Preserving Heritage Textiles - Developing Novel Preservative Fabric for Long-Term Conservation

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Abstract:

Textiles are very delicate and hard to preserve, especially in tropical countries like India, due to the ease with which they can be damaged and the rate of deterioration. Natural fabrics such as cotton, linen, jute, leather, silk, wool, and others, being made up of cellulose and proteins are attractive to microbes, resulting in biological deterioration. Some natural compounds, such as essential oils, can act as biocides, like Azadirachta Indica (commonly known as neem) which has a broad-spectrum antimicrobial action. This study aims to create fabric that will preserve historical textiles when used as a wrapping material in museum storages, lining, backing, and covering material for shelving, drawers, and boxes, and as padding for hangers and rollers used for exhibits. For that purpose, neem essential oil nanoparticles of an average size of 189 nm with 78% entrapment efficiency (EE) and 8.83% loading capacity (LC) were formulated using nano-emulsion and ionic gelation technique and applied to cotton and polyester fabric to give them antibacterial properties. SEM analysis was also performed to understand the surface structure of the nanoparticles. Additionally, stability of the nanoparticle coated fabric over a time period of two months in different storage conditions was also tested. The study also compares the efficiency of the finish applied on cotton and polyester fabric.

Keywords: *Deterioration, Essential oil, Heritage textiles, Nanoparticles, Preservative fabric*

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1. Introduction

Textiles are integral to the lives and customs of people from all cultures, and a museum collection typically displays objects that document the past and present of human life, cultural values, and artistic creations. Many traditional textiles are made from natural fibers, which make them vulnerable to degradation due to a range of biological, chemical and physical factors, such as light, temperature and humidity levels, dust, mishandling, and improper restoration methods [1]. This is especially true for cellulose, silk, and wool fibers, as they provide a favorable environment for microorganisms, insects, and other living organisms. These variables often result in discoloration, brittleness, corrosion of metallic components, a sweet or musty odor, and small irregular holes, which accelerate aging and result in irreversible damage [1,2]. In order to protect textiles from damage, conservators and curators utilize natural and synthetic compounds as biocides and insect repellents. In India, a traditional practice for preserving textiles involves using herbs and spices like clove, cinnamon, carom seeds, camphor, neem leaves, tobacco, tulsi, and eucalyptus [3,4,5]. Although these substances have biocidal and insect repellent properties due to the presence of active compounds, they do not create an extensive protective environment over a large circumference area. Additionally, these compounds are sensitive to light and cannot be applied directly to fabrics or surfaces. Nano

encapsulation of essential oils is a potential solution to this issue, as it allows for controlled release of the active compounds and protects them from oxidation and UV degradation [6]. In encapsulation, chitosan, a polysaccharide derived from chitin has been used widely as a wall material.A cross-linking agent is usually employed as a connection between the ionic polymer and the ion with an opposite charge to create nanoparticles.

This research is conducted with the goal of forming chitosan nanoparticles with neem essential oil, utilizing tripolyphosphate (TPP) for cross-linking. This finish is then coated on a separate fabric for the purpose of wrapping, or using as a lining, padding, or to cover the heritage textiles. This comprehensive approach of having antimicrobial agents leach out of the fabric is necessary for slowing down the degradation process and preserving the distinctive characteristics of heritage textiles and making sure they remain intact for future generations.

2. Materials and Methods

2.1 Materials

Neem oil was purchased from Sigma Aldrich Co. Chitosan well known for its use in encapsulating essential oils and its antimicrobial and mucoadhesive properties, was chosen as the wall material/shell due to its great matrix capabilities. A medium molecular weight chitosan of 84.8% degree of deacetylation purchased from HIMEDIA was selected for the study. Tripolyphosphate (TPP), Tween 80, Glacial acetic acid and solvents such as Dichloromethane, Methanol were also purchased from Sigma Aldrich Co.

