Preparation and characterization of self-healing PVA-H_2SO_4 hydrogel for flexible energy storage

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Supporting Information

Preparation of PANI-PAMPSA

In a 1 L round bottom flask, 300 mL of demineralized water was added, then 2.54 mL of aniline and 58.0 g of PAMPSA were added and stirred for 1 h at room temperature. Successively, 5.8 g of ammonium persulfate (APS) was dissolved in in 25 mL of distilled water, with a fixed aniline to APS molar ratios of 1 and was slowly added dropwise to the stirred suspension. The flask was cooled to 0 °C in an ice bath for 6 h. After 24 h the polymeric suspension, containing the 3.5 % PANI_PAMPSA, was ready.

Preparation of Hydrogel membranes

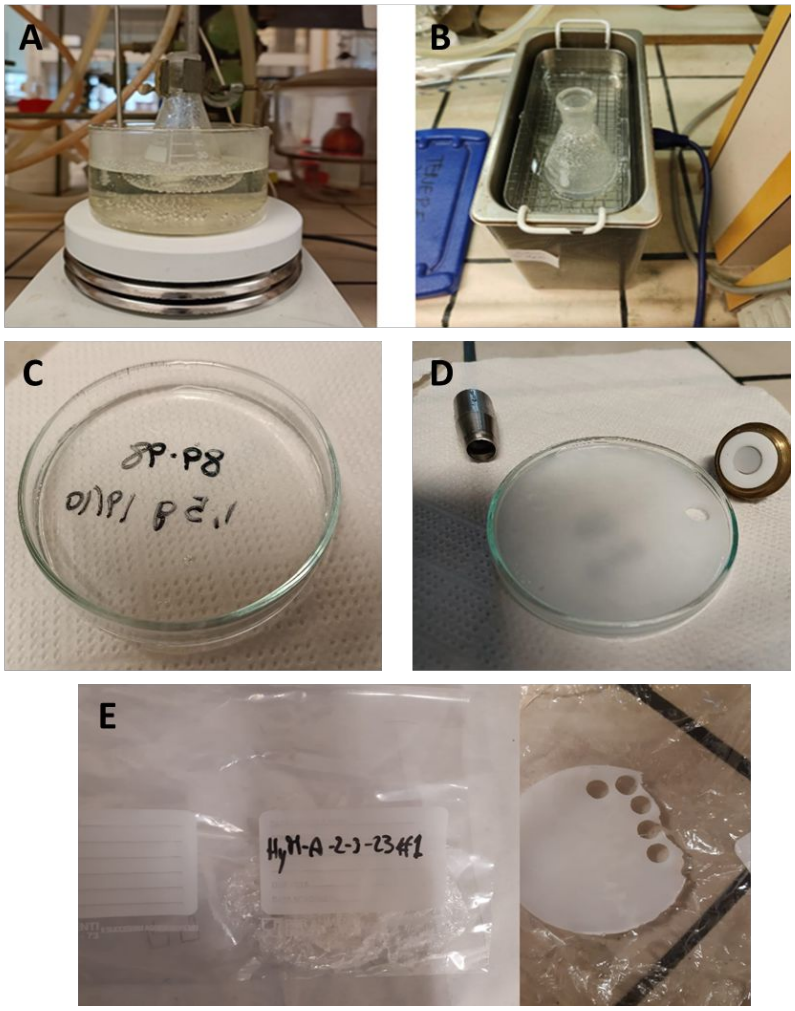

Figure S1. PVA-H_2SO_4 hydrogel preparation: A) PVA powder dissolution in 1.0 M H_2SO_4 at 70°C under vigorous stirring; B) ultrasound treatment to eliminate bubbles; C) hydrogel solution before and D) after 3 freeze-thaw cycles; E) hydrogel membrane storage.

