

Shape-Memory Composites Based on Ionic Elastomers

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TGA

Thermogravimetric analysis (TGA) was carried out on a TA discovery (TA Instruments, USA), by equilibrating at 100 °C for 20 min and with a heating ramp of 10 °C/min under a N₂ atmosphere up to 650 °C and O₂ atmosphere up to 1000 °C.

To verify the correct incorporation of the different fillers to the matrix, a thermogram of the study samples is represented in Figure S1. Two differentiated processes can be seen in these thermograms: (i) weight loss up to 400 °C corresponding to the degradation of the elastomeric matrix; And (ii) the decomposition temperature of the different carbonaceous fillers, corresponding to the degradation of the main carbon structure.

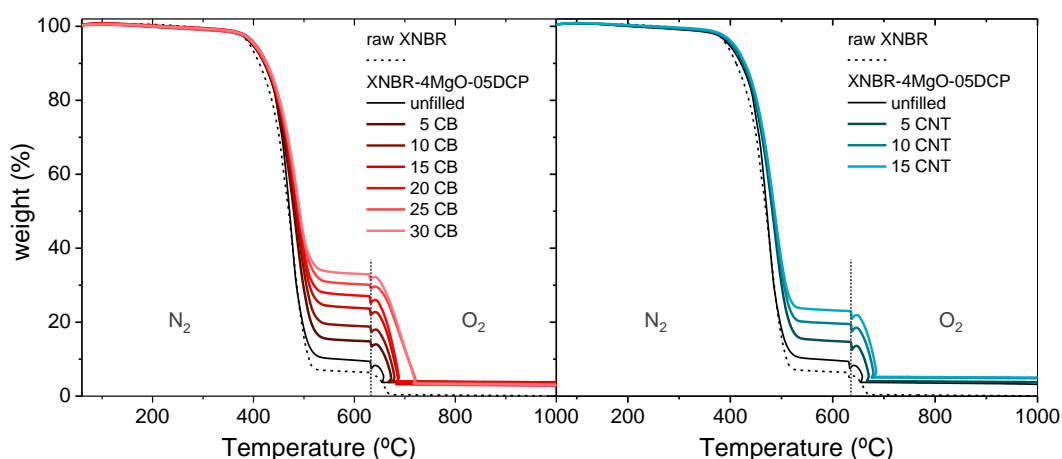


Figure S1. Thermograms of elastomeric samples between 50 and 1000 °C at 30 °C / min in which the degradation of the matrix and the decomposition of the carbon black and carbon nanotubes fillers is observed in an N₂ (up to 650 °C) and O₂ atmosphere (up to 1000 °C).

Comparing the results obtained from the TGA of the vulcanized samples with the filler content introduced in the preparation of the different compounds (raw, before their vulcanization), we obtain the data reflected in Figure S2.

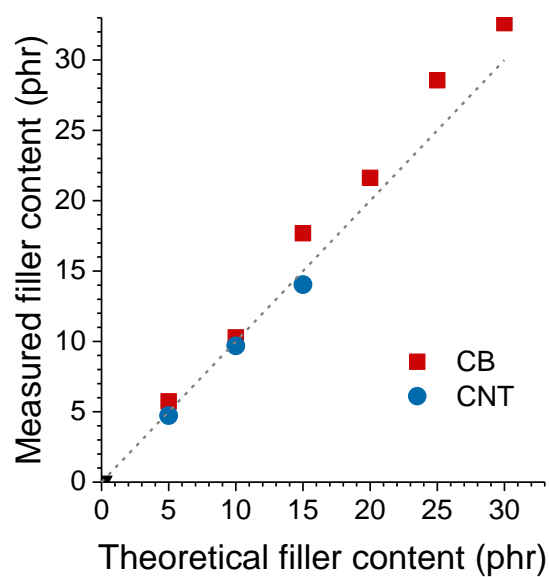


Figure S2. Relationship between the theoretical content introduced in the formulation of the samples and the actual filler content calculated by TGA. Dashed line shows a 1:1 ratio.

Therefore, the incorporation of the filler to the elastomeric compound is totally effective and it is possible to characterize the variation of its properties as a function of these once the pristine sample has been previously characterized.

FEG-SEM

To visualize the size and distribution of fillers in the elastomeric XNBR matrix samples were analyzed by FEG-SEM (Nova NanoSEM FEI 230). Representative images for carbon black and multiwalled carbon nanotubes filled samples are shown in Figures S3 and S4 respectively.

