Valorization of olive leaves extract in Natural Deep Eutectic Solvents for the development of bioactive chitosan films and hydrogels

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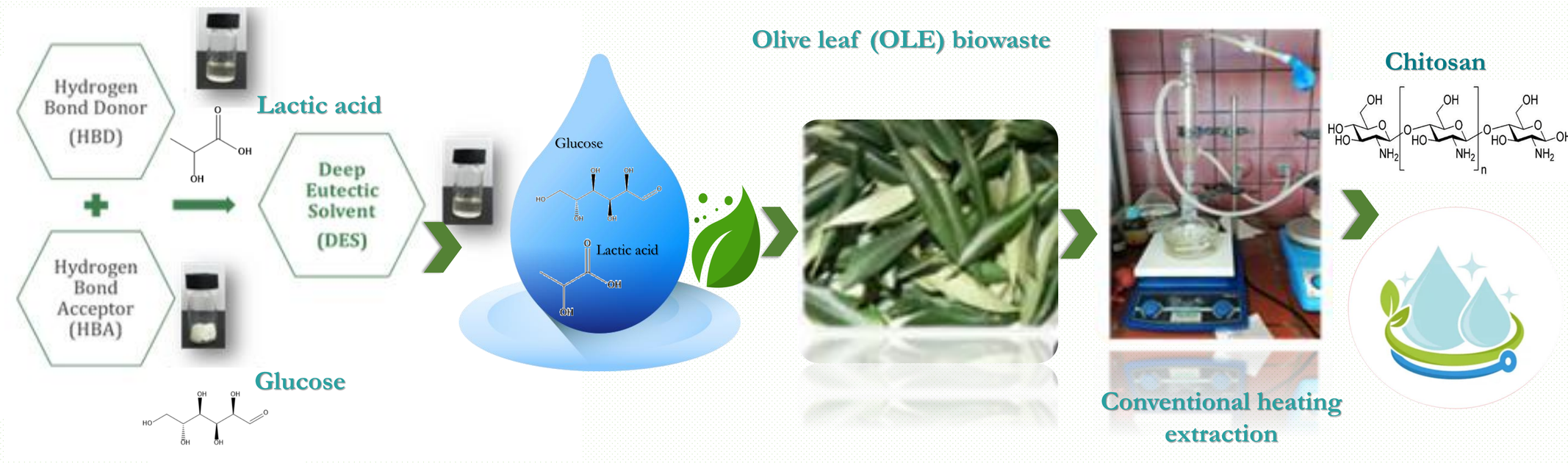
Introduction

Chitosan is a copolymer of N-acetyl-D-glucosamine and D-glucosamine, a partially deacetylated derivative of chitin from the exoskeletons of crustacean shellfish, such as shrimps and crabs. It is non-toxic, biodegradable, and biocompatible with the human digestive system. **Deep Eutectic Solvents (DES)** are mixtures of two or more components, a hydrogen bond acceptor and a hydrogen bond donor, with a low-temperature eutectic point. When the components of the DES are naturally occurring compounds, the solvents are characterized as **Natural Deep Eutectic Solvents (NADES)**. **Maillard reaction** is a spontaneous and non-enzymatic browning reaction between the amine groups of amino acids and the carbonyl group of reducing sugars. In the present work, a greener approach towards the formation of **novel films** and **hydrogels** based on chitosan and Maillard reaction products using **Glucose/Lactic Acid/Water NADES-olive leaf extract**. In an effort to investigate the ability of olive leaf extract (OLE), obtained using NADES as the extraction and storage medium, to act as dissolution and gelating agent.

