

Evaluation of Jute Packages treated using Chitosan Nanoparticles/Neem Leaves Extract

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ABSTRACT

Improvement of people's living standards and need for environmental protection, the demand for natural biodegradable and ecofriendly fibers is rising worldwide day-by-day. Ligno cellulosic natural fibers such as flax, hemp, sisal and jute are an interesting, environmentally friendly. Jute bags were improved by the treatment with different particle sizes of nano chitosan suspensions (20, 67, 93, and 130 nm) and the blend of Neem leaves extract/starch to be used as anti-insect packages for agriculture crops storage. The rheological properties of different coating solution that is used for treatment of Jute, all samples exhibited non-Newtonian pseudoplastic behavior. Water vapor permeability (WVP), water absorption and tensile strength, were determined for all treated jute bags. The results observed that jute bags treated with nano-chitosan and Neem leaves extract /starch blends affects the properties of jute bags. Tensile strength increased as concentration of neem extract increased for jute packages treated with nano-chitosan suspension. Water vapor permeability of jute bags treated with the blend of neem extract/starch was higher than water vapor permeability of control jute bags.

Keywords: *Jute bags, Neem, Chitosan, rheological properties, water vapor permeability, water absorption, tensile strength.*

1. INTRODUCTION

Jute fiber reinforced composite as moderate structural materials have attracted the attention of material scientists all over the world because of their low cost, easy availability, light weight, renewability, and biodegradability. In addition, low density and high specific strength of jute reinforced polymer composites make them the most suitable candidates for low load bearing applications. Jute is a biodegradable, cheap, nontoxic, environment friendly, lignocellulose bast fiber. It is versatile and fast-growing renewable biomass and photo reactive crop with only

120-150 days duration from seed to fiber or maximum biomass. With the advent of cheap synthetic substitutes, bulk handling, containerization and storage in oils, jute and jute goods are losing markets harply in the important countries. Diversified use of jute is therefore essential to prevent further decline of the jute sectors [1].

Improvement of people's living standards and need for environmental protection, the demand for natural biodegradable and ecofriendly fibers is rising worldwide day-by-day. Ligno cellulosic natural fibers such as flax, hemp, sisal and jute are an interesting, environmentally friendly alternative to the use of glass fibers as reinforcement in engineering composites. These fibers are renewable, non-abrasive and can be incinerated for energy recovery since they possess a good calorific value and cause little concern in terms of health and safety during handling of fiber products [2]. In addition, they exhibit excellent mechanical properties, low density and are cheap. This environmentally friendly feature makes the materials very popular in the engineering markets such as automotive and construction industry. Jute fiber is a bast fiber obtained from the bark of jute plant containing three main categories of chemical compounds namely cellulose (58~63%), hemicellulose (20~24%) and lignin (12~15%) and some other small quantities of constituents like fats, pectins, aqueous extracts, etc. Jute fiber is composed of small units of cellulose surrounded and cemented together by lignin and hemi cellulose. Large amount hydroxyl group in cellulose gives natural fiber hydrophilic properties when used to reinforce hydrophilic matrices [3]. Chitosan, a natural linear bio-poly-amino-saccharide is obtained by alkaline deacetylation of chitin, which is the principal component of protective cuticles of crustaceans such a scrabs, shrimps, prawns, lobsters and cell walls of some fungi such as *Aspergillus*. Chitosan is a weak base and is insoluble in water and organic solvent. However, it is soluble in dilute aqueous acidic solution ($\text{pH} < 6.5$), which can convert glucose amine units into soluble form R-NH_3^+ [4]. It gets precipitated in alkaline solution or with poly an ions and forms gel at lower pH. Chitosan is inexpensive, biodegradable, and nontoxic for mammals. This makes it suitable for use as an additive in the food industry [5] and [6], as a hydrating agent in cosmetics, and more recently as a pharmaceutical agent in biomedicine [7-9].

The antimicrobial activity of chitosan was observed against a wide variety of microorganisms including fungi, algae, and some bacteria. However, the antimicrobial action is influenced by intrinsic factors such as the type of chitosan, the degree of chitosan polymerization, the host, the natural nutrient constituency, the chemical or nutrient composition of the substrates or both, and the environmental conditions (e.g., substrate water activity or moisture or both). Although, both native chitosan and its derivatives are effective as antimicrobial agents, there is a clear difference between them. Their different antimicrobial effect is mainly exhibited in live host plants [10].

