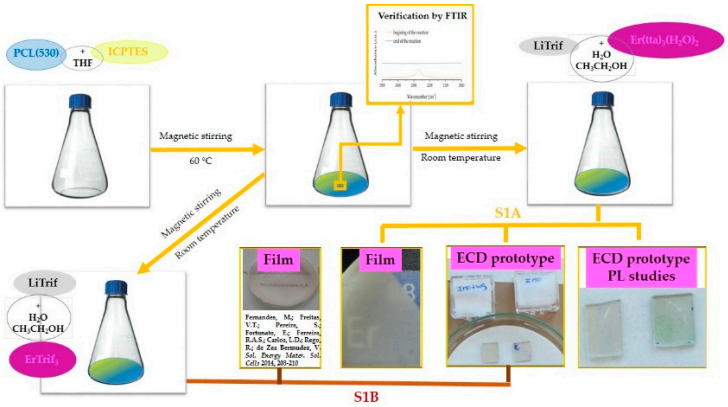
Supporting Information

Luminescent Electrochromic Devices for Smart Windows of Energy-Efficient Buildings

Mariana Fernandes [1,*], Vânia Freitas [2], Sónia Pereira [3], Rita Leones [4,5], Maria Manuela Silva [4], Luís D. Carlos [2], Elvira Fortunato [3], Rute A. S. Ferreira [2], Rosa Rego [1] and Verónica de Zea Bermudez [1,*]

[1] Department of Chemistry and CQ-VR, University of Trás-os-Montes e Alto Douro, 5000-801 Vila Real, Portugal; rrego@utad.pt (R.R.)
[2] Department of Physics and CICECO, University of Aveiro, 3810-193 Aveiro, Portugal; vania.freitas@ua.pt (V.F.); lcarlos@ua.pt (L.D.C.); rferreira@ua.pt (R.A.S.F.)
[3] CENIMAT/I3N, Departamento de Ciência dos Materiais, Faculdade de Ciências e Tecnologia, FCT, Universidade Nova de Lisboa and CEMOP-UNINOVA, 2829-516 Caparica, Portugal; sp@uninova.pt (S.P.); emf@fct.unl.pt (E.F.)
[4] Department of Chemistry, University of Minho, Gualtar, 4710-057 Braga, Portugal; r.d.barros.leones@ifw-dresden.d (R.L.); nini@quimica.uminho.pt (M.M.S.)
[5] Current address: Leibniz Institute for Solid State and Materials Research (IFW) Dresden e.V., Institute for Complex Materials, Helmholtzstr D-01069 Dresden, Germany
* Correspondence: mspf@utad.pt (M.F.); vbermude@utad.pt (V.d.Z.B.); Tel.: +259 350-000 (ext. 4923) (M.F.); Tel.: +351-259-350000 (ext. 4317) (V.d.Z.B.)

Scheme 1: Simplified representation of the work developed; Table S1: Experimental details of the preparation of the d-PCL(530)/siloxane$_{4.9}$LiTrif-ErTrif$_3$ and d-PCL(530)/siloxane$_{9.1}$LiTrif-[Er[(tta)$_3$(H$_2$O)$_2$] electrolytes; Figure S1: Image (a) and structure of the Er(tta)$_3$(H$_2$O)$_2$ complex (b) and the corresponding FT-IR spectrum (c); Figure S2: Absorption spectra of the Er(tta)$_3$(H$_2$O)$_2$ complex (red line) and HTTA (black line) in ethanolic solutions; Figure S3: SEM (a) and 2D AFM (b) images, XRD pattern (c), and UV-visible-NIR spectrum (d) of the IMO layer; Table S2: Electrical data of IMO layers; Table S3: Optical performance of the ECDs; Figure S4: Cyclic voltammograms of the ECD@LiTrif-ErTrif$_3$ (blue) and ECD@LiTrif-[Er(tta)$_3$(H$_2$O)$_2$] (red) devices at scan rates of 50 mV s^{-1} (solid line), 20 mV s^{-1} (dotted line), and 50 mV s^{-1} (dashed line); Figure S5: De-inserted (Q_{out}) (solid symbols) (a) and inserted ($-Q_{in}$) (open symbols) (b) charge density as a function of the number of cycles, for ECD@LiTrif-ErTrif$_3$ and ECD@-LiTrif[Er(tta)$_3$(H$_2$O)$_2$] and $Q_{out}/-Q_{in}$ ratio (semi-solid symbols) (c) for ECD@LiTrif-ErTrif$_3$ (blue symbols) and ECD@LiTrif-[Er(tta)$_3$(H$_2$O)$_2$] (c) (red symbols); Figure S6: CV curves of the ECD@LiTrif-ErTrif (blue line) and ECD@LiTrif-[Er(tta)$_3$(H$_2$O)$_2$] (red line) after the CA test (scan rate: 50 mV s^{-1}).