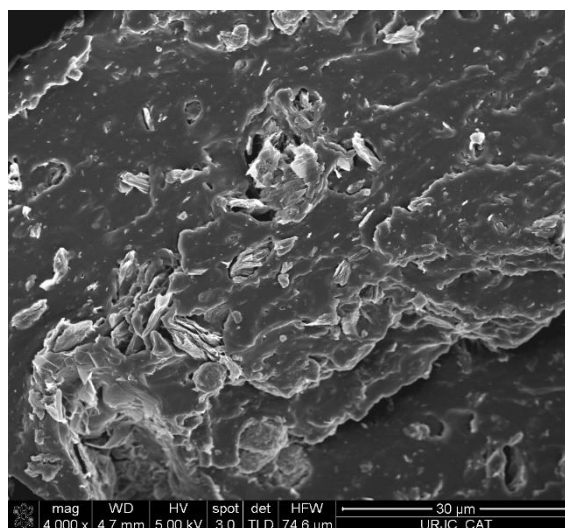


Figure S3. FEG-SEM image of sample “10CB” at low magnification (4000x)

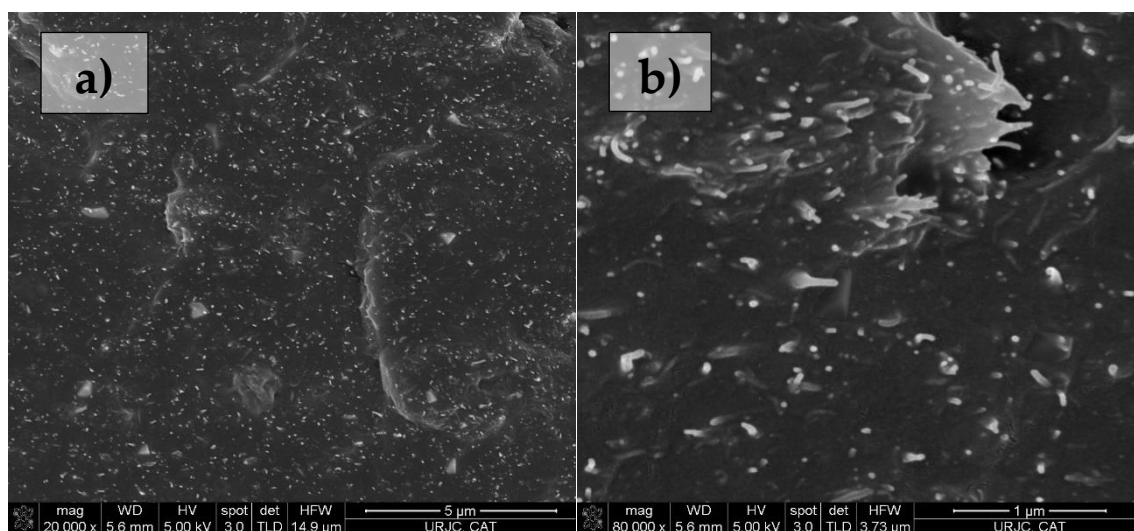


Figure S4. FEG-SEM images of sample “10CNT” at high magnification [a) 20000x, and b) 80000x.]

DMA data results

Table S1. Data results obtained from DMTA study of the samples. T_g bulk and T_{ionic} were obtained as the maximum of the $\tan \delta$ peaks in the study.

Sample	E' at $-25\text{ }^{\circ}\text{C}$	E' at $25\text{ }^{\circ}\text{C}$	T_g bulk	T_{ionic}
	MPa	MPa	$^{\circ}\text{C}$	$^{\circ}\text{C}$
Unfilled	2900	12.0	-5.3	66.9
5CB	2113	15.2	-4.6	66.8
10CB	2442	22.9	-4.1	74.1
15CB	2597	32.4	-3.7	73.8
20CB	2811	54.5	-3.9	79.8
25CB	2890	75.7	-4.6	86.7
30CB	3009	122.5	-4.8	82.0
5CNT	2319	22.5	-3.2	80.1
10CNT	2780	42.6	-2.9	82.8
15CNT	3385	92.5	-2.7	86.0

Shape-memory data results

Table S2. Fixing (R_f) and recovery (R_r) ratios of the shape memory for 4 consecutive cycles in each sample.

Sample	R_f				R_r			
	%				%			
Cycle:	1	2	3	4	1	2	3	4
Unfilled	78.6	80.4	81.4	82.1	81.5	86.6	92.6	95.2
5CB	81.1	81.5	82.0	82.2	91.5	96.0	96.0	96.8
10CB	85.1	85.8	86.2	86.5	87.8	92.7	94.9	95.3
15CB	85.0	85.4	85.7	86.0	84.7	94.5	95.5	95.9
20CB	86.6	87.2	87.4	87.7	91.8	93.9	94.8	95.3
25CB	86.7	87.8	87.9	88.0	85.3	89.5	93.3	94.8
30CB	87.9	88.4	88.7	89.0	75.2	85.3	91.1	93.3
5CNT	82.4	83.1	83.5	83.9	90.2	95.6	96.8	97.0
10CNT	86.6	87.4	87.9	90.0	90.1	93.1	94.6	94.6
15CNT	89.3	90.1	90.3	90.6	86.9	92.0	95.1	95.5