2.2 Preparation of neem essential oil chitosan nanoparticles

A two-step process of Nano-emulsion and Ionic gelation was used to make nanoparticles with an essential oil core and a chitosan polymeric shell. Figure 1 outlines the procedure. Dissolving chitosan in 1% acetic acid (v/v) and stirring the solution on a magnetic stirrer for eight hours at 25℃ ensured complete dissolution. Afterward, Tween 80 surfactant with different concentrations (0.5, 0.75, 1, and 2%) was added to the chitosan solution and was left to stir for one hour. Different chitosan/essential oil ratios (1:1, 1:2, and 1:3) were then prepared in 10 ml of dichloromethane by adding the oil and solvent solution to the chitosan surfactant solution drop by drop using a syringe. This mixture was stirred on a high shear homogenizer at 13000RPM for 10 minutes and then agitated on a magnetic stirrer at 1800 RPM for two hours until the solvent evaporated completely. Subsequently, different concentrations (1, 2, and 3 %) of tripolyphosphate (TPP) were added to the solution drop wise using a syringe and stirred for one hour.

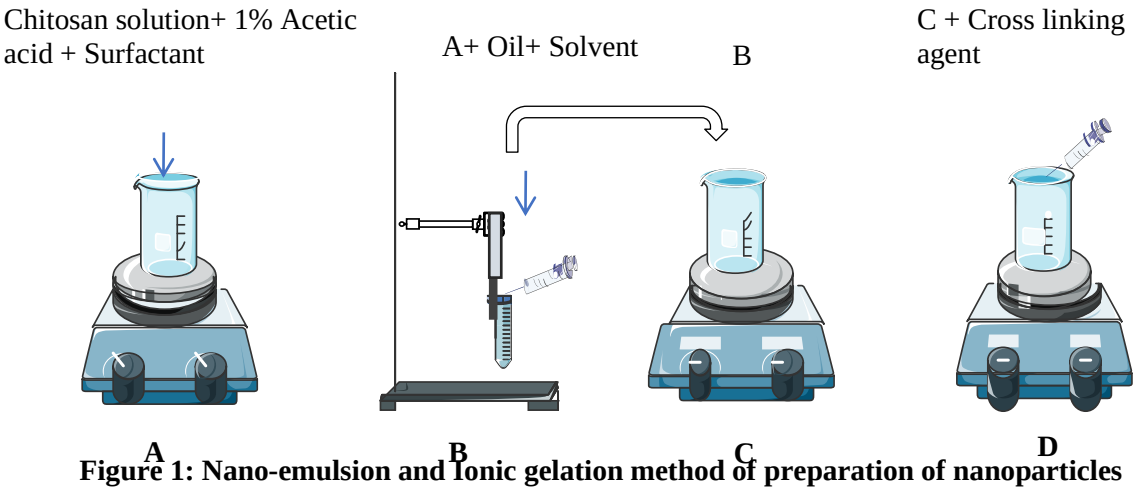

Figure 1: Nano-emulsion and Ionic gelation method of preparation of nanoparticles

2.3 Characterization of nanoparticles:

For determiningthe entrapment efficiency and loading capacity, in 100 µL of freshly prepared essential oil loaded chitosan nanoparticles, 900 µL of the solvent suitable for the essential oil was added to make up 1 ml. This solution was then centrifuged at 18000 rpm for 30 min. The supernatant was separated to estimate drug loading efficiency. Further, the free amount of oil was calculated by measuring the absorbance of all the essential oils at its significant wavelength (nm) using UV–vis spectrophotometer. This was then compared with their standard curve. The drug encapsulation efficiency and drug loading capacity were calculated using the formula below:

$$Entrapment\ efficiency\ (EE) = \frac{Total\ oil - Free\ oil}{Total\ oil}\ X\ 100$$

$$Loading\ capacity\ (LC) = \frac{Total\ oil - Free\ oil}{Total\ content}\ X\ 100$$