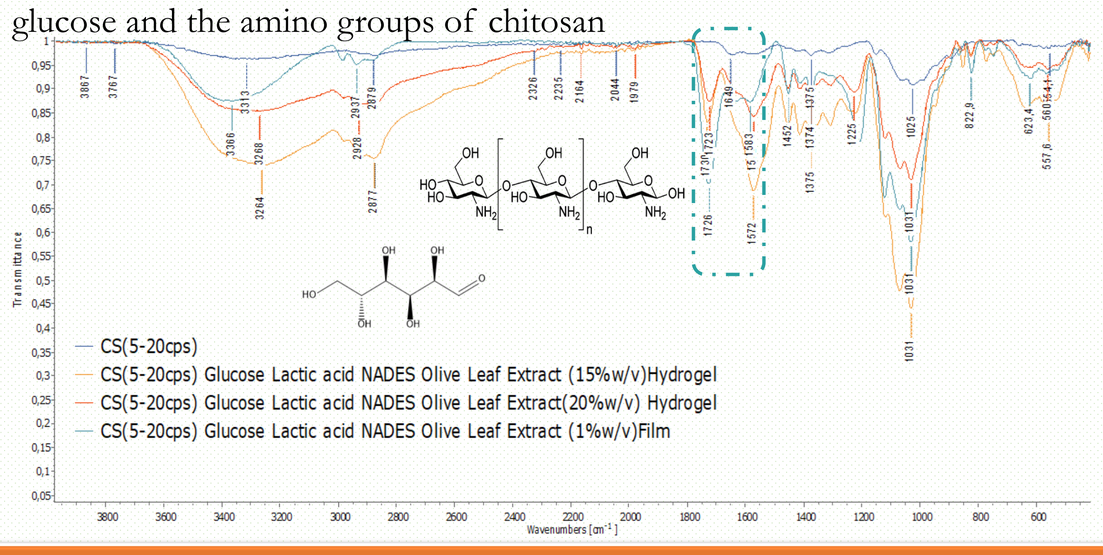
Material & Methods



Results & Discussion

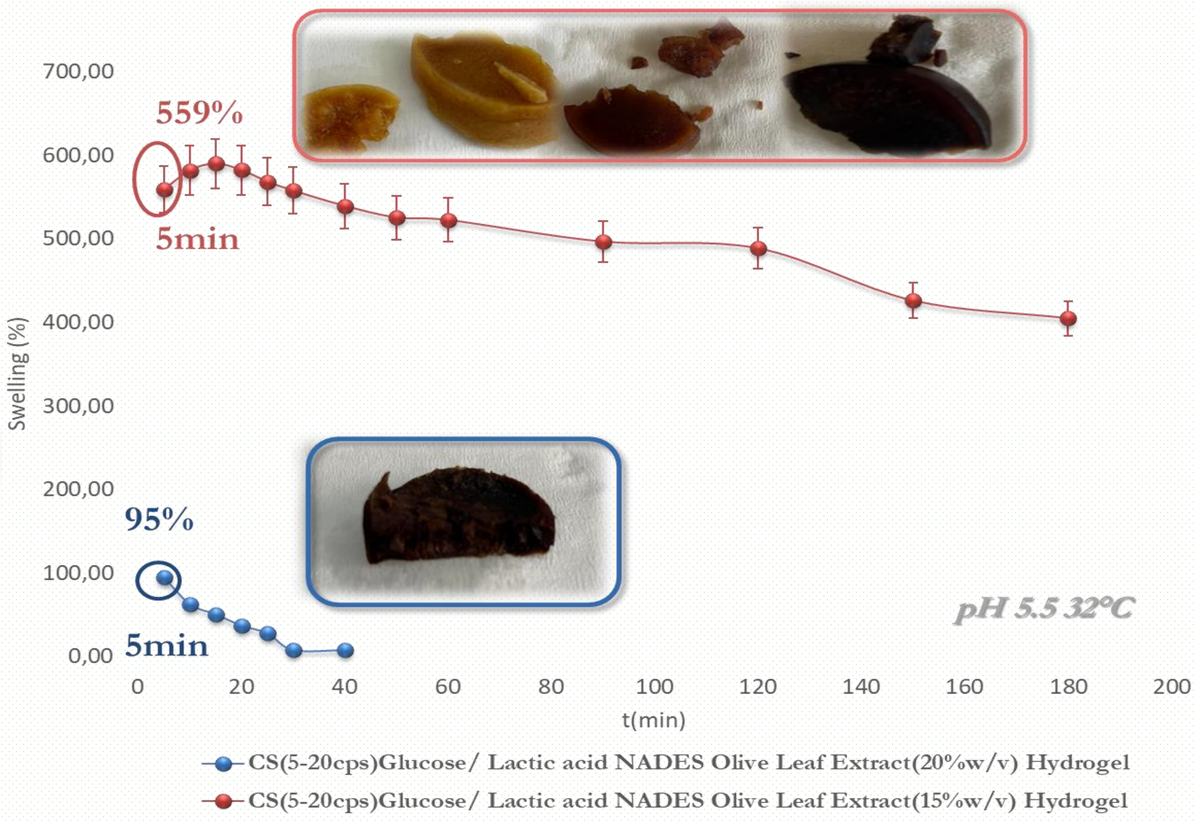
Structural characterization

- Characteristic absorption bands of chitosan observed at around **1649** and **1584 cm^{-1}** were assigned to amide I and primary amino groups, respectively
- After the reaction, it can be observed that these absorption bands of chitosan showed changes
- The absorption peak at **1649 cm^{-1}** decreased and shifted to **1572 cm^{-1}** suggesting that **Schiff base (C=N double bond)** was formed between the reducing termination of glucose and the amino groups of chitosan



Swelling studies of chitosan hydrogels

- Swelling ratio of the chitosan-based Hydrogels-NADES-OLE in aqueous solution increased rapidly in the first 5 min reaching 559%
- The water retention ratio remained over 70%, after keeping the swollen sample in phosphate buffer solution at pH=5.5 for 3 hours



In Vitro Evaluation of Antioxidant Activity

Sample	Inhibition of Linoleic Acid Peroxidation (%)	Trolox Equivalent Antioxidant Capacity/AAPH Assay (μg/mL)
CS(5-20cps) Glucose Lactic acid NADES Olive Leaf Extract (15% w/v) Hydrogel	55.47±4.90	15.46
CS(5-20cps) Glucose Lactic acid NADES Olive Leaf Extract (20% w/v) Hydrogel	53.08±7.10	14.80
CS(5-20cps)Glucose Lactic acid NADES Olive Leaf Extract (1%w/v) Film	59.78±5.10	16.67

Note:% lipid peroxidation inhibition: 89.73 ± 0.90 (concentration of Trolox solution 25.02 μg/mL)

Conclusions

- Olive leaf (OLE) biowaste extracted using the Glucose/Lactic Acid/Water NADES
- The NADES and the NADES-OLE extract acted as efficient cross-linking agents and plasticizers for the production of the chitosan films and hydrogels
- FT-IR confirmed the Maillard reaction of glucose (derived from the NADEs system) and chitosan due to the formation of a Schiff base
- The Maillard reaction products, improved the antioxidant activity of chitosan films and hydrogels
- The obtained films and hydrogels obtained such biological and mechanical properties turning them into good candidates for a variety of applications, especially in those concerning the food industry.

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