Supplementary Information

Biopolymeric hydrogel electrolytes obtained by natural polysaccharide-poly(itaconic acid-co-2hydroxyethyl methacrylate) in deep eutectic solvents for rechargeable Zn-air batteries

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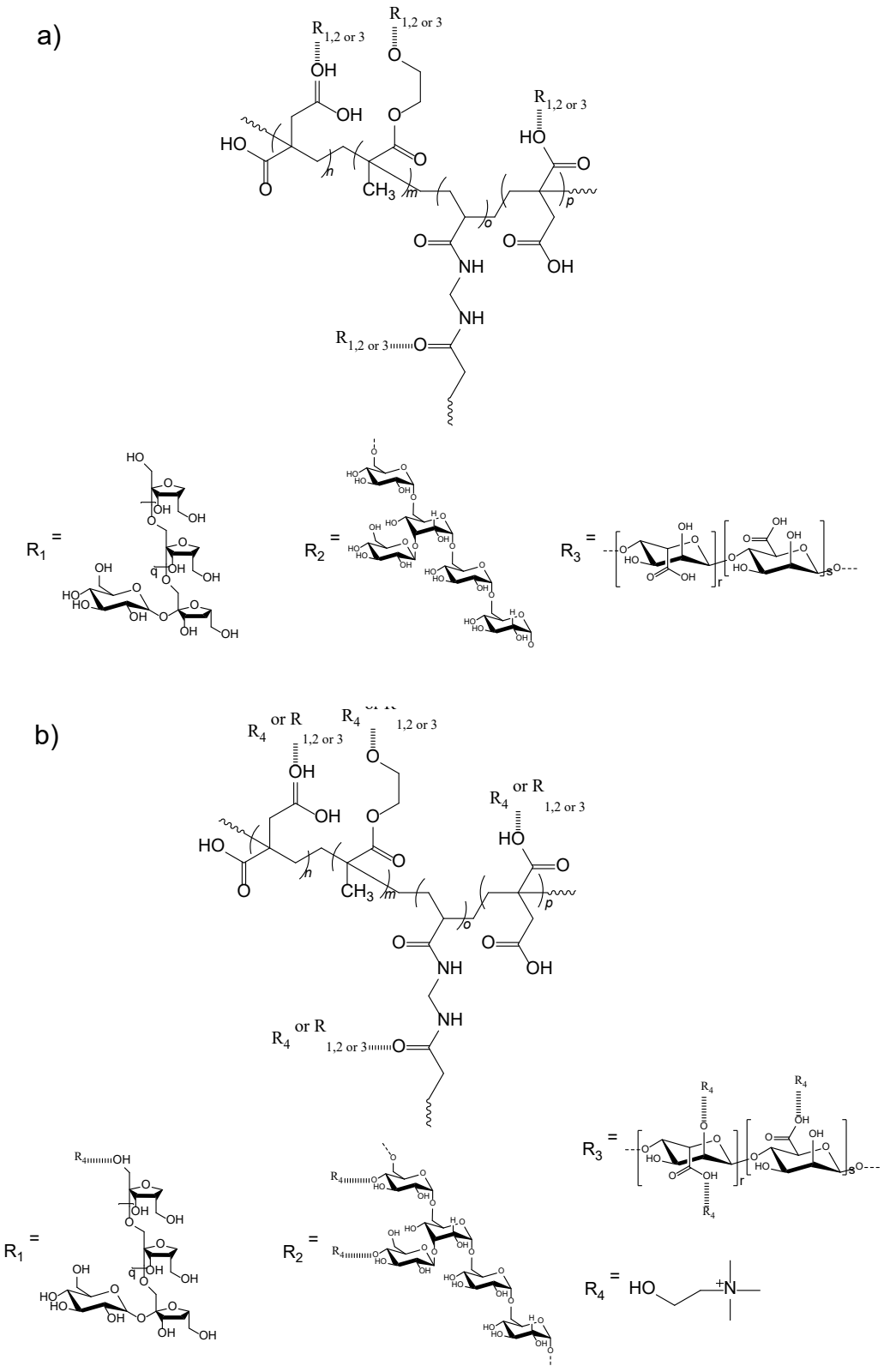

Fig. S1. a) Chemical structure proposed for the biohydrogels structure and b) chemical structure of the biohydrogels with choline moieties in the structure.