Nanocomposites materials are widely used in various fields, among these fields is the application in textiles composite nature and structure where two principle ways can be considered for the use of nanocomposites in textile applications. Melt spinning of nano-composite polymer, which can be subsequently woven or knitted, had been demonstrated as a promising approach

Nanotechnology also has real commercial potential for the textile industry. This is mainly because conventional methods used to impart different properties to fabrics often do not lead to permanent effects and will lose their functions after laundering or wearing. Nanotechnology can provide high durability for fabrics, because nanoparticles have a large surface area-to-volume ratio and high surface energy, thus presenting better affinity for fabrics and leading to an increase in durability of the function. In addition, a coating of nanoparticles on fabrics will not affect their breathability or hand feel. Therefore, the interest in using nanotechnologies in the textile industry is increasing [11]. Coating is a common technique used to apply nanoparticles onto textiles. The coating compositions that can modify the surface of textiles are usually composed of nanoparticles, a surfactant, ingredients and a carrier medium [12]. Several methods can apply coating onto fabrics, including spraying, transfer printing, washing, rinsing, and padding. Padding is the most commonly used, [13-15]. The nanoparticles are attached to the fabrics with the use of a padder adjusted to suitable pressure and speed, followed by drying and curing.

The properties imparted to textiles using nanotechnology include water repellence, soil release, wrinkle resistance, anti-bacteria, anti-static and UV-protection, flame retardant, improvement of dye ability and so on [16]. This technique offers a simple

and mild preparation method in the aqueous environment. First, chitosan can be dissolved in

acetic acid in the absence or presence of stabilizing agent, such as poloxamer, which can be added in the chitosan solution before or after the addition of polyanion. Polyanion or anionic polymers were then added, and nanoparticles were spontaneously formed under mechanical stirring at room temperature. The size and surface charge of particles can be modified by varying the ratio of chitosan and stabilizer [17].

The objective of this research paper is the preparation and evaluation as jute packages using neem extract and chitosan nanoparticles which can be used in storage of agriculture crops.

2. MATERIALS AND METHODS

2.1. Materials

Jute fibers (JF) were purchased from public company of Jute. Sodium hydroxide and acetic acids was obtained from El-Gomhoria, Egypt, Chitosan was obtain from Acros Organics, Ceel, Blgium, Sodium tripolyphosphate (STPP) was obtain from Laboratory Reagents & Fine Chemical and Starch soluble was obtained from NICE Chemicals, Kerala, India and Glycerol was obtained from Piochem.

2.2. Methods

2.2.1. Rheological properties of coating blends

Rheological properties (viscosity, shear stress, shear rate) of different coating solutions and suspensions (nano-chitosan) were measured using Brookfield Engineering Labs DVIII Ultra Rheometer. The samples were placed in a small sample adapter at a constant temperature using thermo stated water bath. The viscometer was operated between 10 and 100 rpm, and the results of shear stress, shear rate, and viscosity data were obtained directly from the instrument; the SC4-21 spindle was selected for the measurement.

2.2.2. Washing and drying of Jute bags

The jute fabrics were washed with 2% detergent solution at 70°C for one hour, then washed with distilled water and finally dried in a laboratory oven at 70°C [18].

2.2.3. Treatment of jute fabric with Sodium Hydroxide

Jute was treated with 18% sodium hydroxide solution at room temperature for 45 min. The material- to liquor ratio was kept at 1:20 (w/w), the treated fabrics were thoroughly washed with running water, neutralized with 1.5% acetic acid solution, again washed with running water and then air dried at room temperature [19].

2.2.4. Preparation of Chitosan Nanoparticles

Chitosan nanoparticles were synthesized *via* the inotropic gelation of chitosan Sodium tripolyphosphate (STPP) anions. Chitosan (0.5%) was dissolved in acetic aqueous solution (1%). Concentration of acetic acid solution was 1.5 time higher than that of chitosan. STPP solution was prepared by double-distilled water. Chitosan nanoparticles were spontaneously fabricated with the drop wise addition of different ratios chitosan to STPP solution (2:1, 3:1, 4:1 and 5:1 (w/w), respectively) under magnetic stirring (1000 rpm, 1 hour) at room temperature [20]. Particles size were determined by Zeta sizer nano (nano ZS), Malvern, UK.

