Supplementary material (ESI) for Journal of Materials Chemistry A This journal is © The Royal Society of Chemistry 2012

Ionic liquid redox electrolytes based on binary mixtures of 1-alkylmethylimidazolium tricyanomethanide with 1-methyl-3propylimidazolium iodide and implication in dye-sensitized solar cells

Maria Bidikoudi^a, Thomas Stergiopoulos^{*,a}, Vlassis Likodimos^a, Georgios Em. Romanos^a, Maria Francisco^b, Boyan Iliev^c, Gabriela Adamová^c, Thomas J. S. Schubert^c and Polycarpos Falaras^{*,a}

 ^a Institute of Advanced Materials, Physicochemical Processes, Nanotechnology and Microsystems (IAMPPNM), Division of Physical Chemistry, National Center for Scientific Research
"Damekrites", 152-10 A chia Paragkavi Attikia, Athana, Crassa, E. mail: pari@aham.damekrites.gr

"Demokritos", 153 10 Aghia Paraskevi Attikis, Athens, Greece. E-mail: papi@chem.demokritos.gr; +30-2106503644; +20-2106511766.

^b Dept. Chemical Engineering and Chemistry, Eindhoven University of Technology, Den Dolech 2, 5612 AZ, P.O. Box 513, STO 1.21, 5600 MB Eindhoven, The Netherlands.

^c IOLITEC, Ionic Liquids Technologies GmbH, Salzstraße 184, D-74076 Heilbronn, Germany.

Corresponding authors: Thomas Stergiopoulos (<u>stergt@chem.demokritos.gr</u>) and Polycarpos Falaras (papi@chem.demokritos.gr).

Table S1: Experime	ntal values	for viscosities	and densities	as a function	of temperature for ionic
liquid blend C2					

T(K)	η (mPas)	kvisc(mm ² s)	$\rho(g/cm^3)$
293.15	172.910	123.860	1.3961
298.15	125.700	90.298	1.3921
303.15	93.884	67.635	1.3881
308.15	71.817	51.887	1.3841
313.15	56.198	40.719	1.3801
318.15	44.811	32.562	1.3762
323.15	36.356	26.495	1.3722
328.15	29.956	21.894	1.3683
333.15	25.024	18.342	1.3643
338.15	21.183	15.571	1.3604
343.15	18.126	13.362	1.3565
348.14	15.664	11.580	1.3527
353.14	13.654	10.123	1.3488
358.14	12.000	8.922	1.3450
363.14	10.629	7.925	1.3412

Table	S2 :	Experimental	values	for	viscosities	and	densities	as a	a function	of t	emperature	for	ionic
liquid l	blen	d C4											

T(K)	η (mPas)	kvisc(mm ² s)	$\rho(g/cm^3)$
293.15	272.21	196.36	1.3863
298.15	192.8	139.48	1.3823
303.15	140.56	101.98	1.3783
308.15	105.28	76.604	1.3744
313.15	80.485	58.727	1.3705
318.15	62.897	46.025	1.3666
323.15	50.074	36.746	1.3627
328.15	40.561	29.85	1.3588
333.15	33.348	24.613	1.3549
338.14	27.825	20.0595	1.351
343.15	23.498	17.442	1.3472
348.14	20.067	14.938	1.3434
353.14	17.284	12.903	1.3396
358.14	15.022	11.245	1.3358
363.15	13.148	9.8706	1.3321

Supplementary material (ESI) for Journal of Materials Chemistry A This journal is © The Royal Society of Chemistry 2012

Table S3: Experimental values for viscosities and densities as a function of temperature for ionic liquid blend C6

