

Supporting Information

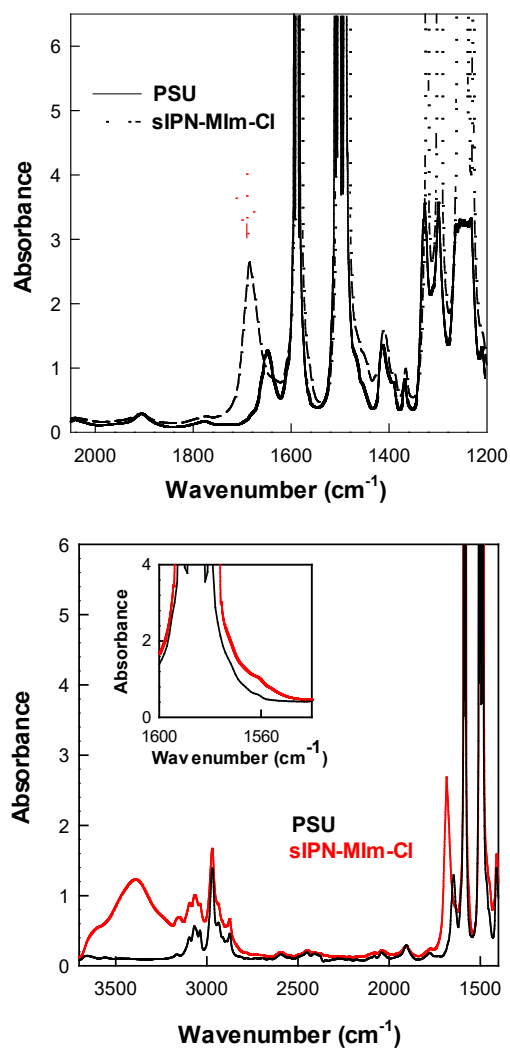


Figure S1. FTIR spectra of PSU and sIPN-MIm-Cl membranes in different regions of the spectrum.

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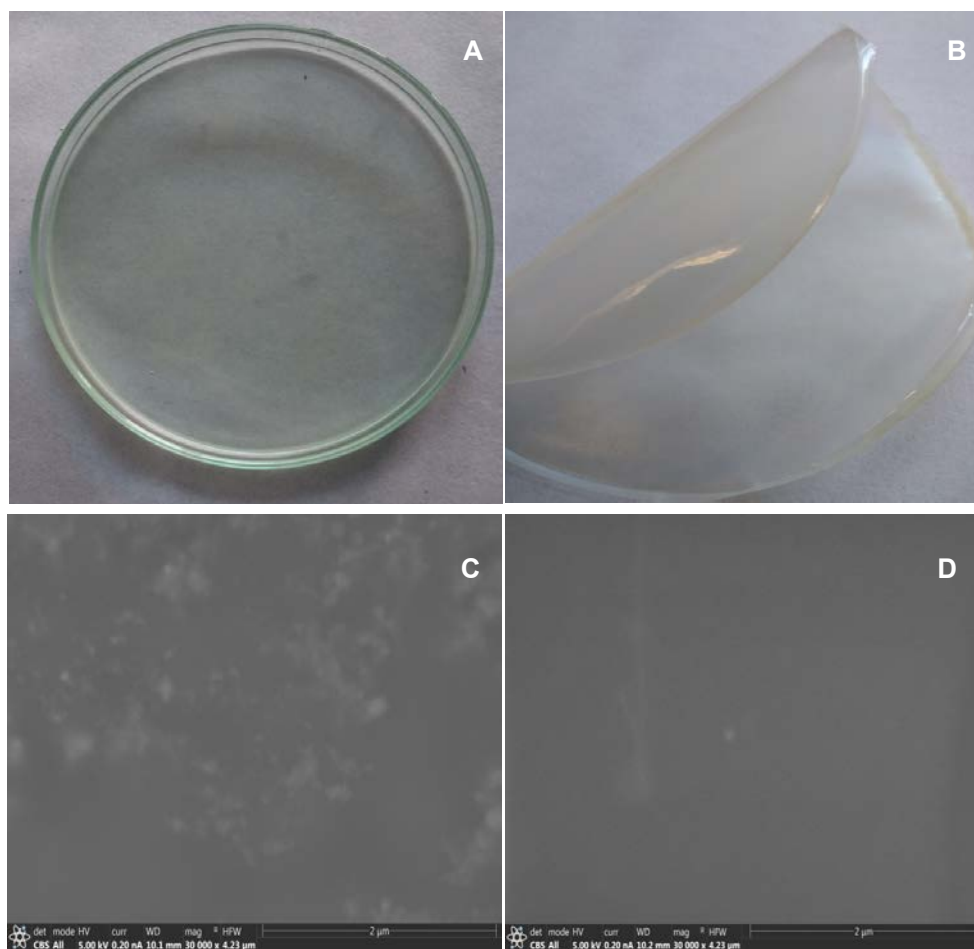