Characterization

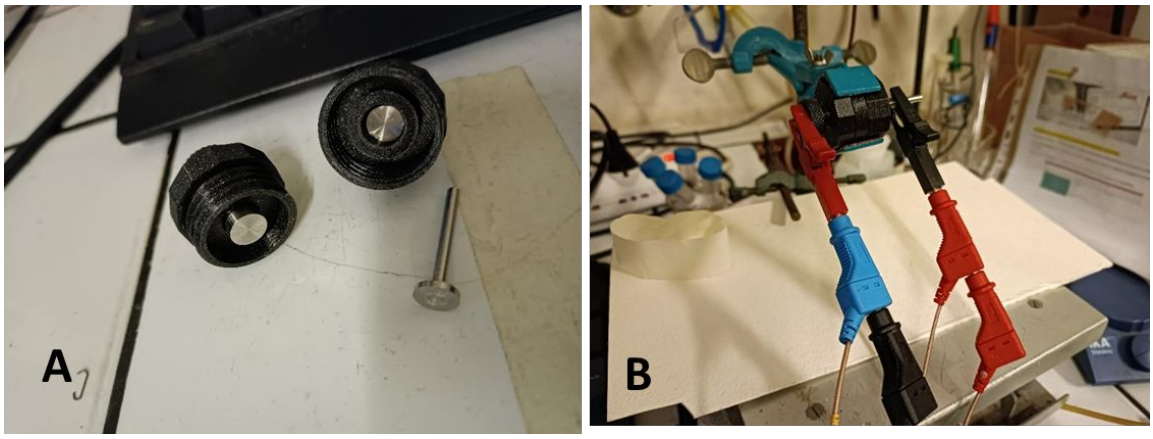

Figure S2. A) Swagelok type cell with a 316 stainless steel caps of 1.0 cm diameter; B) configuration for electrochemical measurements.

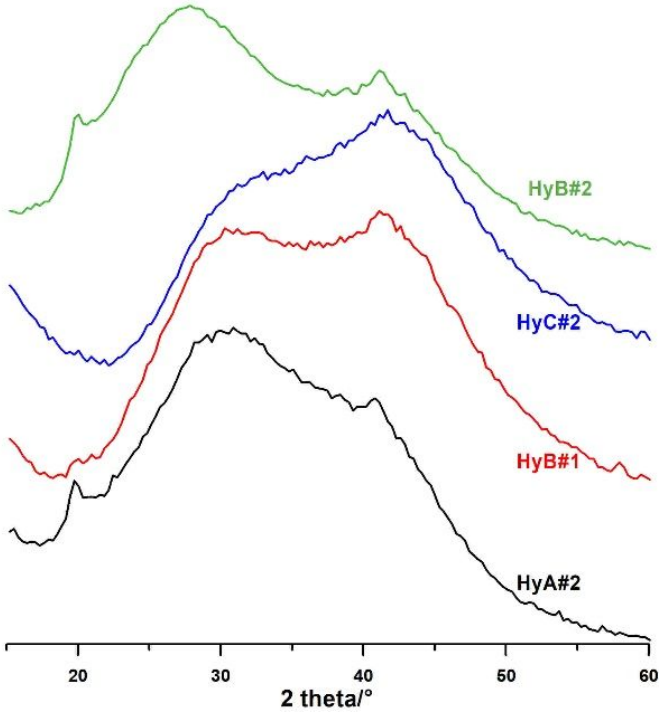

Figure S3. X-Rays diffraction patterns of the investigated samples.