2.2.5. Preparation of Neem aqueous extract

Neem aqueous extract was prepared by mixing 5, 10, 15, 20 g of dry powder of Neem leaves with 100 ml of distilled water in round flask with occasional shaking overnight. The flask was kept in the dark to avoid effect of the light on the ingredients of Neem. The extract was then filtered through a muslin cloth for coarse residue and finally through Whatman No.1 filters paper and kept in an airtight amber colored container [21].

2.2.6. Preparation of Neem aqueous extract and starch blend

Starch sample (8%) was gelatinized using microwave. It was prepared by stirring the starch gently just before heating starch in microwave oven in order to ensure that starch was fully suspended in water, glycerol (2%) was added as plasticizer. Different concentration of Neem aqueous extract (5, 10, 15, 20%, respectively) were added to gelatinization starch at 50°C and stirring them for 10min.

2.2.7. Test procedure of Moisture absorption 24 Hour/Equilibrium ASTM D570

For water absorption test, the specimens are dried in an oven for a specified time and temperature and then placed in a desiccator to cool. Immediately upon cooling the specimens are weighed. The material is then emerged in water at

agreed upon condition, often 23°C for 24 hours or until equilibrium. Specimens are removed, patted dry with a lint free cloth, and weighed.

1.1.1. Determination of Water vapor permeability (WVP)

The water vapor permeability of the samples has been measured using the cup method, according to ASTM 96 (procedure B) testing standard. This method is the straightforward one, involving the determination of weight loss, with evaporation time (24) of water contained in a cup, the top of which is covered by the cover ring. In this method, test fabric is placed in an airtight manner over the top of a cup. Another cup contains the reference fabric secured in the same airtight manner. The dimensions of the cup were calculated to give a 10 mm deep layer of air between surface of the water and underside of the specimen. The technique compares the rate of water mass transfer through fabric and other from cups, which are covered with reference fabric and other test samples. The weight of the cups was measured firstly at the starting of the test and then periodically after a certain time interval by the balance with resolution of 0.01g to determine how much water has been lost from each one. The difference in water loss between a cup covered with the standard fabric and one with test fabric enable to study the relative rate of moisture movement through the test fabrics, so that the moisture vapor permeability of the test specimen can be calculated.

The water vapor permeability index was calculated by expressing the water vapor permeability (WVP) of the fabric as a percentage of the (WVP) of reference, as shown below:

$$WVTR = \frac{\Delta m}{\Delta t A}$$

(1)

$$WVP = \frac{L}{\Delta RH}$$

(2)

Where, $\frac{\Delta m}{\Delta t}$ is the moisture gain weight per time (g/s), A is the surface area of the film m², L is the film thickness (mm) and ΔRH is the difference in relative humidity [22].

2.3. Experimental procedure

2.3.1. Treatment of Jute packages

All Jute bags were treated with sodium hydroxide and then divided into sets:

Jute bags treated with different concentrations of chitosan, Neem extract, chitosan nano particles.

2.3.2. Jute bags treated with different particle size of nano-chitosan

Jute bags were soaked in different particle size of nano chitosan (20, 67, 93 and 130 nm, respectively).

2.3.3. Mechanical properties of Jute bags

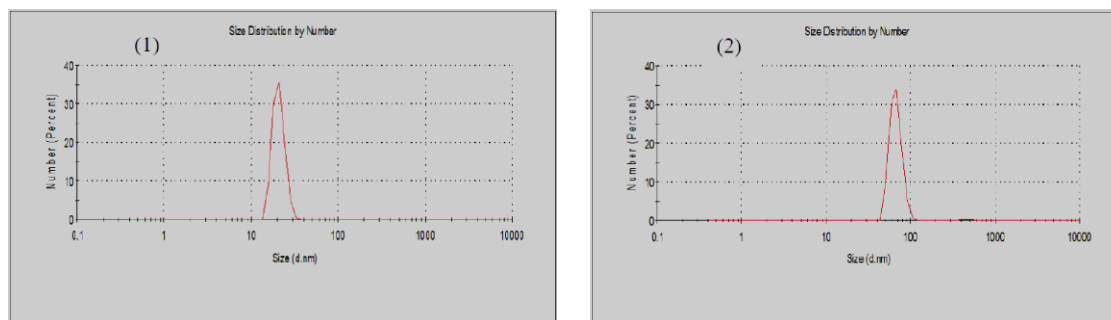
Tensile properties of treated Jute bags were examined from stress strain curves measured by using a tension meter carried out with use of (Mecmesin Ltd), tension speed was 50/100 mm/min, load range 2500 N and extension range 1500mm.