100 μL of freshly prepared nanoparticles was centrifuged at 18000 rpm for 15 minutes. The supernatant was diluted with distilled water to a concentration of (1 mg/mL), and viewed under a Zetasizer by Malvern in order to assess the particle size through dynamic light scattering (DLS).The nanoparticles obtained at optimized conditions were lyophilised and observed under SEM to understand the surface structure. The Minimum Inhibitory Concentration (MIC) was then calculated using two-fold dilutions in a 96-well plate containing Mueller Hinton broth (MHB). A series of dilutions of the nanoparticles (125 ppm) were prepared in a 96-well plate using Mueller Hinton broth (MHB). Bacterial suspensions were created and diluted in MHB until a turbidity of 0.5 McFarland was achieved, then added to each well. The plates were then incubated at 37°C for 24 hours. The lowest concentration that prevented visible growth of the test strains were then applied to scoured cotton and polyester fabrics with a citric acid binder via spraying and padding. The antibacterial activity of the fabrics was assessed using the AATCC 147 Parallel streak method against *Bacillus cereus, Staphylococcus aureus, Pseudomonas, and Escherichia Coli.* The stability of the nanoparticles was tracked over a time period of two months in different storage conditions. 2X2 cm of the nanoparticle coated cotton fabric was evaluated for 1 and 2 months. One set of samples was kept in a closed environment in a large petri dish, while the other set was exposed to an open environment at 25 ± 2°C. The amount of oil released from the nanoparticle-coated samples was determined by soaking them in a suitable solvent for 24 hours, and the absorbance was calculated using a UV spectrophotometer at278 nm and compared to that of the initial sample.

3. Results and Discussion

Neem essential oil chitosan nanoparticles were prepared using nano-emulsion followed by ionic gelation method. Chitosan was selected as a wall material to encapsulate the essential oil because of its ability of being biodegradable, cationic charge and being muco-adhesive nature proving best candidate to encapsulate. Negative charge TPP was used as a cross-linking agent to form the nanoparticles. It is important for the nanoparticle system to be successful that it has high entrapment efficiency, which allows the drug to be administered at smaller or more effective doses while reducing the amount of matrix components [6]. Therefore, the effect of change in the amount of chitosan, neem essential oil, Tween 80, and TPPwas optimized on the basis of highest entrapment efficiency of the neem essential oil observed in the nanoparticles.

3.1 Effect of Chitosan concentration on entrapment efficiency

By varying the chitosan concentration between 0.5, 1, 1.5 and 2 %, and while keeping the surfactant and TPP constant at 0.75% and 1% respectively. The percent entrapment efficiency of the neem essential oil was seen to be highest at 1% concentration with 79.24 ± 2.34 (Figure 2A). There was an increase of 7% in entrapment when compared to 0.5 % concentration, suggesting that with the increase of polymer, more oil could be encapsulated in the emulsion. However, further increase of chitosan concentration led to decreased entrapment efficiency. This could be attributed to the increased viscosity of the solution caused by the higher concentration of polymer, which impeded the formation of the emulsion.Nguyen,G., and Le, X. (2021) noted a similar trend for Palmarosa essential oil encapsulated with chitosan; the efficiency of encapsulation increased from 22.8% to 34.0% when the concentration of chitosan ranged from 5.0 to 10.0 g/L, then decreased to 26.1% at 12.5 g/L [8].

3.2 Effect of % surfactant on the entrapment efficiency

Tween 80 was added at different concentrations (0.5, 0.75, 1, and 2%) to the chitosan solution. Tween 80 is a non-ionic surfactant that aids in the formation of uniform oil dispersion by reducing the interfacial tension between the oil and the aqueous phase. This allows for a greater degree of oil droplet formation and stabilization, resulting in improved entrapment efficiency.Sun, W., et al. in their research demonstrated that when tween 80 is added, it results in a minor increase in the effectiveness of the encapsulation [9]. This occurs because tween 80 is adsorbed onto the surface.As observed in figure 2B, the encapsulation efficiency had an initial growth from 71.55 % to 80.36% which could be because of the adsorption of Tween 80 which might have caused decrease in surface tension leading to high

entrapment efficiency. However, reduction to 64.61% and 54.38% at %1 and 2% surfactant concentration was observed with further increase of Tween 80. Formation of foam was noted by the researcher with the increase in 2% of Tween 80. This might have caused a barrier around the nanoparticles, leading to less entrapment efficiency.