Scheme 1. Simplified representation of the work developed.

Table S1. Experimental details of the preparation of the d-PCL(530)/siloxane$_{4.9}$LiTrif-ErTrif$_3$ and d-PCL(530)/siloxane$_{9.1}$LiTrif-[Er(tta)$_3$(H$_2$O)$_2$] electrolytes.

Mass PCL(530) (g)	Mass LiTrif (g)	m	Mass ErTrif$_3$ (g)	m′	Mass Er(tta)$_3$(H$_2$O)$_2$ (mg)	m″	*n*	Reference
0.95	0.12	8.7	0.40	11.2			4.9	[21]
1.10	0.13	9.2			9.1	764	9.1	This work

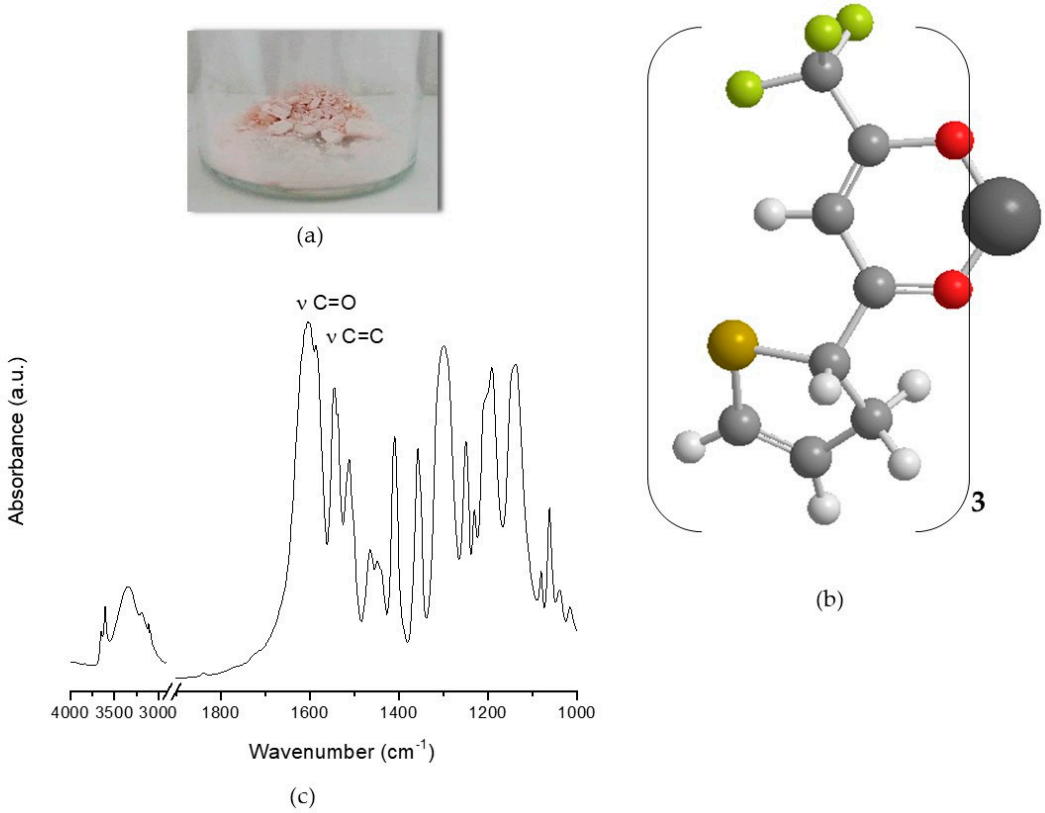


Figure S1. Image (**a**) and structure of the Er(tta)$_3$(H$_2$O)$_2$ complex (**b**) and the corresponding Fourier transform infrared (FT-IR) spectrum (**c**).

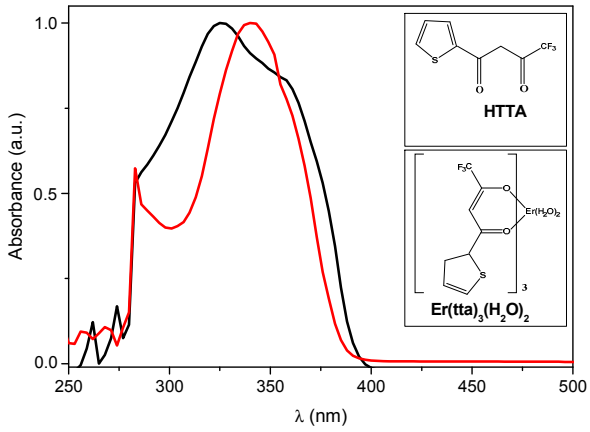


Figure S2. Absorption spectra of the Er(tta)$_3$(H$_2$O)$_2$ complex (red line) and HTTA (black line) in ethanolic solutions.