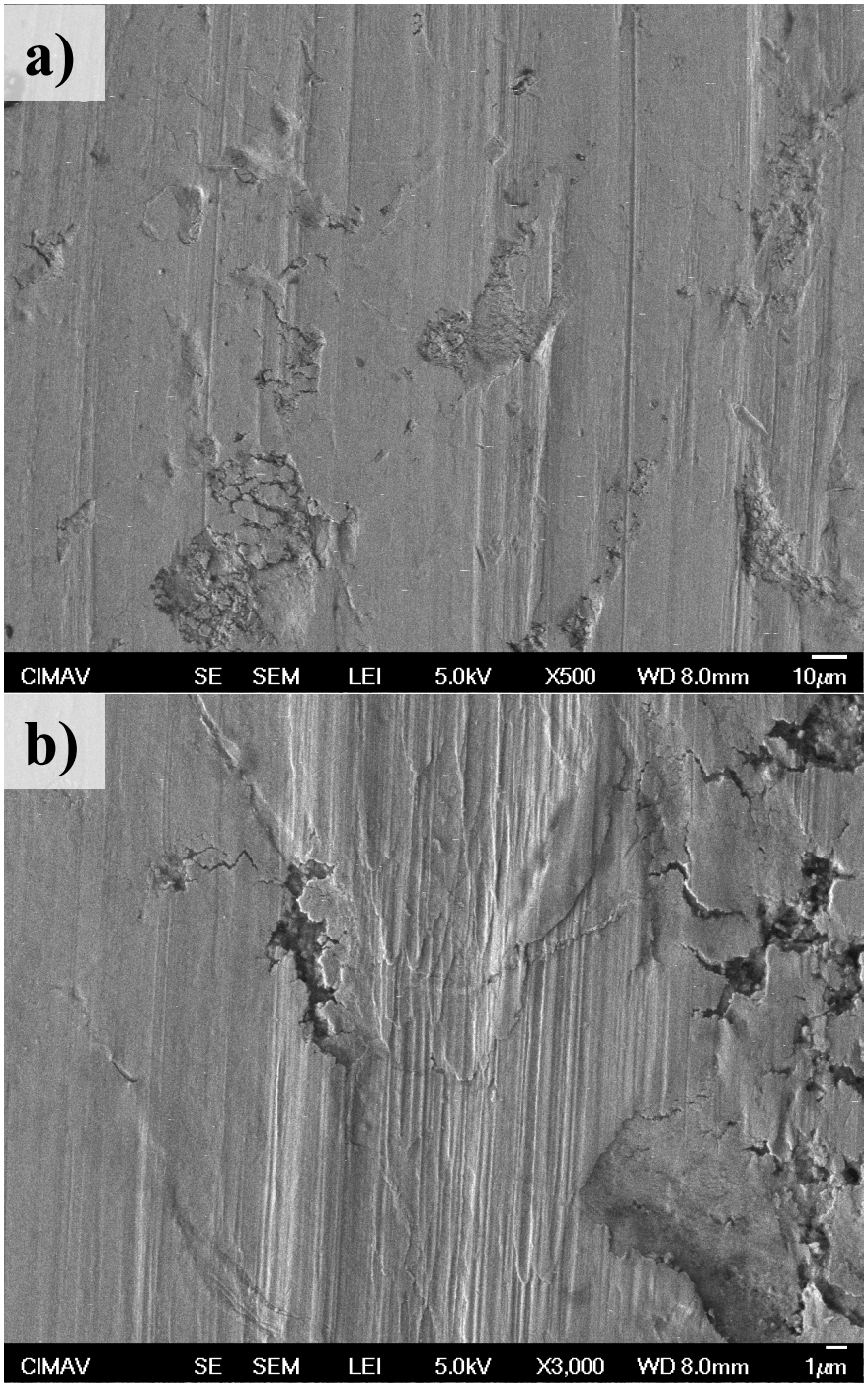

Fig. S2. SEM images of pure Zn foil before its use in the Zinc-air battery.