3.3 Effect of polymer: oil ratio on entrapment efficiency

Oil concentration is one of the factors that affect the entrapment efficiency of oil-loaded nanoparticles. Three different concentration of neem essential oil was used to encapsulate with 1% chitosan concentration. the highest entrapment efficiency was observed at 1% oil concentration (78.65 ± 1.28), according to the figure 2C. As the oil concentration increased to 2% and 3%, the entrapment efficiency drastically decreased to 69.36 ± 2.54 and 62.12 ± 2.71, respectively. This decrease in efficiency at higher oil concentrations can be attributed to the oil droplets becoming more difficult to encapsulate within the nanoparticle matrix. With higher oil concentrations, the likelihood of oil droplets coalescing or merging increases, leading to larger droplets that are more challenging to encapsulate. This ultimately reduces the overall efficiency of the encapsulation process, resulting in lower entrapment efficiency. Several studies have investigated the effect of oil concentration on the entrapment efficiency of oil-loaded nanoparticles. For example, a study by Wong et al. (2010) investigated the effect of oil concentration on the EE of poly(lactic-co-glycolic acid) (PLGA) nanoparticles loaded with curcumin. They found that the EE decreased from 75% to 50% as the oil concentration increased from 0.5% to 2.5% (w/v) [11].

3.4 Impact of % TPP on the entrapment efficiency

Three concentrations of TPP (1,2, and 3%) was added as a crosslinking agent to the chitosan neem oil solution. The entrapment efficiency of 78.65% was highest when 1% of TPP solution was added as a crosslinking agent to the chitosan neem oil solution (Figure 2D). This could have been caused by the presence of a greater amount of cross-link bonds, which enhanced the amount of oil inside the chitosan capsules. As the concentration of TPP was increased to 2 and 3%, the encapsulation efficiency decreased to 69.36% and 62.12% respectively. This could be due to the formation of a more compact and cohesive structure, leading to a reduction in the size of the pores [8].

