SYNTHESIS OF LIGNIN NANOCAPSULES FOR INCORPORATION OF ESSENTIAL OILS

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ABSTRACT

The use of renewable resources in the development of sustainable nanomaterials is on the rise. Lignin and essential oils (EOs) are natural compounds that have several properties of interest, such as antioxidant, antifungal, and antibacterial activities. Therefore, the present study proposes the use of lignin and EO as raw materials to obtain sustainable functional nanomaterials. For this, lignin extracted from persimmon tree pruning waste (PTPW) and EO from cinnamon bark (*Cinnamomum verrum*) were used. Nanoparticles were obtained from antisolvent precipitation, with an average diameter of 320 to 630 nm, a low polydispersity index (PDI < 0.2), and a zeta potential below -30 mV, with encapsulation efficiency in the range of 77 to 90% (*v/v*). The results suggest the technical feasibility of encapsulating EO in lignin-based nanostructures, being a biodegradable and low-cost alternative for obtaining nanometric structures for future applications, e.g., nanoactives in cosmetic and therapeutic formulations.

Keywords: Biomaterials. Lignin. Nanocapsules. Essential oils.

1 INTRODUCTION

The pharmaceutical industries are constantly moving towards the use of renewable resources focused on the development of sustainable materials. Food derivatives and agricultural waste, such as lignin, have gained more attention due to their abundance, biocompatibility, and interesting biological characteristics. This aromatic macromolecule, found in the cell walls of terrestrial plants, has thermal stability, ultraviolet ray absorption capacity, antioxidant, and antimicrobial activity.[1,2].

Other natural compounds of interest are essential oils (EOs), which are made up of various natural active ingredients that can be extracted from medicinal, aromatic, spicy, and/or forest plants. They are volatile substances that evaporate when exposed to air at room temperature and pressure and tend to oxidize when in contact with sunlight and heat. The extracted compounds have numerous properties, such as: antioxidants, antimicrobials, antifungals, insecticides, anti-inflammatories, etc.[3,4,5]. Although EOs have important properties, their constituents are highly volatile, making the encapsulation of EOs a relevant alternative for preserving their biological activity, reducing their oxidation and volatilization.[6].

For this study, lignin extracted from PTPW and EO from cinnamon bark (*Cinnamomum verrum*) were used, whose main compound in this EO is cinnamaldehyde (C_9H_8O) - a fast-acting antifungal and antimicrobial agent, capable of inhibiting, in low concentrations, the growth of *Pseudomonas aeruginosa* present in the skin microbiota.[7,8].

2 MATERIAL & METHODS

For the synthesis of lignin nanocapsules for incorporation of EOs, 50 mg of freeze-dried lignin from PTPW was dissolved in 10 mL of a mixture of solvents (acetone (C_3H_6O), ethanol (C_2H_6O) and deionized water (H_2O)), obtaining a concentration of 5 mg.mL^{-1}. The solution was placed on a magnetic stirrer at 600 rpm for 10 min and then filtered through a 0.45 µm syringe filter to remove undissolved solids. 10, 30, and 50 µL of cinnamon EO were added for each 1 mL of the previously prepared lignin solution, obtaining final concentrations of 1, 3 and 5% (*v/v*), respectively. These final lignin and EOs solutions were mixed on a magnetic stirrer for 40 min.[9].

Deionized water (antisolvent) was then added to the lignin and EO solution in a fixed ratio of 9:1, with a flow rate of 250 µL.s^{-1} using a peristaltic pump. This system was maintained under constant stirring at 600 rpm until the total volume of water was added. The overall process is illustrated in Figure 1.[9].

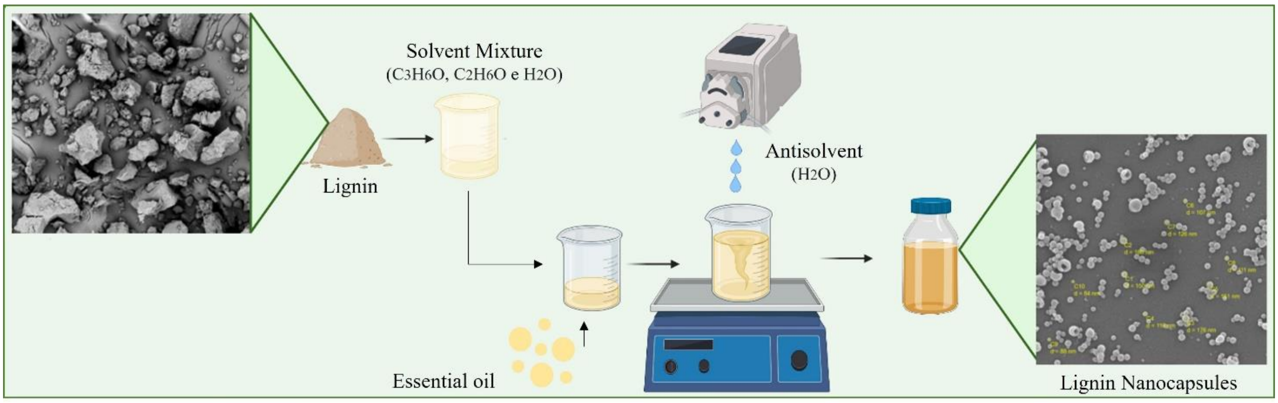

Figure 1 Diagram showing the main stages in the production of lignin nanocapsules with essential oil (Adapted from LONGO, MARCHEZAN & CAMASSOLA, 2023).

