Towards Smart and Sustainable Binders: The BIONIC Project on Chitosan-Based Self-Healing Materials for Lithium-Ion Batteries

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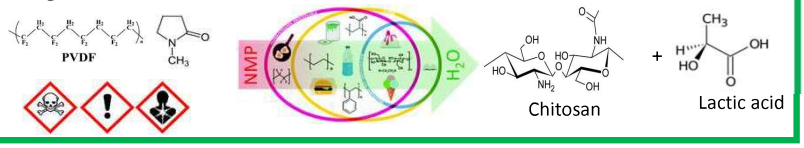
BIONIC – BInders with high iONIc Conductivity for fully sustainable Li-ion cells is a PRIN project aligned with the Battery2030+ European Research Initiative. The project aims to design **eco-friendly, low-cost polymeric binders** with **self-healing functionalities** and **ionic conductivity** for Li-ion battery electrodes.
These innovative materials will: enhance safety and stability of cells, improve energy density and cycle life, reduce costs and environmental impact by replacing PVdF/NMP with water-soluble green binders.
BIONIC integrates multi-scale computational modelling with advanced experimental characterizations to link structure–properties–performance and provide guidelines for the design of next-generation binders.

Motivation

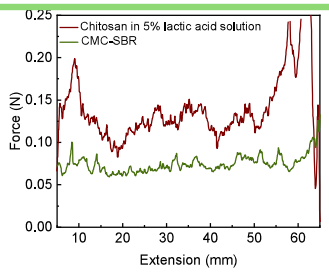
Current Li-ion cells still rely on PVdF/NMP binders, which are toxic, costly and unsustainable. Binders strongly influence electrode adhesion, SEI stability, safety and cycle life. Greener, water-processable and functional binders are crucial for safer and more sustainable batteries.

The BIONIC approach is to develop eco-friendly, low-cost polymeric binders with ionic conductivity and self-healing properties to replace PVdF/NMP with green, natural, water-based formulations based on **chitosan**



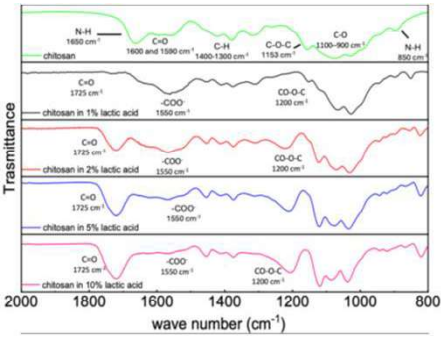
Experimental results

The experimental investigation demonstrated that chitosan, when solubilized in lactic acid, provides a remarkable improvement in electrode properties compared to conventional binders. Adhesion tests revealed a stronger interaction with the copper current collector than the benchmark CMC/SBR system, ensuring better mechanical stability of the coating under cycling.



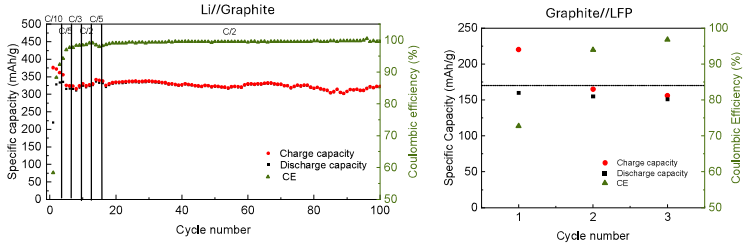
90° peel tests on graphite electrodes with chitosan binder show stronger adhesion to the copper collector compared to the CMC:SBR (2:1) benchmark, as evidenced by higher force values in the force–extension curve

FTIR spectroscopy confirmed the formation of ester bonds between chitosan and lactic acid, which are expected to reinforce the polymer network and contribute to enhanced structural integrity of the electrode.



FTIR spectra of chitosan films solubilized in lactic acid show a decrease of carboxyl bands (1550 cm^{-1}) and an increase of ester bands (1725 cm^{-1}) with rising acid concentration. This indicates that part of the lactic acid remains bound in the polymer matrix, enhancing interfacial adhesion. So, 5 wt% lactic acid solution was selected for electrode fabrication.

Half-cell tests showed stable cycling (~330 mAh g^{-1}, ~99% CE), comparable to state-of-the-art binders.
Preliminary full-cell tests (LFP cathode, graphite–chitosan anode) showed promising performance, with initial C/10 cycling delivering capacities close to the theoretical value, highlighting the promise of chitosan–lactic acid binders for sustainable Li-ion anodes



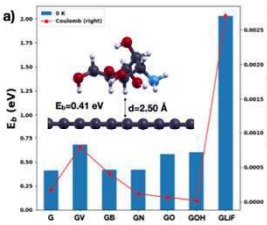
Binder: chitosan in 5% lactic acid solution
Graphite: binder: C45 (90: 7: 3)
electrolyte: LP30 + 10% FEC + 2% VC - Celgard 2325 separator

Modeling

The adhesion of chitosan and graphene can be calculated through the binding energy defined as:

$$E_b = -[E(G + molecule) - E(G) - E(molecule)]$$

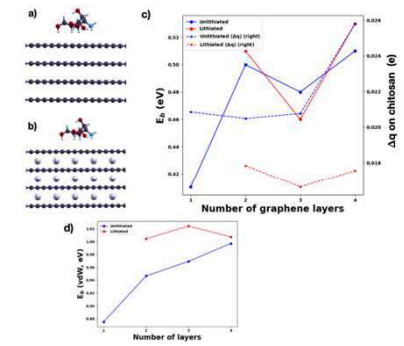
With E total energy. The calculations of E are based on DFT using a van der Waals exchange-correlation functional. The binding properties of the chitosan monomer on single and multi-layer, pure, defective, and lithiated graphene were studied.



b) Chitosan-graphene distance d and charge Δq (e) on chitosan

	d (Å)	$\Delta q(e)$
G	2.51	-0.021
GV	1.92	-0.039
GB	2.35	-0.031
GN	2.51	-0.017
GO	2.15	-0.012
GOH	2.02	+0.006
GLiF	2.35	-0.080

Binding energies of chitosan on pure graphene (G), graphene with carbon vacancies (GV), substitutional boron (GB), substitutional nitrogen (GN), adsorbed oxygen (GO), adsorbed OH (GOH), adsorbed LiF (GLiF). d is the equilibrium chitosan-grapgene distance and Δq is the transferred electronic charge from graphene to chitosan (i.e. 0.021 e is transferred from G to chitosan)



(c) Calculated binding energies of chitosan to multilayer graphene (figure (a)) and on fully lithiated multilayer graphene (figure (b)) and corresponding Bader charge transfers.
(d) Contribution of the van der Walls interaction to the binding energies as a function of the number of graphene layers.

CONCLUSIONS

- Chitosan–lactic acid binder ensure strong adhesion and structural integrity, outperforming conventional systems.
- Electrochemical tests show stable cycling (~330 mAh g^{-1}, 99% CE) and promising full-cell performance with LFP.
- Modelling (MD, ReaxFF, DFT) will complement experiments, clarifying adhesion, ionic transport and self-healing mechanisms.
- BIONIC paves the way for sustainable, water-processable binders aligned with the Battery2030+ vision.

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