Towards Eco-Friendly Lithium-Ion Batteries: Optimizing Chitosan as a Binder for Graphite Anodes

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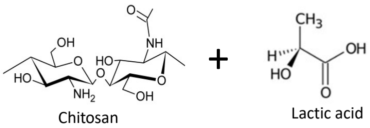
1 INTRODUCTION

Lithium-ion batteries have significantly improved in performance in recent years. However, all components must be optimized to achieve further advancements, not only in performance but also in sustainability and environmental impact [1]. One crucial component of the electrodes is the binder, which ensures adhesion and guarantees mechanical stability. Therefore, it is essential to explore new environmentally friendly and water-processable binders to reduce production costs and enhance environmental and worker safety during cell manufacturing [2].

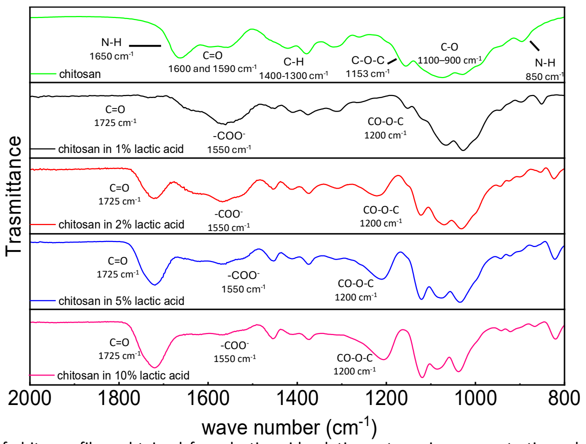
This work focuses on the design and development of a new eco-friendly and cost-effective polymeric binder for graphite anodes: chitosan. Chitosan is a polymer derived from the partial N-deacetylation of chitin, which is abundant in nature. To enable its processing, chitosan must be solubilized in an acidic aqueous solution, which typically employ acetic acid [3]. Different carboxylic acids were evaluated for use in the chitosan processing, with the aim of introducing a functional component to enhance the mechanical and electrochemical performance of the electrode.

2 CHITOSAN SOLUBILITY TEST



Chitosan is not soluble in water, so the binder must be prepared in an acidic environment to ensure complete solubilization of the polymer. Carboxylic acids are used to prepare an acidic solution. In addition to ensuring solubilization of the polymer, these acids could lead to the formation of hydrogen bonds with the hydroxyl and amino groups of chitosan. Chitosan was solubilized in lactic acid at various concentrations to understand the type of bonds formed and then study the effects on the mechanical properties of the electrode. To our knowledge, it is the first time that lactic acid is used in combination with chitosan as an anodic binder.

3 FTIR TEST

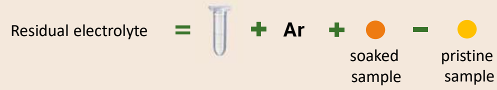


FTIR spectra of chitosan films obtained from lactic acid solutions at varying concentrations show that the increase of the acid concentration leads to a reduction in the absorption bands associated with the carboxyl group of the acid (1550 cm^{-1}), while the intensity of the band attributed to ester stretching vibrations increases at 1725 cm^{-1}. This suggests that a fraction of the lactic acid remains bound within the polymer matrix [4]. Such chemical interactions are expected to enhance the interfacial adhesion between the graphite electrode and the current collector. Based on these results, chitosan solubilized in 5 wt% lactic acid was selected as the binder for subsequent physicochemical characterization and electrode fabrication.

4 ELECTROLYTE UPTAKE

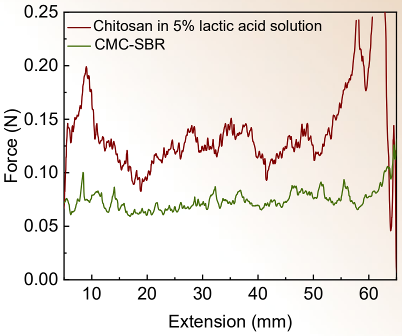
For electrolyte uptake measurement chitosan was solubilized in 5 wt% lactic acid solution, then coated on copper foil (14 mil). After drying in the desiccator at room temperature, eight discs (diam = 9 mm) were punched out. The average thickness of the samples was 20 µm.

The average weight of the electrolyte retained is 0.97 mg, the average retained electrolyte % is 127. This indicates that the binder displays a good affinity with the electrolyte and improves the wettability of the electrode, without retaining it too much, which could cause premature depletion of the electrolyte.



