

SUPPLEMENTARY INFORMATION

Unlocking the Potential of Self-Healing and Recyclable Ionic Elastomers for Soft Robotics Applications

S. Utrera-Barrios^{1,*}, N. Steenackers^{2,3}, S. Terryn^{2,3}, P. Ferrentino³, R. Verdejo¹,
G. Van Asche², M. A. López-Manchado¹, J. Brancart², M. Hernández Santana^{1,*}

¹ Institute of Polymer Science and Technology (ICTP), CSIC, Juan de la Cierva 3, 28006 Madrid, Spain

² Physical Chemistry and Polymer Science (FYSC), Department of Materials and Chemistry (MACH), Vrije Universiteit Brussel (VUB), Pleinlaan 2, B-1050 Brussels, Belgium

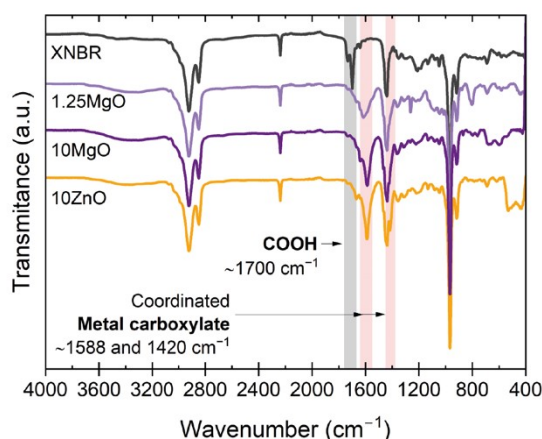
³ Brubotics, Vrije Universiteit Brussel (VUB) and Imec, Pleinlaan 2, B-1050 Brussels, Belgium

* corresponding authors: sutrera@ictp.csic.es; marherna@ictp.csic.es

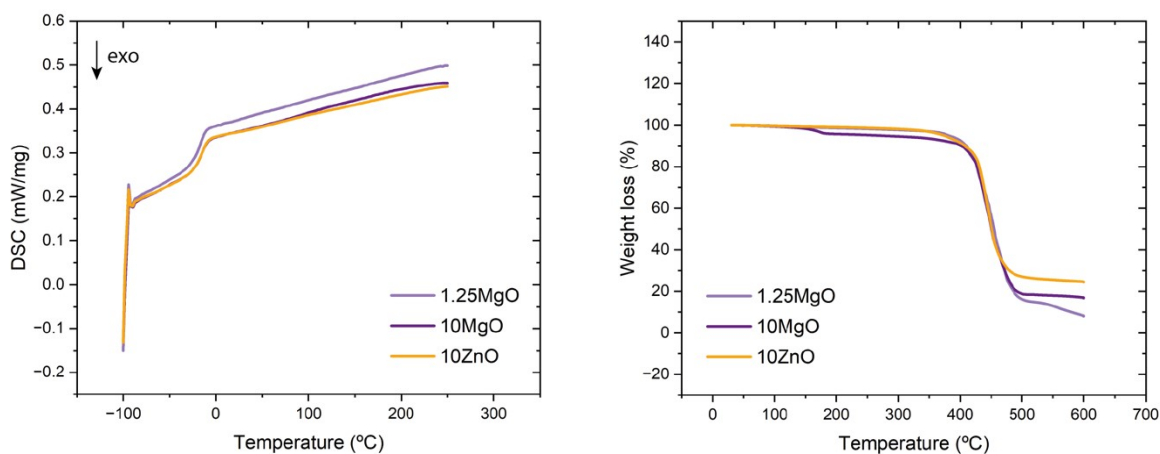
Supplementary Information S1. Product data for Krynac X 750 supplied by Arlanxeo.

Property Name	Nominal Value	Test Method
Mooney viscosity	47 MU	ISO 289 / ASTM D 1646 / ML(1+4) 100 °C
Acrylonitrile (ACN) content	27.0 wt. %	ISO 24698
Carboxyl (COOH) groups	7.0 wt. %	-
Density	0.99 g/cm ³	Internal method

Supplementary Information S2. IR spectra of the ionic elastomers.



Supplementary Information S3. DSC and TGA results.