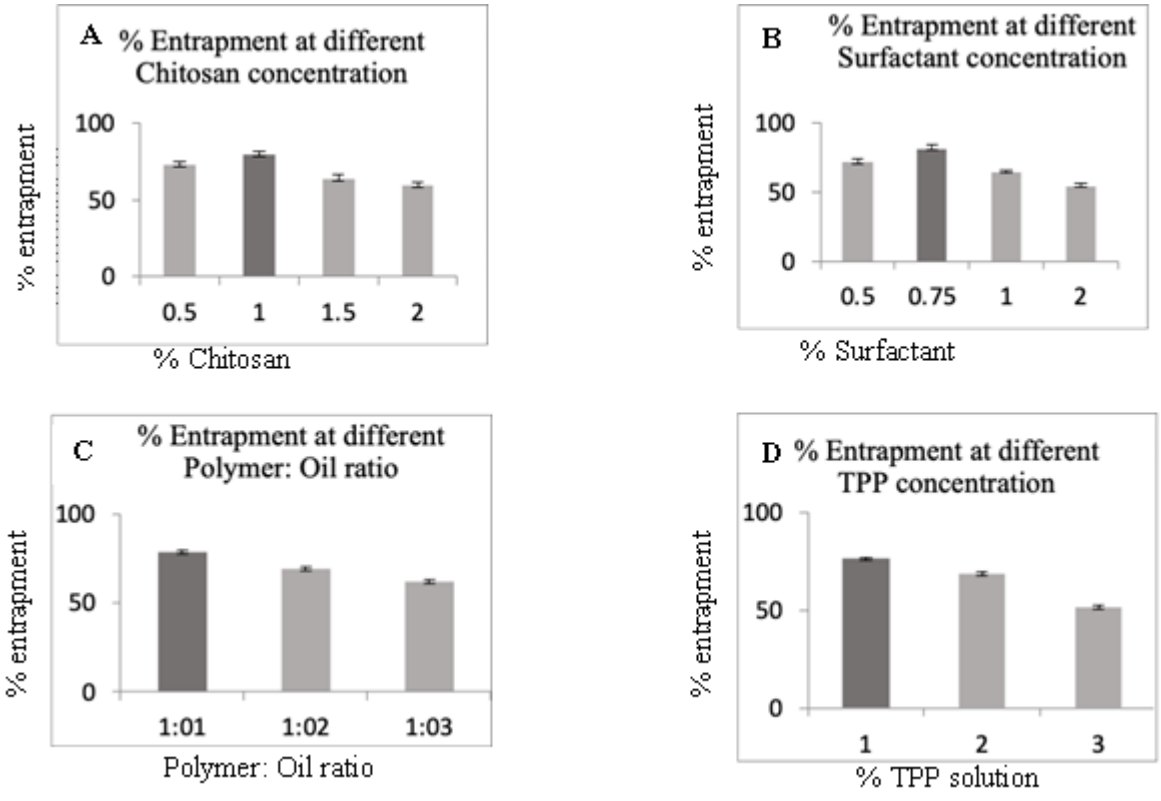

Figure 2: Impact of different parameters on entrapment efficiency of neem essential oil (Fig. 2A: %entrapment efficiency at different chitosan concentration, Fig.2B: % entrapment efficiency at different surfactant concentration, Fig. 2C: % entrapment efficiency at different oil concentration, and Fig.2D: % entrapment efficiency at different TPP concentration)

3.5 Characterization of the neem essential oil chitosan nanoparticles

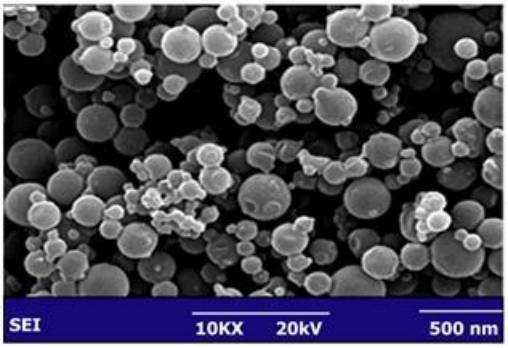

Figure 3: SEM image of Neem oil chitosan nanoparticles

The neem essential oil chitosan nanoparticles obtained at the optimized conditions when observed under scanning electron microscopeshowed spherical structures, absence of cracks and formation of a continuous wall layer (Figure 3). The average size of the nanoparticles was observed as 189 ± 26 nm.The average polydispersity index (PDI) detected was 0.2±3 using zetasizer, suggesting that the majority of the nanoparticles in the samples are relatively uniform in size

Whereas, the loading capacity was8.83±1.24.Table 1 represents values that are the mean of triplicate measurements of the entire test performed.

Table 1: Characteristic features of neem essential oil nanoparticle at optimized conditions

Essential oil Nanoparticles				**Results**						
Polymer: Oil Concentration	Surfactant (Tween 80) %	TPP %	RPM	Size (nm)	% EE	% LC	Antimicrobial Activity in Zone of inhibition- (mm) for cotton fabric			
							BC	SA	EC	PD
1:1	0.75	1	13000	189 ± 26	78.42 ± 1.56	8.83± 1.24	2.25 ±0.5	1.45± 0.39	0.57± 0.32	0.65± 0.3

3.6 Antibacterial assessment

The test bacteria used in this study includes two Gram-positive bacteria *Bacillus cereus*(BC), and *Staphylococcus aureus (SA) and Gram-negative bacteria Escherichia coli (EC), and Pseudomonas (PD).*Table 1 shows that the largest zone of inhibition observed was 2.25 mm against *Bacillus cereus* (BC). Figure 4 displays the antibacterial activity of neem chitosan nanoparticle-treated cotton fabric (on the left) and the no bacterial repellency on polyester fabric (on the right), as indicated by the absence of a zone of inhibition indicating no absorbance of the finish on the polyester fabric because of its hydrophobic nature. Staphylococcus aureus displayed 1.45 mm of inhibition, while both Gram-negative bacteria exhibited very little zone of inhibition.The limited zone of inhibition observed in Gram-negative bacteria may be attributed to their rigid and complex outer membrane, hindering the diffusion of hydrophobic compounds through it.In contrast, Gram-positive bacteria lack this extra complex membrane and instead have a wall that is not dense enough to resist small antimicrobial molecules, allowing for easier access to the cell membrane [10].