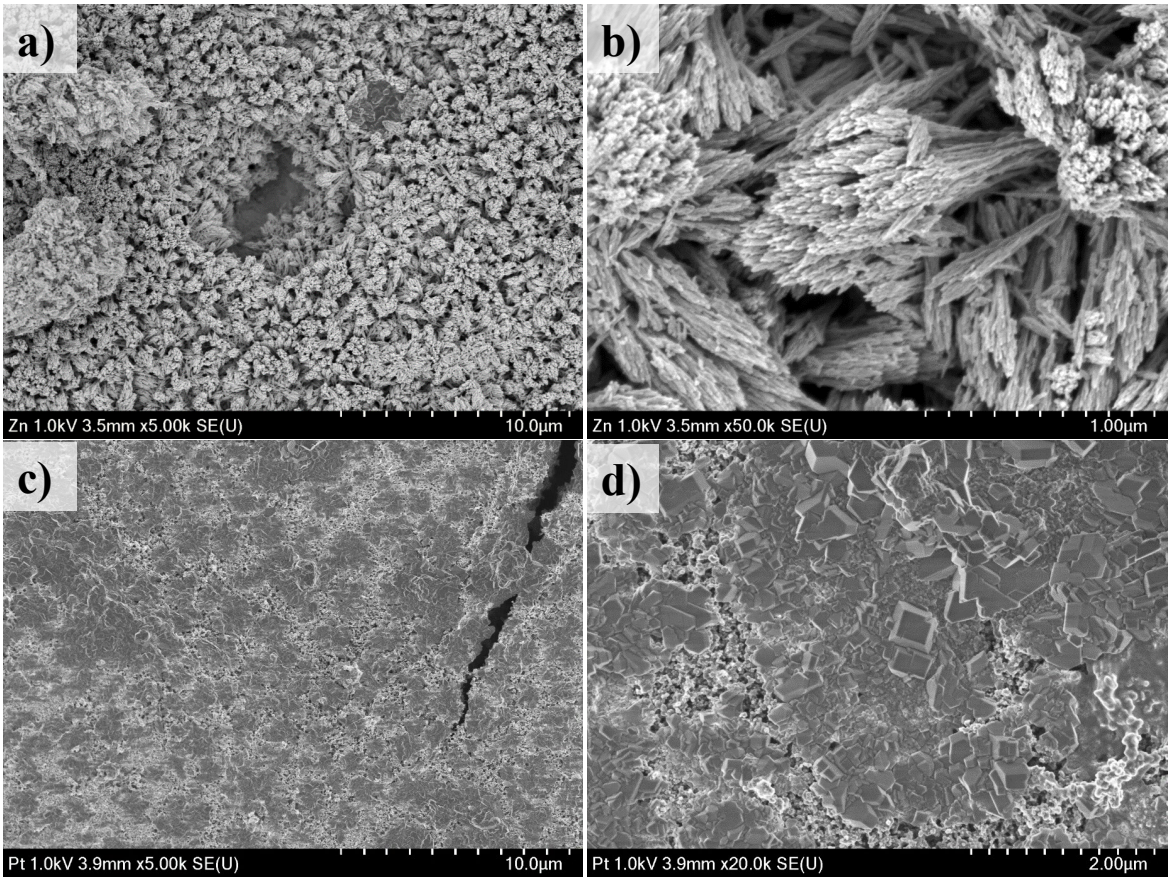

Fig. S3. SEM micrographs at different magnifications of (a, b) Zn anode, and (c, d) Pt electrode for the *post-mortem* analysis of the ZAB discharged at 6.6 mA cm^{-2} during 5 h.