Figure S2. sIPN membranes (A, B), SEM images of the surface of sIPN-TMA-Cl membrane (C) and sIPN-MIm-Cl membrane (D).

Physically the membranes are transparent and flexible (Fig. S2A and B). It is directly related to their good mechanical robustness. SEM is employed to evaluate the morphology of the membranes in a microstructural level. Homogeneity of the membrane is essential to obtain good ionic conductivity. In this context, as can be seen in the images, the surface of the membranes sIPN-TMA-Cl and sIPN-MIm-Cl (Fig. S2C and D) is dense and without pores. Both types of membranes show a compact, uniform and homogeneous

morphology in the large area observed. Phase separation is not observed even at a high magnification. Generally, crosslinking systems exhibit higher homogeneity. The lack of interaction between polymers creates phase separation and membranes without uniformity.^[1,2] In this case, the difference between the composition of the crosslinked and non-crosslinked polymer is negligible and, just in the Fig. S2C, two zones with different contrast are appreciable. The lighter zones could be associated to regions with higher percentage of crosslinked polymer (%N is lightly higher in these zones), while the free polymer distributed in the sIPN being related to darker zones. Samples with different degree of chloromethylation as well as crosslinking degree are observed by SEM but non-significant differences are detected. All membranes exhibit homogeneous microstructures.

References

- [1] X. Lin, Y. Liu, S. D. Poynton, A. L. Ong, J. R. Varcoe, L. Wu, Y. Li, X. Liang, Q. Li, T. Xu, Cross-linked anion exchange membranes for alkaline fuel cells synthesized using a solvent free strategy, *J. Power Sources*, 233 (2013) 259-268.
- [2] X. Lin, M. Gong, Y. Liu, L. Wu, Y. Li, X. Liang, Q. Li, T. Xu, A convenient, efficient and green route for preparing anion exchange membranes for potential application in alkaline fuel cells, *J. Membr. Sci.* 425-426 (2013) 190-199.

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Table S1. AEMs based on sIPNs

Membrane	Parameters	
sIPN-TMA-OH DCL 20% and 6/4	DC 58, 80, 111, 118, 130, 143	
sIPN-TMA-OH DC 111% and 6/4	DCL 0, 5, 10, 15, 20	
sIPN-TMA-OH DC 100% and DCL 20%	Crosslinked polymer/PSU 6/4 7/3 9/1	
sIPN-MIm-OH DC 80%	DCL 0, 5, 15	Crosslinked polymer/PSU 6/4, 9/1
sIPN-MIm-OH DC 100%	DCL 0, 5, 15	Crosslinked polymer/PSU 6/4, 9/1
sIPN-MIm-OH DC 143%	DCL 0, 5, 15	Crosslinked polymer/PSU 6/4, 9/1
sIPN-DMIm-OH DC 80%	DCL 0, 5, 15	Crosslinked polymer/PSU 6/4, 9/1
sIPN-DMIm-OH DC 143%	DCL 0, 5, 15	Crosslinked polymer/PSU 6/4, 9/1