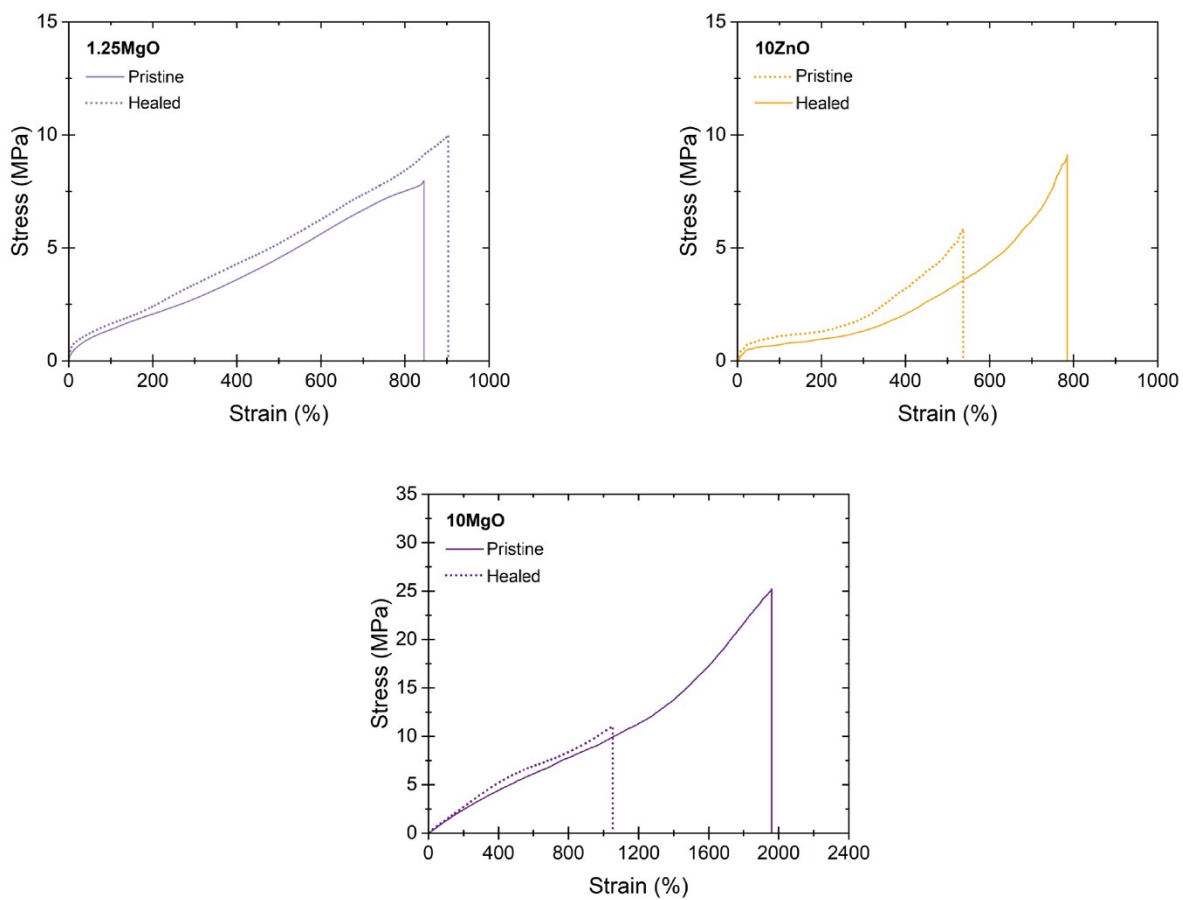
Differential scanning calorimetry (DSC): A differential scanning calorimeter (DSC 214 Polyma, Netzsch) was used to determine the thermal properties of the compounds in a nitrogen atmosphere. A heating sweep (from $-100\text{ }^{\circ}\text{C}$ to $250\text{ }^{\circ}\text{C}$) was performed at $10\text{ }^{\circ}\text{C min}^{-1}$.

Thermogravimetric analysis (TGA): A thermal analyzer (TGA 2, Mettler Toledo) was used. A heating sweep from room temperature to $600\text{ }^{\circ}\text{C}$ (in a nitrogen atmosphere) was performed, using a heating rate of $10\text{ }^{\circ}\text{C min}^{-1}$. The recorded spectra were analyzed using TA Universal Analysis software.