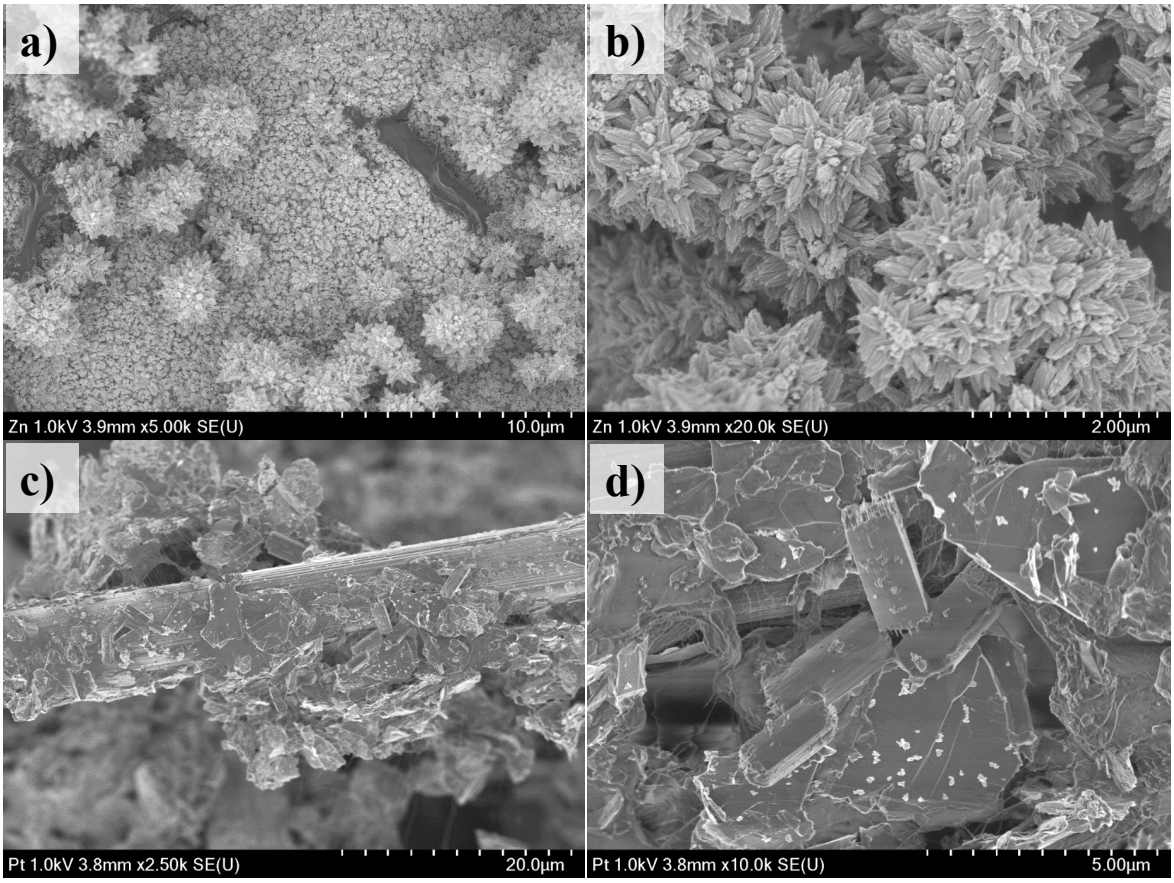

Fig. S4. SEM micrographs at different magnifications of (a, b) Zn anode, and (c, d) Pt electrode for the *post-mortem* analysis of the ZAB discharged at 13 mA cm^{-2} during 5 h.