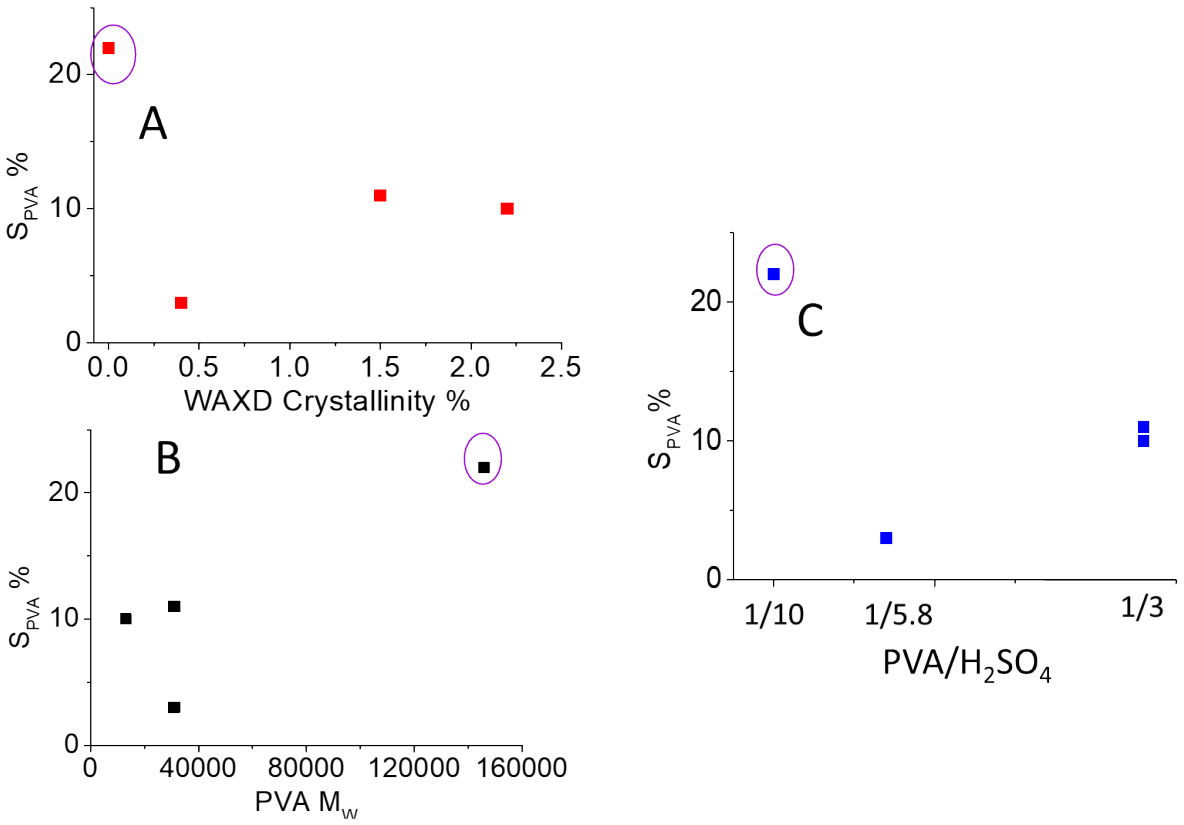

Figure S4. SPVA as a function of: A) WAXD Crystallinity %; B) PVA Mw and C) PVA/H_2SO_4 w/w ratio

Table S1. T_g values of PVA hydrogels for each freeze-thaw cycle.

Sample	Cycle- Tg (°C)*
HyA#2	II-24
	III-26
	IV-29
HyB#1	I-38
	II-42
	III-42
HyB#2	I-37
	II-41
	III-43
HyC#2	I-42
	II-42
	III-42

*Evaluated using the midpoint of the step-change

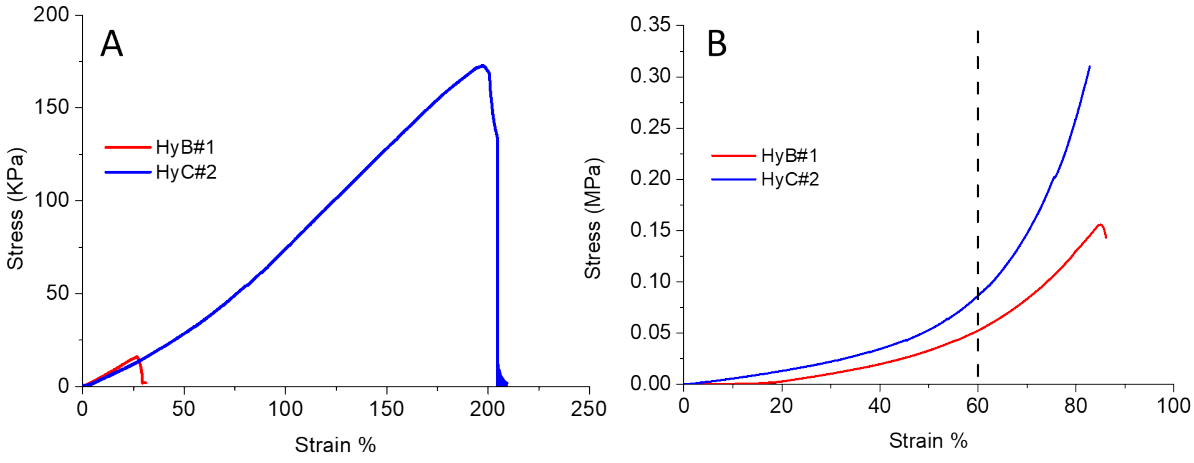

Figure S5. A) Tensile stress/strain curves and B) compression stress/strain curves for HyB#1 and HyC#2.