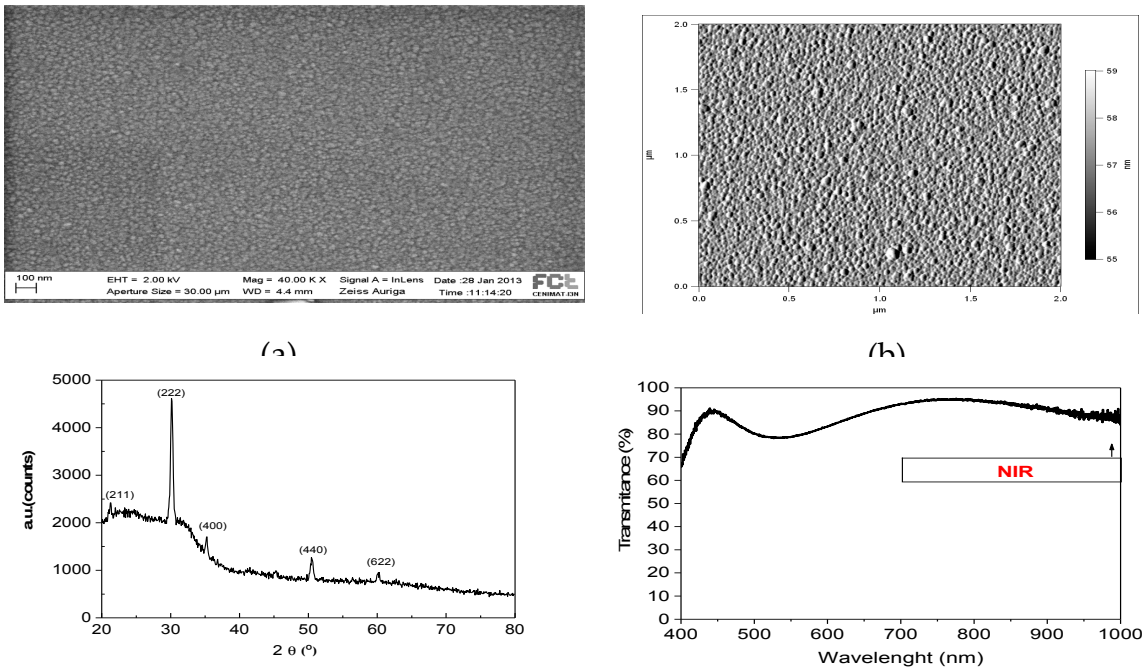


Figure S3. SEM (**a**) and 2D AFM (**b**) images, X-ray diffraction (XRD) pattern (**c**), and UV-visible-NIR spectrum (**d**) of the indium molybdenum oxide (IMO) layer.

Table S2. Electrical data of IMO layers.

Bulk resistivity (ρ) .cm)	Band gap (Eg) (eV)	Mobility ( ) (cm ^{2}V^{-1} s^{-1})	Carrier concentration (N)	Partial pressure Argon (Pa)	Partial pressure Oxygen (Pa)	Reference
7.87×10^{-3}	3.8	11.5	6.87×10^{19}		2.0×10^{-3}	This work
	3.69		2.08×10^{21}		1.0×10^{-3}	
4.05×10^{-3}	3.88				2.0×10^{-3}	
1.71×10^{-3}		~18.1		0.3	3.0×10^{-3}	[19]
		~19.1			3.6×10^{-3}	
			~1.11×10^{20}		4.0×10^{-3}	
1.80		1.4	2.53×10^{18}		6.0×10^{-3}	

Table S3. Optical performance of the electrochromic devices (ECDs).

	ECD@LiTrif-ErTrif$_3$		**ECD@LiTrif-[Er(tta)$_3$(H$_2$O)$_2$]**	
	T $_{555\ nm}$ (%)	**T $_{visible}$ (%)**	**T $_{555\ nm}$ (%)**	**T $_{visible}$ (%)**
As-deposited (not shown)	51.0	51.3	64.1	66.6
Colored	41.2	37.8	55.2	56.7
Bleached	46.3	41.4	64.1	66.1
ΔT	5.1	3.6	8.9	9.4
ΔOD	0.05	0.04	0.07	0.07

