

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

Rossella Petruzzelli¹, Burak Ozdemir², Giampaolo Lacarbonara¹, Daniele Versaci³, Carlotta Francia³, Matteo Gandolfo³, Catia Arbizzani¹, Rita Magri^{2,4}, Silvia Bodoardo³

1. Università di Bologna, Dipartimento di Chimica "Giacomo Ciamician" via P. Gobetti 85, 40129 Bologna
2. Dipartimento di Scienze Fisiche, Informatiche e Matematiche, Università di Modena e Reggio Emilia, Via Campi 213/A, 41125 Modena, Italy
3. Dipartimento di Scienza Applicata e Tecnologia – Politecnico di Torino – Corso Duca degli Abruzzi 24- 10129 Torino
4. Centro S3, Istituto Nanoscienze-Consiglio Nazionale delle Ricerche (CNR-NANO), Modena, Italy

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.

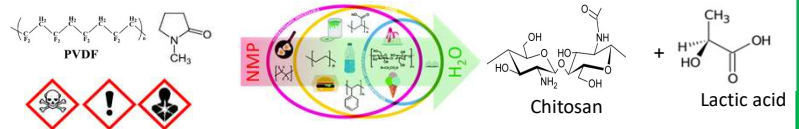
These innovative materials will: enhance safety and stability of cells, improve energy density and cycle life, reduce costs and environmental impact by replacing PVdF/NMP with water-soluble green binders.

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

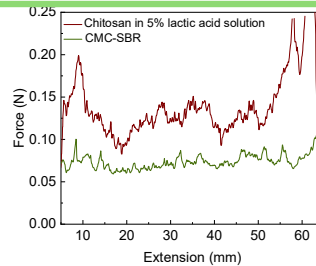
Current Li-ion cells still rely on PVdF/NMP binders, which are toxic, costly and unsustainable. Binders strongly influence electrode adhesion, SEI stability, safety and cycle life. Greener, water-processable and functional binders are crucial for safer and more sustainable batteries.

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**



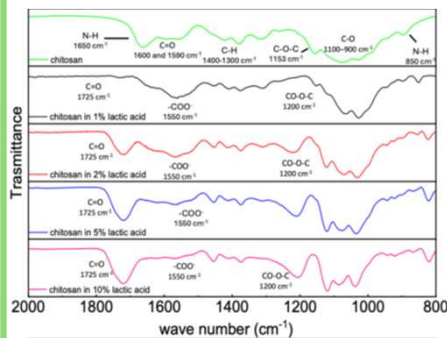
Experimental results

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.



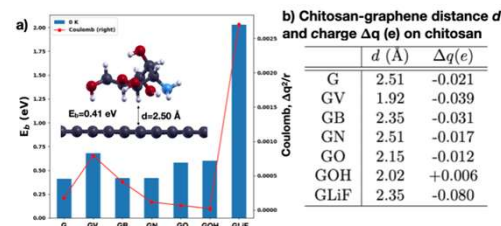
FTIR spectra of chitosan films solubilized in lactic acid show a decrease of carboxyl bands (1550 cm⁻¹) and an increase of ester bands (1725 cm⁻¹) with rising acid concentration. This indicates that part of the lactic acid remains bound in the polymer matrix, enhancing interfacial adhesion. So, 5 wt%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

Modeling

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

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

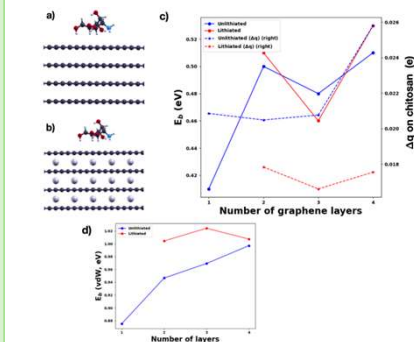
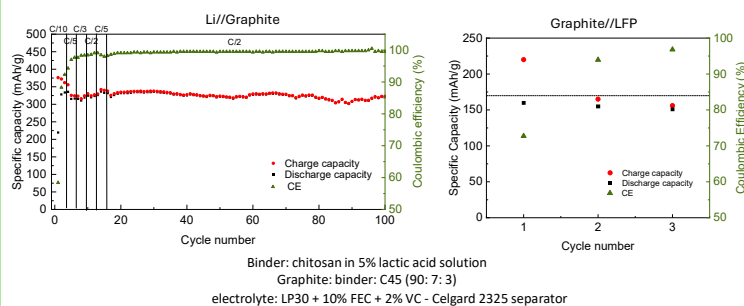
With E total energy. The calculations of E are based on DFT using a van der Waals exchange-correlation functional. The binding properties of the chitosan monomer on single and multi-layer, pure, defective, and lithiated graphene were studied.



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-graphene distance and Δq is the transferred electronic charge from graphene to chitosan (i.e. 0.021 e is transferred from G to chitosan)

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



(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 Waals 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.