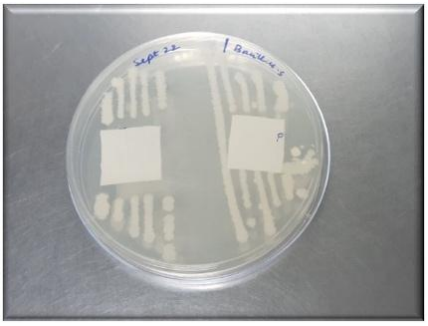

Figure 4: Activity of Neem chitosan nanoparticle against *Bacillus cereus* on treated cotton fabric (left) and polyester fabric (right) side of the petri dish

3.7 Effect of storage conditions on % oil retention

The % oil retention of neem chitosan nanoparticle treated cotton fabric was assessed with regards to its end use in both open and enclosed environments such as drawers or shelves, at room temperature referring the closed and open petri dish condition. The calculation was done after two months and compared to the initial zero time (after nanoparticle preparation). The findings revealed that the treated fabric samples that were kept covered in a petri dish demonstrated better stability compared to the ones left uncovered and exposed to the environment. The nanoparticles

showed a 2.7% change in entrapment efficiency after two months in a covered condition and a 12.8% change in uncovered conditions.

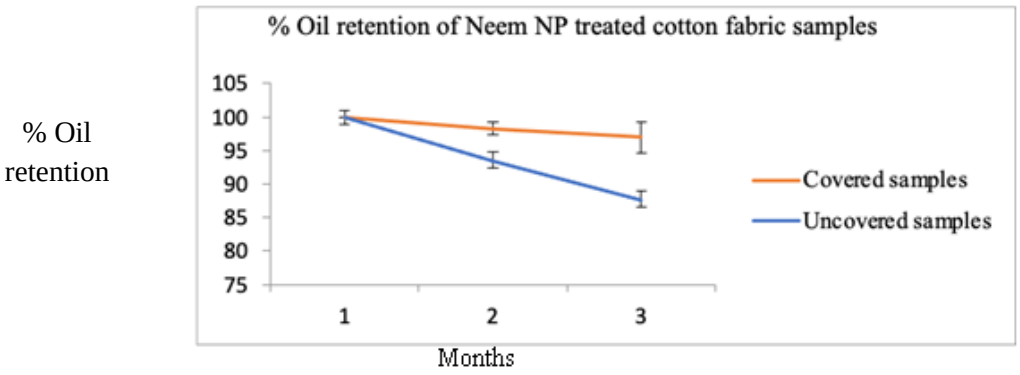

Figure5: % oil retention of the treated cotton fabric when stored in both covered and uncovered conditions

4. Conclusion

Neem essential oil was successfully loaded in the chitosan nanoparticle. As mentioned by Supraba, W., et al.,the good percentage of entrapment efficiency has to be more than 60% [7], the current study has observed entrapment efficiency of 78.42 ± 1.56 % with 189 ± 26 nm as an average size of the nanoparticle. The treated cotton fabric showed satisfactory antibacterial activity. Due to the control and release mechanism, the finish showed satisfactory stability in an open environment. Thus, the treated cotton fabric can be use as a preservative fabric for covering heritage textiles to protect from microbes. The fabric can also be used as a backing or a lining material when stored flat in shelves or drawers or as a padding on hangers and rollers. By doing so, it prevents direct application of the finish causing stress to the ancient textiles and helps preserve them for future generations.

5. Acknowledgement

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