4. RESULTS AND DISCUSSION

4.1. Preparation of chitosan nanoparticles

Macromolecular chitosan, however, encounters several challenges, while its applications to textiles, in particular. Investigations have shown that the inherent properties of cotton fabric such as appearance and handle and other essential properties like fastness of dyes to various agencies due normal chitosan treatment were found to be affected [23].

These detrimental effects of chitosan are mainly attributed to its lack of penetration into fabric structure causing the surface deposition of film [24] and [25]. One possible way to enhance its effectiveness is to reduce the particle size closer to nano level, which facilitates the greater penetration of CHT into fabric structure. The characterization was done by the determination of particle size. Particle size distribution of nanochitosan was shown in figure (1).



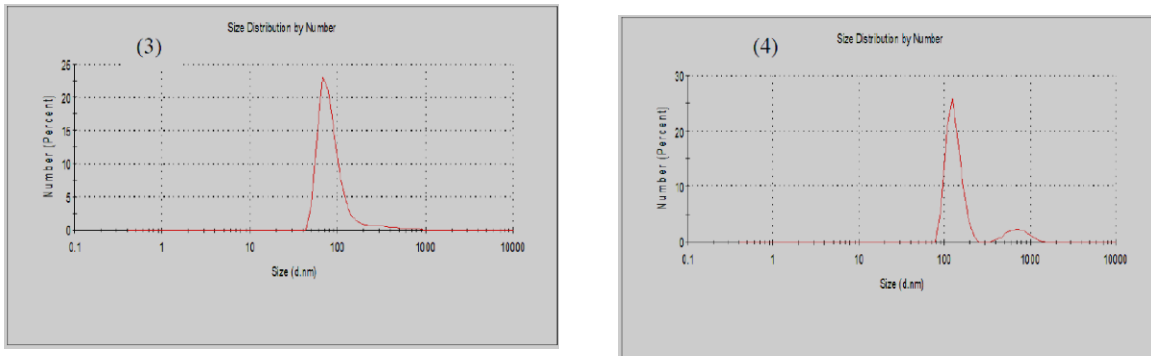


Fig. (1) Particle size distribution of nanochitosan
Chitosan nanoparticles (1) 20 nm (2) 67 nm (3) 93 nm (4) 130 nm

4.2. Rheological properties of different edible coating solutions

4.2.1. Effect of Shear rate on apparent viscosity

Figure (2) shows the effect of shear rate on apparent viscosity of the blend of different particle size of Nano chitosan suspension. The results observed that viscosity decreases as shear rate increased. The lower apparent viscosity of chitosan nanoparticles suspensions may be due to TPP-cross linked chitosan molecules turned into more dense particles whose hydrodynamic volumes were small, [26]. Apparent viscosity decreases as particle size increased this may be due to that the larger the particle size the higher will be the resistance offered for the flow of liquid and hence the higher will be the viscosity and vice versa [27]. Also, apparent viscosity of Neem leaves extract and starch decreased with increasing shear rate.

The shear thinning region curve can be expressed by the following power law equation

$$\mu = k \gamma^{n-1}$$

Where, μ is the apparent viscosity, Pa.s, k is the consistency index, γ is the shear rate, 1/sec, and n is the flow behaviour index.