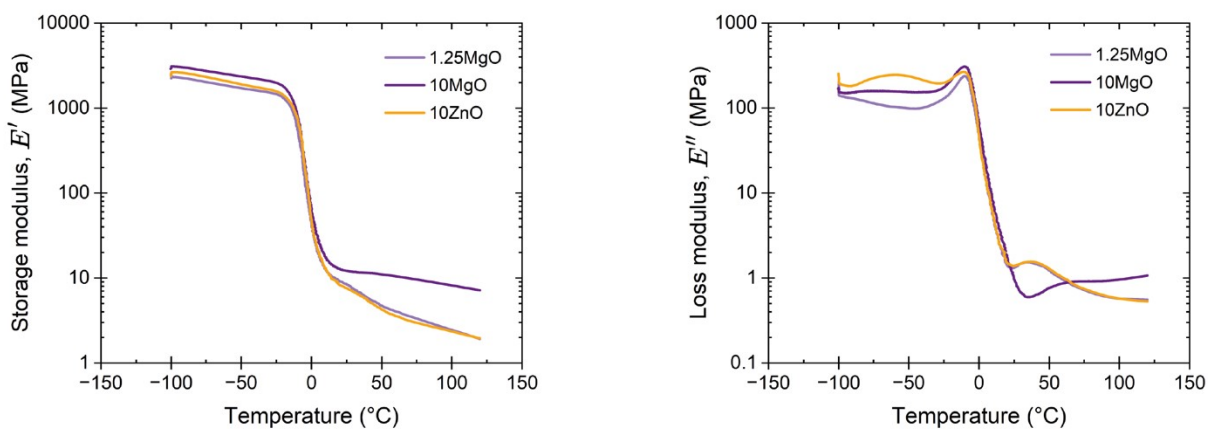
Supplementary Information S4. Healing efficiency based on M300 values of the ionic elastomers.

Time (h)	1.25MgO	10MgO	10ZnO
3	106 ± 5	106 ± 7	165 ± 54
5	103 ± 1	-	120 ± 15
7	78 ± 5	-	140 ± 7

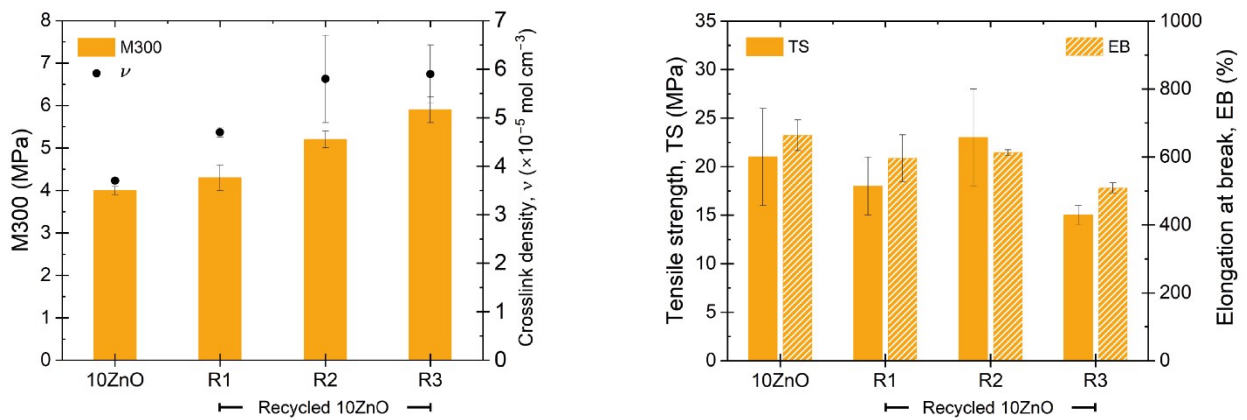
Supplementary Information S5. Stress-strain curves of the ionic elastomers in pristine and healed state (healing protocol: 110 °C, 3 h, rectangular specimens).



Supplementary Information S6. Storage (E') and Loss moduli (E'') by DMA.



Supplementary Information S7. Evolution of the mechanical properties (M300, TS, and EB) and crosslink density of 10ZnO through three recycling cycles.



Adapted from *Materials & Design*, 2023, 233, 112273, DOI: 10.1016/j.matdes.2023.112273 with permission from Elsevier.

Supplementary Information S8. Video of the actuation.

https://drive.google.com/file/d/1kmG13XX5t1Q-3gGtjqhq1kDD0_2-dhPz/view?usp=sharing