Sample	Sample (mg)	Vial + Ar (g)	Vial + Ar + soaked sample (g)	Electrolyte retained (mg)	Average weight of the electrolyte retained (mg)	Electrolyte retained %
1	7.09	1.05367	1.06176	1.00	0.97	127
2	7.06	1.08031	1.08833	0.96		
3	7.01	1.07388	1.08193	1.04		
4	6.85	1.03968	1.04717	0.64		
5	6.66	1.06172	1.06945	0.75		
6	7.12	1.06204	1.07098	1.82		
7	6.83	1.04529	1.05336	1.24		
8	7.01	1.08061	1.08837	0.75		

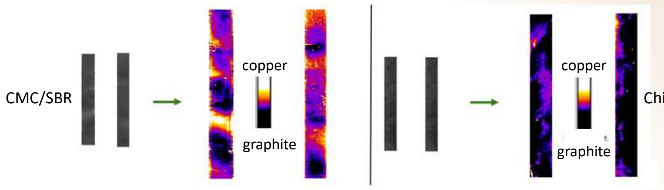
5 T-PEEL TEST



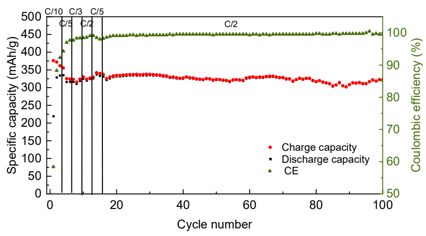
90° Peel tests of single-coated tapes, was conducted to evaluate the adhesion of graphite containing chitosan to the copper collector [5] . Graphite containing CMC:SBR (2:1) as the binder was used as a benchmark.

Graphite containing chitosan as binder shows an adhesion higher than that of the reference graphite, as shown by the graph of the force (N) versus the extension.

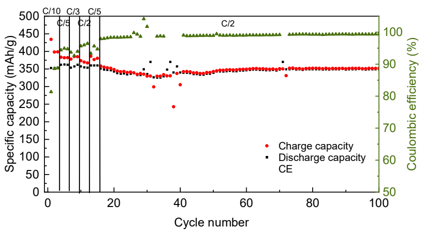
Using the program 'ImageJ', pre- and post-measurement samples were graphically processed to evaluate the amount of graphite remaining attached to the current collector. In the colour scheme, white represents copper and black represents graphite; intermediate colours highlight the presence of both. The images show that less copper is exposed in the graphite containing chitosan, indicating greater adhesion of the coating to the substrate.



6 ELECTROCHEMICAL TEST: RATE CAPABILITY AND STABILITY TEST



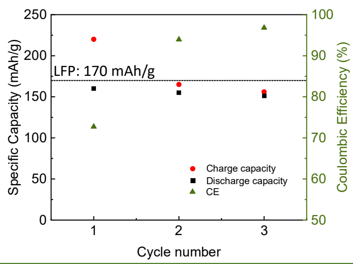
Binder: chitosan in 5% lactic acid solution
Graphite: binder: C45 (90: 7: 3)



Benchmark
Binder = CMC : SBR: (2:1)

The electrochemical tests were carried out in an EL-CELL, featuring the graphite of interest as the working electrode and metallic lithium as the counter electrode. The electrolyte consisted of LP30 with the addition of 10% fluoroethylene carbonate (FEC) and 2% vinylene carbonate (VC) as additives. A Whatman GFA glass fiber separator, 260 µm in thickness, was utilized. The results indicate that the graphite maintains, over the first 100 cycles, a stable discharge capacity of approximately 330 mAh g^{-1} and a coulombic efficiency of 99%. These values are comparable to those of the benchmark electrode, which exhibits a discharge capacity of 350 mAh g^{-1} within the first 100 cycles

7 PRELIMINARY ELECTROCHEMICAL TEST IN FULL CELL



Preliminary electrochemical test was carried out in full-cell configuration, employing $LiFePO_4$ (LFP) as the cathode and the graphite-based anode formulated with chitosan as the binder. The electrolyte consisted of LP30 + 10% fluoroethylene carbonate (FEC) + 2% vinylene carbonate (VC), while a Celgard 2325 membrane was used as the separator. The initial galvanostatic cycling at C/10 over the first three cycles exhibited promising performance, with the measured specific capacity approaching the theoretical value.

8 CONCLUSION

Chitosan, a biopolymer widely employed in green chemistry, has demonstrated promising applicability as a binder material. When solubilized in lactic acid, FTIR analysis revealed the formation of ester bonds between chitosan and lactic acid, which are expected to enhance the mechanical integrity of graphite electrodes. Mechanical adhesion tests (T-peel) confirmed excellent adhesion and cohesion within the graphite structure. Furthermore, electrochemical evaluations showed performance comparable to that of conventional CMC/SBR binders, which are currently standard in commercial graphite anodes. These findings highlight chitosan–lactic acid systems as viable green alternatives for binder applications in lithium-ion batteries.

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