T(K)	η (mPas)	kvisc(mm ² s)	$\rho(g/cm^3)$
293.15	364.63	264.7	1.3776
298.15	253.74	184.74	1.3734
303.15	181.88	132.81	1.3695
308.15	134.08	98.187	1.3656
313.15	101.22	74.331	1.3617
318.15	78.069	57.493	1.3579
323.15	61.398	45.345	1.354
328.15	49.3128	36.387	1.3502
333.15	39.944	29.669	1.3463
338.15	32.927	24.526	1.3425
343.15	27.519	20.556	1.3387
348.14	23.27	1.3387	1.3349
353.14	19.877	14.932	1.3312
358.14	17.142	12.914	1.3274
363.15	14.913	11.266	1.3237

Supplementary material (ESI) for Journal of Materials Chemistry A This journal is © The Royal Society of Chemistry 2012

Table S4: Experimental values for viscosities and densities as a function of temperature for ionic liquid blend C8

T(K)	η(mPas)	kvisc(mm ² s)	$\rho(g/cm^3)$
293.15	419.49	305.57	1.3728
298.15	292.57	213.76	1.3687
303.15	209.04	153.17	1.3648
308.15	153.33	112.68	1.3609
313.15	115.15	84.857	1.357
318.15	88.351	65.292	1.3532
323.15	69.11	51.21	1.3493
328.15	55.033	40.901	1.3455
333.15	44.531	33.19	1.3417
338.15	36.558	27.325	1.3379
343.15	30.394	22.785	1.3341
348.15	25.581	19.229	1.3303
353.15	21.76	16.403	1.3266
358.15	18.694	14.132	1.3229
363.15	16.208	12.286	1.3192



Figure S1. Raman spectrum of the PMII/ $[C_6mim][TCM]$ blend compared to those of pure IL components. The dashed curve corresponds to the calculated mole fraction weighted sums of the neat PMII and $[C_6mim][TCM]$ components.



Figure S2. Raman spectrum of the PMII/ $[C_8mim]$ [TCM] blend compared to those of pure IL components. The dashed curve corresponds to the calculated mole fraction weighted sums of the neat PMII and $[C_6mim]$ [TCM] components.



Figure S3. Raman spectrum of the 4-tert-butyl pyridine at 785 nm.



Figure S4. Stokes–Einstein ratios for ionic liquid 1-alkyl-3-methylimidazolium tricyanomethanide (C_v mimTCM)-based blends at room temperature as a function of the increasing alkyl length (and thus the viscosity) of the C_v mimTCM. A linear trend line is shown as a guide to the eye.

Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A This journal is © The Royal Society of Chemistry 2013



Figure S5. Specific conductivity (κ) as a function of the viscosity (by increasing the alkyl length of C_v) of the C_v mimTCM-based blends at room temperature. A linear trend line is shown as a guide to the eye.

Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A This journal is C The Royal Society of Chemistry 2013



Figure S6. Short-circuit photocurrent (J_{sc}) plotted against the light power density (P_{in}) for the different ionic liquid electrolytes. The black dotted line represents the ideal linear behavior that should have been observed for C2-electrolyte.

Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A This journal is C The Royal Society of Chemistry 2013



Figure S7. Electron transit times (τ) calculated from IMPS versus J_{sc} (in log-log scale) delivered by the DSCs, incorporating the different ionic liquid electrolytes.

Supplementary material (ESI) for Journal of Materials Chemistry A This journal is © The Royal Society of Chemistry 2012



Figure S8. Characteristic macroscopical images of liquid and solidified electrolytes (the reference ionic-liquid based electrolyte is depicted on the left while the one solidified with silica nanopowder is shown on the right).

Supplementary material (ESI) for Journal of Materials Chemistry A This journal is © The Royal Society of Chemistry 2012



Figure S9. (a) Chemical capacitance (C_{μ}) plotted against V_F and (b) recombination resistance (R_{rec}) plotted against V_{ecb} for various solidified electrolytes.

Supplementary material (ESI) for Journal of Materials Chemistry A This journal is © The Royal Society of Chemistry 2012



Figure S10. Resistance at the counter electrode/electrolyte interface (R_{pt}) plotted against the applied potential $(V_{applied})$ for electrolytes C2 (a) and C8 (b) prior and after the addition of SiO₂ nanoparticles.