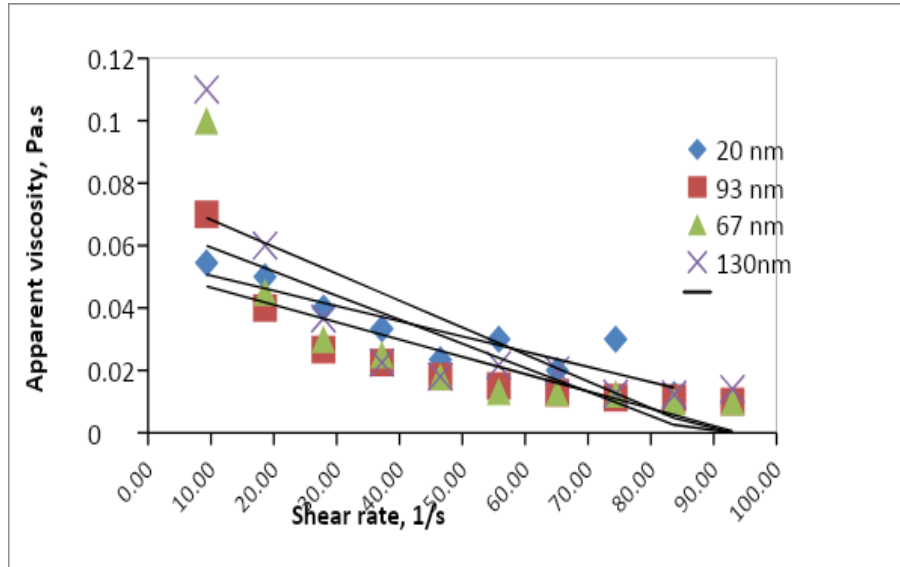


Fig. (2) Effect of shear rate on apparent viscosity for (1) different particle size of nano chitosan solution, (2) different blends of neem leaves extract and starch.

4.1. Thixotropic effect

4.1.1. Thixotropic effect of chitosan solution

The characterization of time-dependent rheological properties of food systems is important to establish relationships between structure and flow, and to correlate physical parameters with sensory evaluation [28].

4.1.2. Thixotropic effect of chitosan nanoparticles solution

Figure (3) shows the thixotropic effect of different particle sizes (20, 67, 93, and 130 nm, respectively). The results observed that thixotropic behavior is evident in these samples more than chitosan, neem extract or even the blend of Neem extract and starch, this means that jute bags treated with nano chitosan solutions will stick more than others.