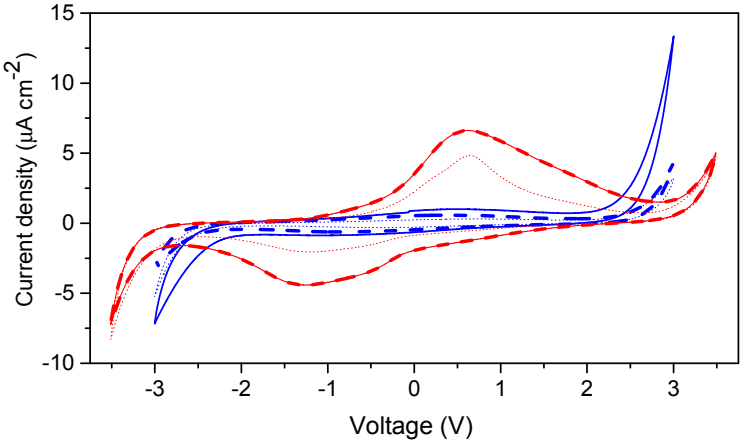


Figure S4. Cyclic voltammograms of the ECD@LiTrif-ErTrif$_3$ (blue) and ECD@LiTrif-[Er(tta)$_3$(H$_2$O)$_2$] (red) devices at scan rates of 50 mV s^{-1} (solid line), 20 mV s^{-1} (dotted line), and 50 mV s^{-1} (dashed line).

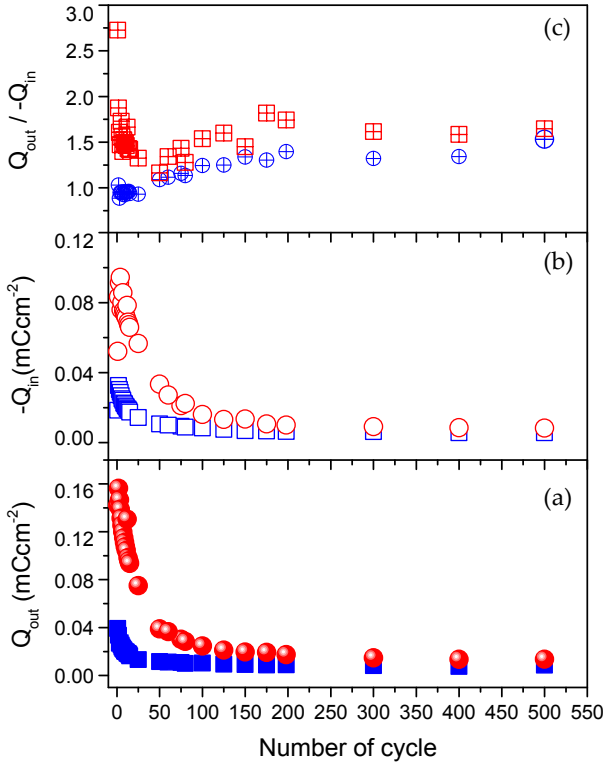


Figure S5. De-inserted (Q_{out}) (solid symbols) (**a**) and inserted ($-Q_{in}$) (open symbols) (**b**) charge density as a function of the number of cycles for ECD@-LiTrif-ErTrif$_3$ and ECD@-LiTrif-[Er(tta)$_3$(H$_2$O)$_2$] and the $Q_{out}/-Q_{in}$ ratio (semi-solid symbols) (**c**) for ECD@-LiTr-ErTrif$_3$ (blue symbols) and ECD@LiTrif-[Er(tta)$_3$(H$_2$O)$_2$] (c) (red symbols).

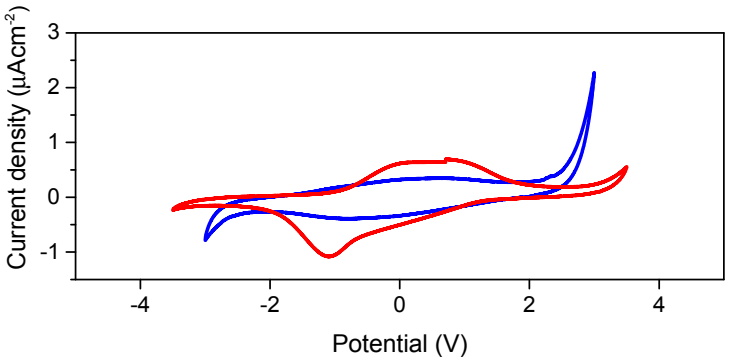


Figure S6. Cyclic voltammetry (CV) curves of the ECD@LiTrif-ErTrif (blue line) and ECD@LiTrif-[$Er(tta)_3(H_2O)_2$] (red line) after the chronoamperometric (CA) test (scan rate: 50 mV s^{-1}).