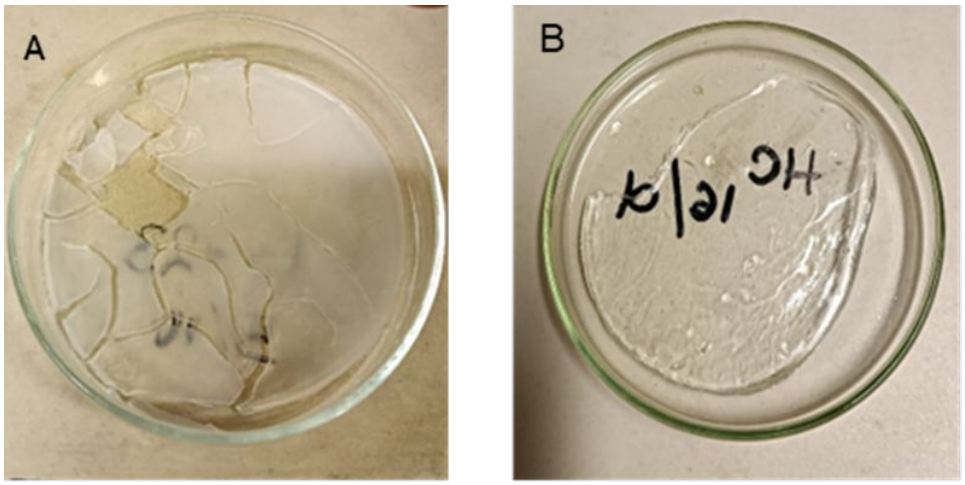

Figure S6. Pictures of the hydrogels after 3 months of storage in cling film for food at room temperature: A) HyA#2 and B) HyC#2.

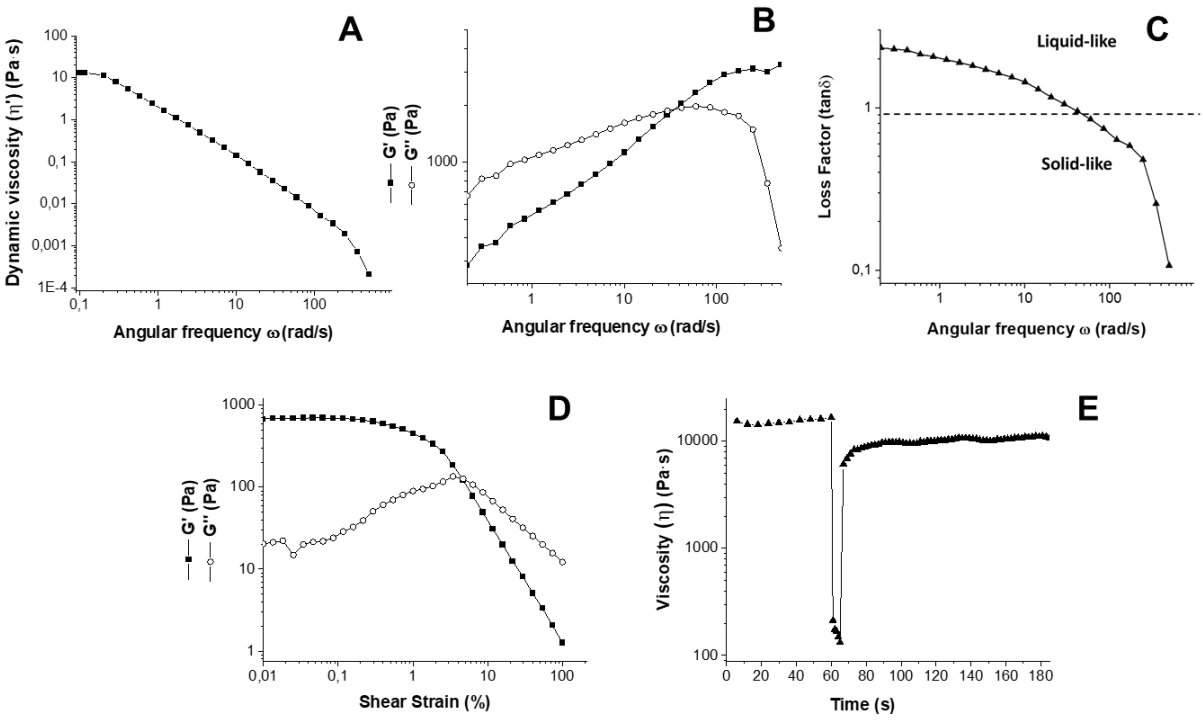

Figure S7. A) Dynamic viscosity (η') versus angular frequency curve; B) Frequency sweep of PVA-based hydrogel; C) Tanδ versus angular frequency curve; D) Amplitude sweep curves of PVA-based hydrogels; E) 3ITT test.

Table S2. Specific capacitance and Current density for PANI-PVA based supercapacitors.

Sample	Specific Capacitance ($mF\ cm^{-2}$)	Current density ($mA\ cm^{-2}$)	Ref
PPH_{003}	602	1.0	[1]
PH-A	237	0.5	[2]
PANI-PVA	260.1	0.5	[3]

* PPH_{003}= PVA/PANI hydrogel with the molar ratio of Maba to aniline equal to 0.03; PH-A = PANI-PVA hydrogel.

References

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(3) Lai, F.; Fang, Z.; Cao, L.; Li, W.; Lin, Z.; Zhang, P. Self-Healing Flexible and Strong Hydrogel Nanocomposites Based on Polyaniline for Supercapacitors. *Ionics* **2020**, *26*, 3015–3025. https://doi.org/10.1007/s11581-020-03438-3.