Electronic Supplementary Information

Bio-based Chitosan and Cellulose Ionic Liquid Gels: Polymeric Soft Materials for the Desulfurization of Fuel

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Figure S1. DSC thermograms of ILGs, exo up.



Figure S2. Rheological measurements of ILGs.



Figure S3. POM images of a) CS/[bmpyrr][OAc] and b) CS+CE/[bmim][OAc] at 10x.



Figure S4. Picture of self-healing test performed on CEOAc/[bmim][PF₆]; **a)** uncolored and dyed gel cut, **b)** diffusion of Rhodamine B to the uncolored gel, **c)** complete joint of the two pieces of gel in 24 h.



Figure S5. Picture of self-healing test performed on CS+CE/[bmpyrr][OAc]; **a)** uncolored and dyed gel cut, **b)** initial contact between gel and dyed one, **c)** diffusion of Rhodamine B to the uncolored gel, **d)** homogeneous diffusion of dye in all gel and complete joint of the two pieces of gel in 24 h.



Figure S6. Picture of load-bearing test performed on CS+CE/[bmim][OAc] **a**) gel before casting weights, **b**) gel with 2 dkg weight cast on it, **c**) gel partially deformed after loading 20 dkg. **d**) CS+CE/[bmpyrr][OAc] gel with 2 dkg weight cast on it, **e**) intact CS+CE/[bmpyrr][OAc] gel after loading 20 dkg, **f**) flowing CEOAc/[bmim][PF₆] gel after reaching the maximum weight on it.



Figure S7. NMR spectra of hexane cast for 48 h on ILGs adding DMSO-d₆ as internal standard.



Figure S8. Trend of AE, corresponding to each sulfur compound, from ternary mixtures as function of total concentration of sulfur in the mixture. Tests at 24h, room temperature and with 0.5 mL of solution. AE is based on triplicate runs with a reproducibility of 3%.



Figure S9. Trend of AE, corresponding to each sulfur compound, from ternary mixtures as function of volume cast on gels. Tests at 24h, room temperature and with 1500 ppm of total sulfur concentration. AE is based on triplicate runs with a reproducibility of 3%.



Figure S10. Trend of AE, corresponding to each sulfur compound, from ternary mixtures as function of gel diameter. Tests at 24h, room temperature and with 1500 ppm of total sulfur concentration. AE is based on triplicate runs with a reproducibility of 3%.



DBT BT T

Figure S11. Trend of AE, corresponding to each sulfur compound, from ternary mixtures in static or dynamic conditions (400 rpm). Tests at 24h, room temperature and with 1500 ppm of total sulfur concentration. AE is based on triplicate runs with a reproducibility of 3%.



Figure S12. Trend of AE, corresponding to each sulfur compound, from ternary mixtures for a series of three syringes. On the left, ternary mixture and binary mixture on the right. Tests after 15 min of contact, room temperature and with 1500 ppm of total sulfur concentration. AE is based on triplicate runs with a reproducibility of 3%.



Figure S13. Trend of AE, corresponding to each sulfur compound, from ternary mixtures on the same gel to test recycling ability. Tests at 24 h, room temperature and with 1500 ppm of total sulfur concentration. AE is based on triplicate runs with a reproducibility of 3%.



Figure S14. Rheological measurements performed on regenerated ILGs.

	AE (%) of DBT	AE (%) of BT	AE (%) of T
[bmim][OAc]	48	33	69
CS+CE/[bmim][OAc]	34	34	64
CS/[bmim][OAc]	19	18	73
[bmpyrr][OAc]	20	23	28
CS+CE/[bmpyrr][OAc]	30	20	22
CS/[bmpyrr][OAc]	20	17	54
[bmim][Cl]	8	12	8
CS/[bmim][Cl]	13	13	26
CEOAc/[bmim][Cl]	13	12	21
[bmim][PF6]	18	30	34
CEOAc/[bmim][PF6]	6	55	37

Table S1. Trend of AE as function of ILs or ILGs for adsorption of sulfur compounds from hexane solutions, at initial concentration of 500 ppm. AE is based on triplicate runs with a reproducibility of 3%.

Table S2. Percentage of sulfur compounds recovered after ILGs washing with 2-Me-THF. different aliquots of gels.

Gel	V2-Me-THF (mL)	Recovered T (%)	Recovered BT(%)	Recovered DBT(%)
CEOAc/[bmim][PF6]	3 x 0.5 mL	100	100	36
CS+CE/[bmpyrr][OAc]	4 x 0.5 mL	98	100	25
CS+CE/[bmim][OAc]	4 x 0.5 mL	61	100	74

Table S3. Rheological parameters of ILGs determined at 25 °C, $\gamma = 1$ % and f = 1 Hz, after washing with 2-Me-THF. Values determined as average values on three different aliquots of gels.

Gel	<i>G</i> ' (Pa)	G" (Pa)	tan δ	γ at crossover point
				(%)
CS+CE/[bmim][OAc] CS+CE/[bmpyrr][OAc]	(2300±500) (2000±1000)	(1000±100) (400±200)	(0.37±0.12) (0.20±0.01)	(23±3) (77.6±0.01)