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Table S2. WU and IEC of AEMs in the proportion 6/4 crosslinked polymer/PSU

DCL	sIPN-TMA-OH				sIPN-MIm-OH				sIPN-DMIm-OH			
	WU ^a		IEC		WU ^a		IEC		WU ^a		IEC	
	%		(mmol g ⁻¹)		%		(mmol g ⁻¹)		%		(mmol g ⁻¹)	
	DC											
	87	>100	87	>100	75	143	75	143	80	143	80	143
20%	0.9± 0.1	12±4	0.78± 0.02	-	-	-	-	-	-	-	-	
15%	-	5±1	-	0.62±0. 02	-	-	-	-	33±4	34±4	-	-
5%	-	0.9±0. 1	-	0.77±0. 03	9± 1	22± 2	1.45± 0.05	2.60± 0.04	10±1	27±3	1.51± 0.01	1.48± 0.03
0%	-	-	-	-	8± 1	-	1.20± 0.03	-	14±3	17±3	1.84± 0.04	1.48± 0.02

^a *T* = 25 °C.

To substantiate the influence of DC and DCL on dimensional stability of the sIPN membranes, we further studied WU. The obtained results are gathered in Table S2. WU values are relatively low in all membranes, i.e., in all cases are lower than 35%.

Highly functionalized membranes show higher hydrophilicity and consequently, higher WU%. This fact can be easily explained taking into account that the cationic group anchored to the backbone increases polarity.

WU% is significantly higher when the DCL% increased. This tendency could be related to the increase in polarity due to the presence of TMEDA, a diamine with two aliphatic nitrogen atoms with high hydration ability.

The capacity of the functionalized-sIPN membranes to exchange ions is also studied Table S2. As expected, WU and IEC data exhibit a correlation. However, non-clear trends are observed when both DC and DCL vary.

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Table S3. WU and IEC for AEMs at different proportion of crosslinked polymer/PSU

DCL	sIPN-MIm-OH ^a				sIPN-DMIm-OH ^a			
	WU ^b		IEC		WU ^b		IEC	
	%		(mmol g ⁻¹)		%		(mmol g ⁻¹)	
	Crosslinked polymer/PSU							
	6/4	9/1	6/4	9/1	6/4	9/1	6/4	9/1
15%	14±3	18±3	1.18±0.04	1.82±0.05	33±4	-	-	-
5%	19±2	24±3	1.59±0.01	1.56±0.03	10±1	-	1.51±0.01	1.45±0.03
0%	35±1	41±1	1.29±0.02	1.60±0.03	14±3	-	1.84±0.04	-

^a DC ~ 80-103%; ^b T = 25 °C.

The crosslinked polymer/free PSU ratio has a direct effect in both WU% and IEC parameters. Thus, WU and IEC values increase as the proportion of crosslinked and functionalized polymer (hydrophilic part) increased in the blend. This behaviour can be explained in polarity terms. The polar segment absorbs higher amount of water than the less, or -non- polar one, and it is also responsible for the ion exchange in the membrane. sIPN-DMIm membrane with a ratio of 9/1 could not be prepared due to its high fragility.

Supporting Information

Table S4. Mechanical properties of sIPN-TMA membranes (DC = 100% and DCL = 20%. Tensile Strength (TS) with different crosslinked polymer/PSU ratio, counter ion and hydration conditions

sIPN-TMA									
Crosslinked polymer/PSU									
6/4		7/3				9/1			
TS (MPa)									
OH^-_{wet}	Cl^-_{dry}	Cl^-_{wet}	OH^-_{dry}	OH^-_{wet}	Cl^-_{dry}	Cl^-_{wet}	OH^-_{dry}	OH^-_{wet}	
36.1±1.0	43.2±2.6	48.9±0.4	36.8±0.4	43.8±1.8	25.9±6.7	36.5±11.1	28.1±4.1	23.3±10.6	