Encapsulation efficiency was determined using a Field Emission Scanning Electron Microscope (FE-SEM). The average diameter, polydispersity index (PDI), and zeta potential of suspended particles were analyzed on a Zetasizer Advance Series Pro (Blue Label).

3 RESULTS & DISCUSSION

Firstly, SEM analysis was carried out, obtaining micrographs of the lignin extracted from PTPW and its nanoparticles prepared from the best antisolvent precipitation condition, without the incorporation of cinnamon EO. Next, the SEM analysis was performed, now with the incorporation of the EO into the lignin nanoparticles already formed, as illustrated in Figure 2.

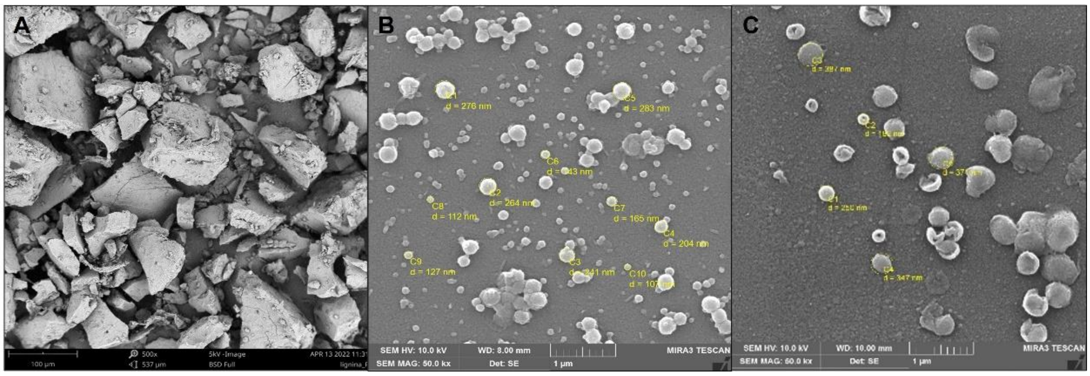

Figure 2 Micrographs of lignin extracted from persimmon tree pruning waste (A); nanoparticles formed (B); lignin nanocapsules with cinnamon EO (C).

Figure 2 shows that the lignin nanoparticles extracted from PTPW have a spherical shape, including small clusters (B). The nanostructures incorporated with cinnamon EO showed spherical and almost spherical nanostructures (C).

The characterization analysis of lignin nanocapsules with incorporation of EO in the matrix was then carried out using the Zetasizer equipment. The data obtained are shown in Table 1.

Table 1 Characterization of lignin nanocapsules extracted from persimmon tree pruning waste with the incorporation of EO.

Initial EO Charge (%, *v/v*)	Mean Hydrodynamic Diameter (nm)	Polydispersity Index (PDI)	Zeta Potential (mV)	Encapsulation Efficiency (%, *v/v*)
1	324.5 ± 1.9	0.07 ± 0.01	-49.6 ± 1.8	77.3 ± 0.3
3	429.2 ± 1.0	0.13 ± 0.07	-50.6 ± 3.9	85.6 ± 3.2
5	630.8 ± 31.6	0.11 ± 0.04	-51.0 ± 2.5	89.9 ± 3.4

The nanoparticles produced had an average diameter in the range of 320 to 630 nm, and a low PDI (PDI < 0.2) was obtained, indicating the formation of a homogeneous system with uniform particle size dispersion.[11] For the zeta potential, a value below -30 mV was obtained, indicating that the particles presented colloidal stability in the suspension.[12].

In terms of encapsulation efficiency, the values were between ~77 and ~90% (*v/v*). Thus, it was observed that the EOs were effectively encapsulated by lignin-based nanostructures. Similarly, ZIKELI et al. 2020 also successfully used EOs trapped in lignin nanocapsules. It was observed that this procedure was satisfactory since the release of encapsulated EOs from common thyme and wild thyme was slower compared to the release of EO that was not subjected to the nanoencapsulation process.[9].

4 CONCLUSION

The data obtained show that it is technically viable to use lignin as a biodegradable and low-cost source for obtaining nanometric structures with EO to be used in future applications. The encapsulation efficiency of up to 90% also provides robustness to the process developed in the present work.

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