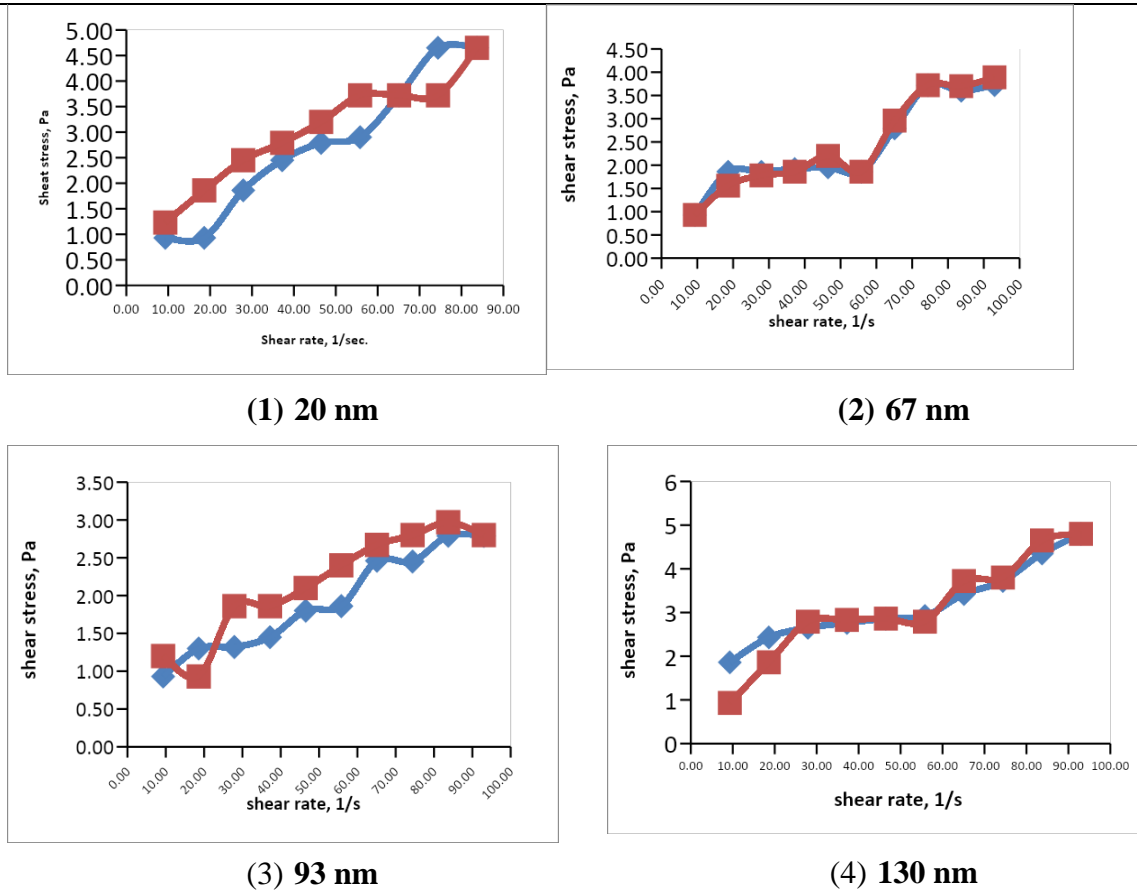


Fig. (3) Thixotropic effect of chitosan nanoparticles

4.3. Treatment of Jute bags

4.3.1. Treatment of Jute bags was done at two steps:

The first step was treating jute bags with NaOH (18%) to remove artificial impurities and improve interfacial adhesion. The second step was that jute treated with different concentration of chitosan (0.25, 0.5, 0.75 and 1%), the adhesion of chitosan on the fiber surface depends on the hydrogen bonding between amide group and cellulose hydroxyl group [28]. After these two steps jute bags was treated with different concentrations of Neem extract and the Neem/starch blend.

4.1. Water absorption

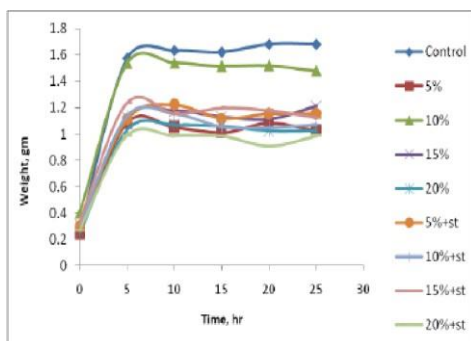
4.1.1. Water absorption of Jute bags treated with chitosan nanoparticles suspensions

Absorption is the ability to retain a liquid in the fabric. It consists of several parts, first the fabric surface is wetted and the liquid is transported into the

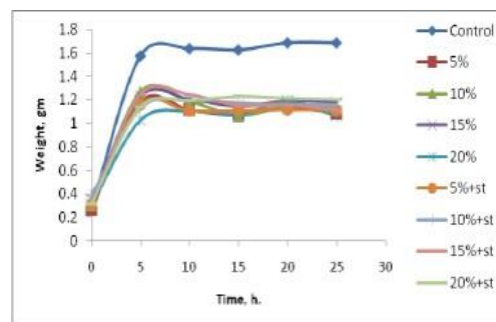
voids between the fiber and are absorbed into the fibers and diffuse. The initial absorption is very quick, and then the absorption slows down until the fabric is saturated. The liquid can be retained in the weave intersections in the capillary spaces between the fibers or in the capillary spaces in the fibers. There are two aspects of absorption. Both are important: how quick a fabric absorbs a certain amount of liquid and how much it absorbs.

Figure (4) shows the water absorption for jute bags treated with different chitosan nanoparticles (20, 67, 93 and 130 nm) then treated with different concentrations of neem extract and neem extract/starch blend. The results observed that samples treated with 10% neem and control sample, moisture content of jute samples increased rapidly for the first 5 hours which indicates early stage of the water sorption of the jute samples. After around 24 hours and above, the moisture content of the jute bags reached a plateau indicating that moisture content is equilibrated with the relative storage humidity for all samples studied as shown in Figures (55-58), with respect to control jute bags except for jute bags while samples treated with 10% Neem extract was near to control bags. Samples treated with the blend of Neem extract/starch; moisture content increased less rapidly.

(1) 20 nm



(2) 67 nm



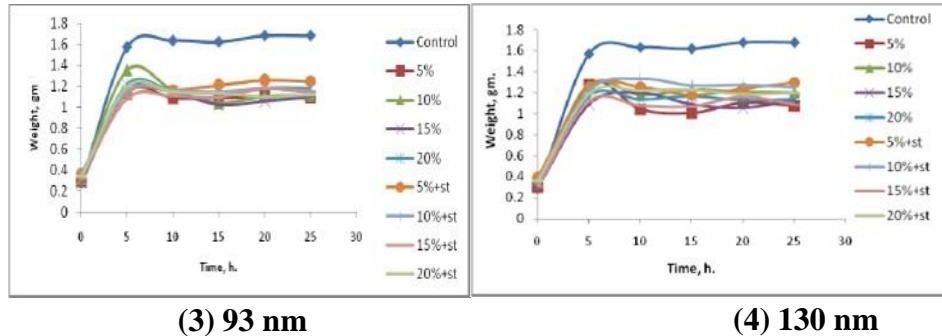


Fig. (4) Water absorption for jute bags treated with chitosan nanoparticles, then different concentrations of Neem extracts and the blend of Neem extract/starch