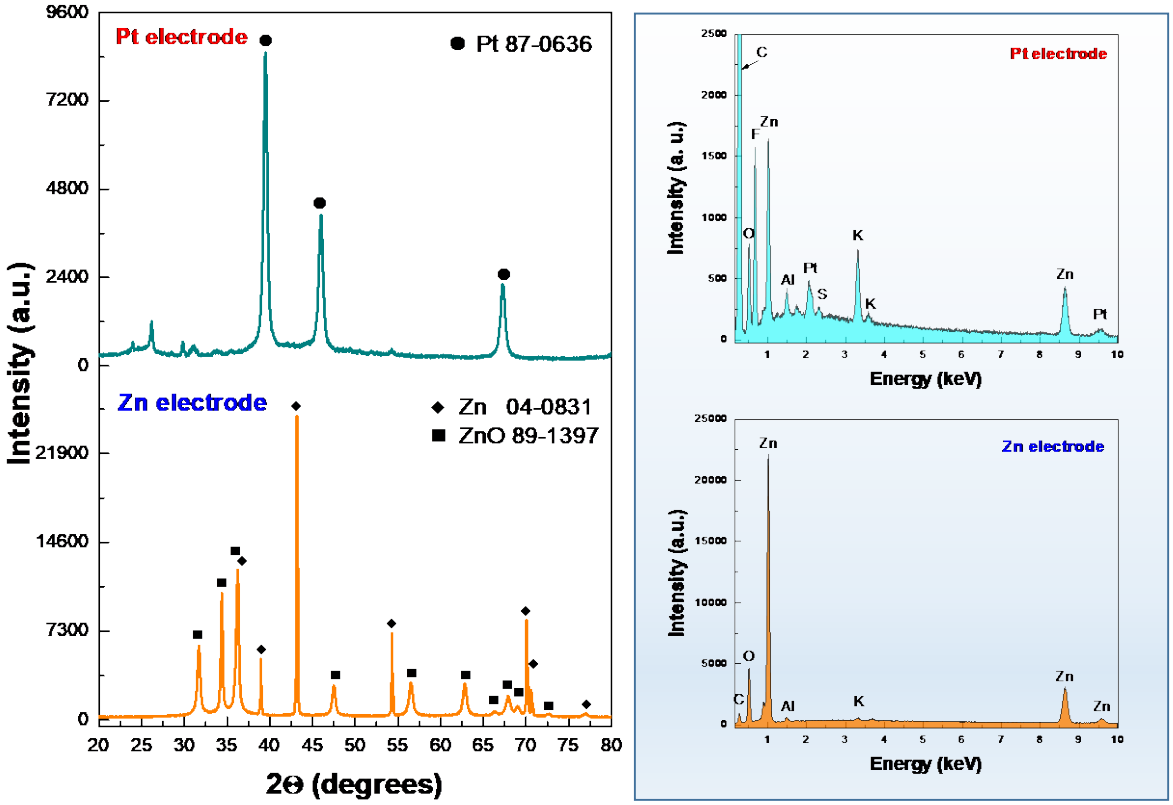

Fig. S5. a) X-ray diffraction patterns of Pt and Zn electrodes, and b) EDS analyzes after discharging the ZAB at 13 mA cm^{-2} during 5 h.

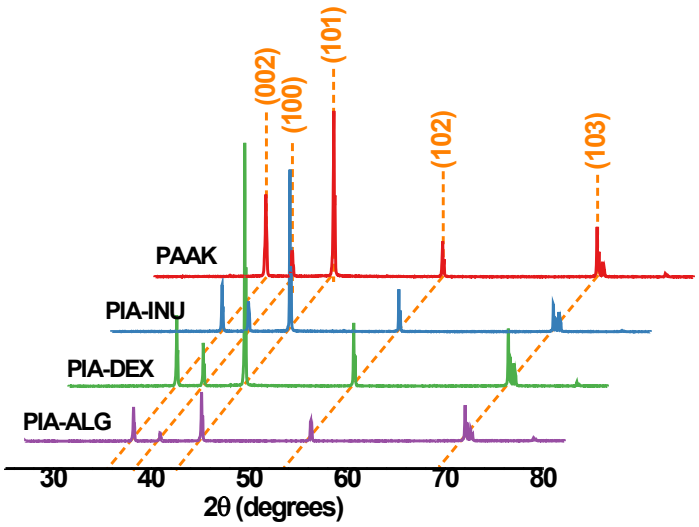

Fig. S6. X-ray diffraction patterns for the Zn anodes after performing the stability tests on the ZAB by demanding different current densities.

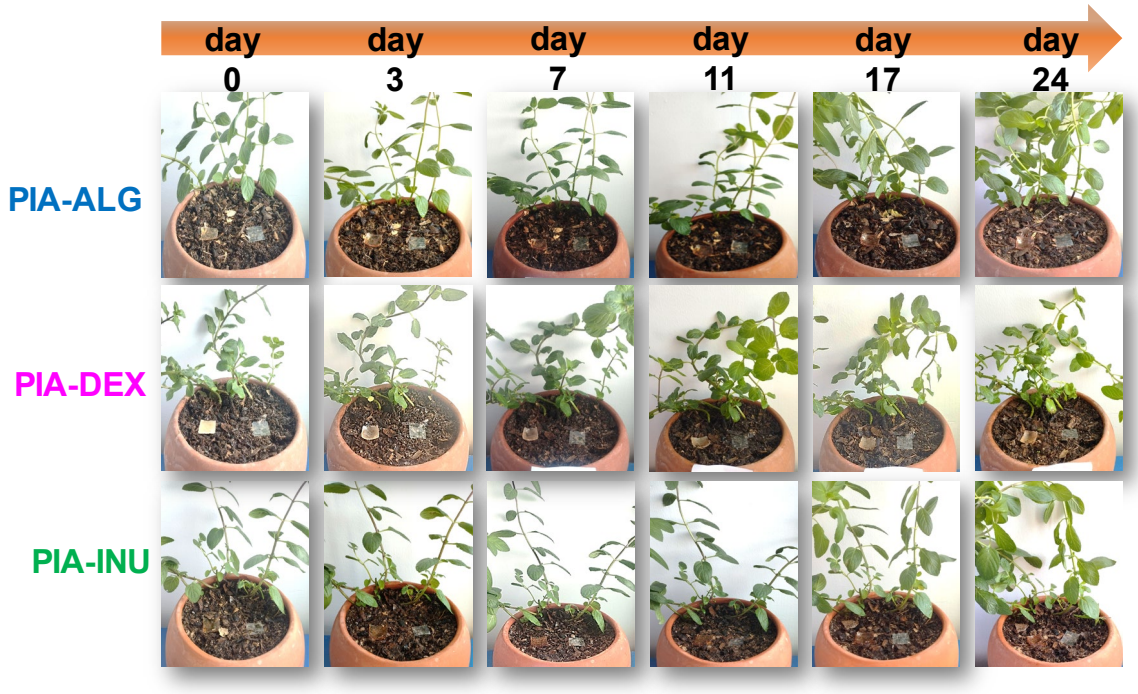

Fig. S7. Biodegradability photographs of biohydrogels using *Mentha piperita* pots as environmental degradation model, after 0, 3, 7, 11, 17 and 24 days.

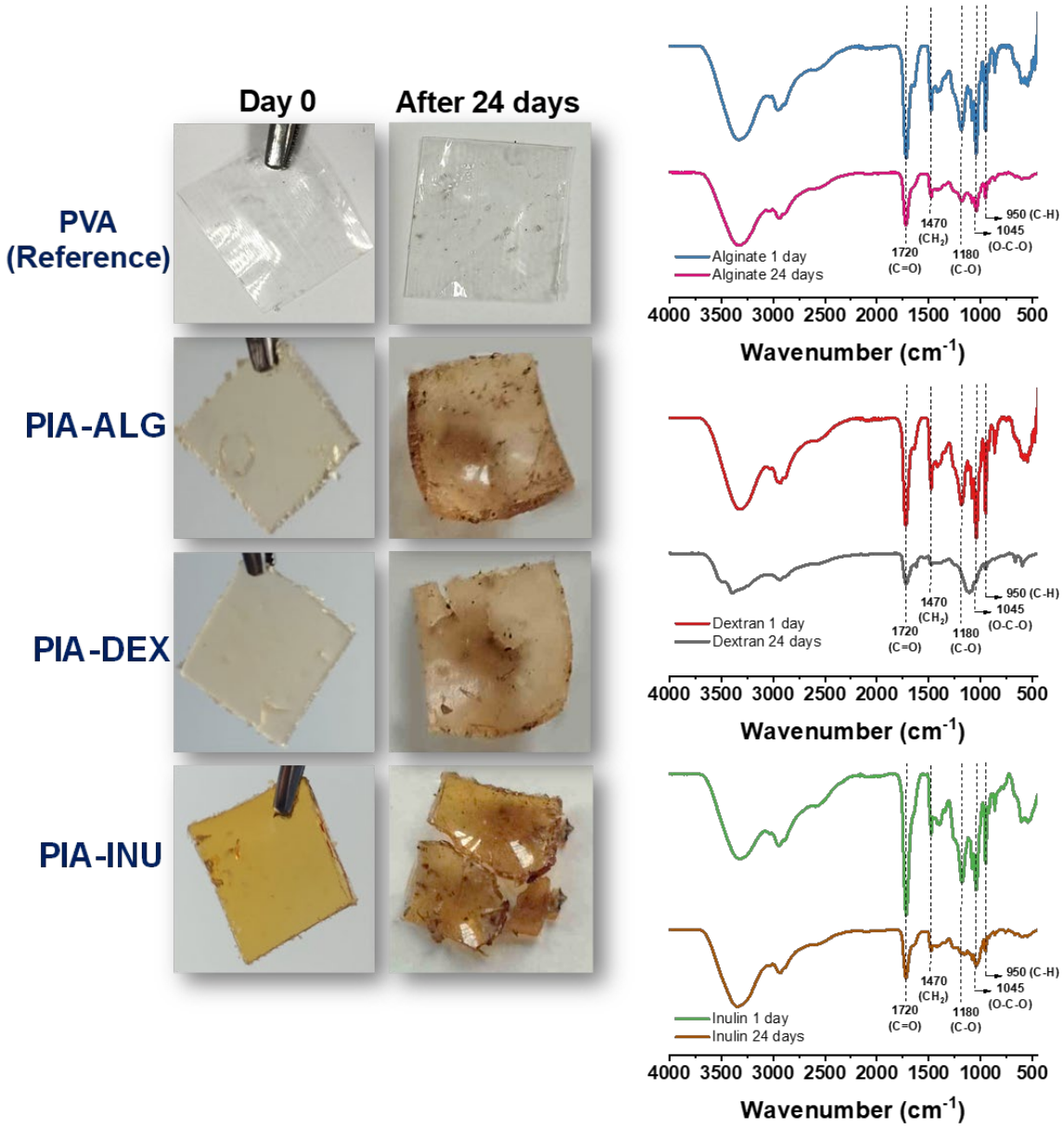

Fig. S8. a)Photographs of biohydrogels after being exposed to the soil surface after 0 and 24 days, and PVA membrane as reference. b) FTIR of the biohydrogels after exposure to the soil surface after 0 and 24 days.

Table S1. Summary of resistances found for the ZABs operated with the biohydrogels at 3 and 10 wt%.

Biohydrogel	Biohydrogel concentration	R1 (Ω)	R2 (Ω)
PIA-INU	3 wt. %	2.75	3.85
PIA-DEX	3 wt. %	3.27	8.7
PIA-ALG	3 wt. %	3.62	9.09
PAAK	3 wt. %	3.32	8.80
PIA-INU	10 wt. %	3.06	22.70
PIA-DEX	10 wt. %	3.03	6.80
PIA-ALG	10 wt. %	5.84	9.56
PAAK	10 wt. %	3.48	61.82

Table S2. Battery performance comparison with recently reported Zinc-air batteries operated with hydrogels.

Hydrogel	Air-electrode mass loading (mg cm^{-2})	Open-circuit potential (V)	Current density @ 0.6 V (mA cm^{-2})	Power density (mW cm^{-2})	Number of functional cycles of charge/discharge	Ref.
Polyacrylic acid (PAA) soaked in 6 M KOH	0.6 (mixture of 40 % Pt, 20 % Ru, 40 % C)	Not provided	Not provided	Not provided	25 cycles demanding 0.5 mA cm^{-2}; time= 10 min/cycle; total duration of the ZAB= 4.1 h	1
Polyvinyl alcohol (PVA) soaked in 6 M KOH	0.6 (mixture of 40 % Pt, 20 % Ru, 40 % C)	Not provided	Not provided	Not provided	24 cycles demanding 0.5 mA cm^{-2}; time= 10 min/cycle; total duration of the ZAB= 4 h	1
Starch gel (from flour) soaked in 6 M KOH	2.0 (Mn-Co-Fe@carbon nanotubes)	1.48	90 @ 0.95 V	~84	Cycles were performed at 2 mA cm^{-2} (2.2 % of the total current density); time= 10 min/cycle; total duration of the ZAB = 35 h	2
PAA + 11.25 M KOH + 0.25 M ZnO	Not provided (Pt/C + Ir/C)	1.3 V	~90	~50	80 cycles performed at 1 mA cm−2 (1.1 % of the total current density); time= 20 min/cycle; total duration of the ZAB= 26.7 h	3
Sodium polyacrylate (PANa) + cellulose soaked in 6 M KOH	Not provided (Polypyrrole-coated-graphene @ ZIF-8 to obtain Fe-embodied porous nitrogen-doped carbon)	1.48	~330	~200	220 cycles performed at 5 mA cm−2 (1.5 % of the total current density); time= 30 min/cycle; total duration of the ZAB= 110 h	4
Sodium polyacrylate + 0.5 g starch	2 (mixture of 50 % Pt/C and 50 % RuO_2/C)	1.40	~100	68	132 cycles performed at 1 mA cm−2 (1 % of the total current density); time= 20 min/cycle; total duration of the ZAB= 44 h	5
Agar + PVA + graphene oxide in 6 M KOH	1 ($ZnFe_2O_4$ spinel on carbon microspheres)	~1.1	~150	123.7	180 cycles performed at 0.5 mA cm−2 (0.33 % of the total current density); time= 10 min/cycle; total duration of the ZAB= ~30 h	6
Inulin + 6 M KOH	1 (Ni-Co-Mn trimetallic spinel)	1.44	112	62	1600 cycles performed at 0.16 mA cm^{-2} (0.15 % of the total current density); time= 240 s/cycle; total duration of the ZAB= 106.6 h	This work

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