4.3.2. Vapor permeability of chitosan nanoparticles treated jute bags

Jute bags were treated first with chitosan nanoparticles solution (20, 93, 67 and 130 nm), then divided into two groups, the first group was treated with different concentrations of neem extract (5, 10, 15 and 20%) and the second group was treated with the blend of neem extract and starch. Thickness of different packages affects other characteristics such as tensile strength, elongation and water vapor permeability, the thickness depends on processing parameters (97, 98). The results observed that thickness for all samples didn't give a good trend with increasing the concentration of Neem extract solution and nanoparticles suspensions as shown in table (2). water vapor permeability of Jute bags treated with chitosan nanoparticles solution (20nm), then neem extract and neem extract/starch blend.

Water vapor permeability of jute bags treated with chitosan nanoparticles solution (20nm), then neem extract and neem extract/starch blend were determined. The results observed that water vapor permeability of jute bags (nano chitosan treated jute bags) treated with different concentrations of neem extract (5, 10, 15 and 20%) increased as concentration of neem extract increased, the lower water vapor permeability with respect to control bags was (0.747g. mm/m². mmHg) for jute bags treated with 5% neem extract. Water vapor permeability of jute bags treated with the blend of neem extract/starch didn't give a good trend (0.99, 0.88, 0.968 and 0.907 g.

mm/m².mmHg) and was higher than water vapor permeability of control jute bags (0.842 g.

mm/m².mmHg). Water vapor permeability of jute bags treated with chitosan nanoparticle suspension (67, 93, 130nm) the lower water vapor permeability with respect to control bags was (0.823, 0.67, 0.726 g. mm/m². mmHg) for jute bags treated with 5, 15% neem extract and the blend of 20% neem extract / 8% starch respectively.

Table (1) Water vapour permeability and thickness of different treated jute bags

Items	Thickness, mm	WVP, g.mm/m ² .mmHg	Thickness, mm	WVP, g.mm/m ² .mmHg	Thickness, mm	WVP, g.mm/m ² .mmHg	Thickness, mm	WVP, g.mm/m ² .mmHg
Control	1.236	0.842	1.236	0.842	1.236	0.842	1.236	0.842
5% Neem Extract	0.788	0.747	0.996	0.823	1.096	0.986	1.154	0.823
10% Neem Extract	1.11	0.775	1.146	0.957	1.035	0.763	1.343	0.957
15% Neem Extract	1.008	0.89	1.272	1.022	0.857	0.67	1.192	1.022
20% Neem Extract	1.152	0.92	1.055	1.541	1.047	0.753	1.078	1.541
Starch+5% Neem	1.226	0.99	1.091	1.918	1.124	0.957	1.114	1.918
Starch+10% Neem	1.128	0.88	1.186	1.186	1.208	1.048	1.119	1.186
Starch+15% Neem	1.397	0.968	1.063	1.063	1.177	1.546	1.075	1.063
Starch+20% Neem	1.147	0.907	0.938	0.938	0.96	1.062	1.068	0.938

4.1. Mechanical properties of treated jute bags

4.1.1. Tensile strength of jute bags treated with chitosan nanoparticles

Figure (5) shows the tensile strength of jute bags treated 20, 76, 93, 130 nm chitosan nanoparticles solution then different concentrations of neem extract (5, 10, 15 and 20%) and the blend of neem extract/8%starch. The results observed that tensile strength increased from (56.62 to 80.3619 MPa), (51.863 to 61.775 MPa), (59.02 to 107.439 MPa), (70.215 to 94.07 MPa) as concentration of neem extract increased for jute packages treated with 20, 76, 93, 130 nm of nano-chitosan respectively, the higher tensile strength was 80.3619, 61.775, 107.439 and 94.07 MPa for jute bags treated with for jute packages treated with 20, 76, 93, 130 nm of nano chitosan respectively and 20% neem extract. As starch added to neem extract, the tensile strength increased from (86.526 to 100.66 MPa), (65.079 to 75.19 MPa), (118.503 to 151.695 MPa) and

(96.179 to 115.652 MPa) for jute packages treated with 20, 76, 93, 130 nm of nano chitosan respectively and different

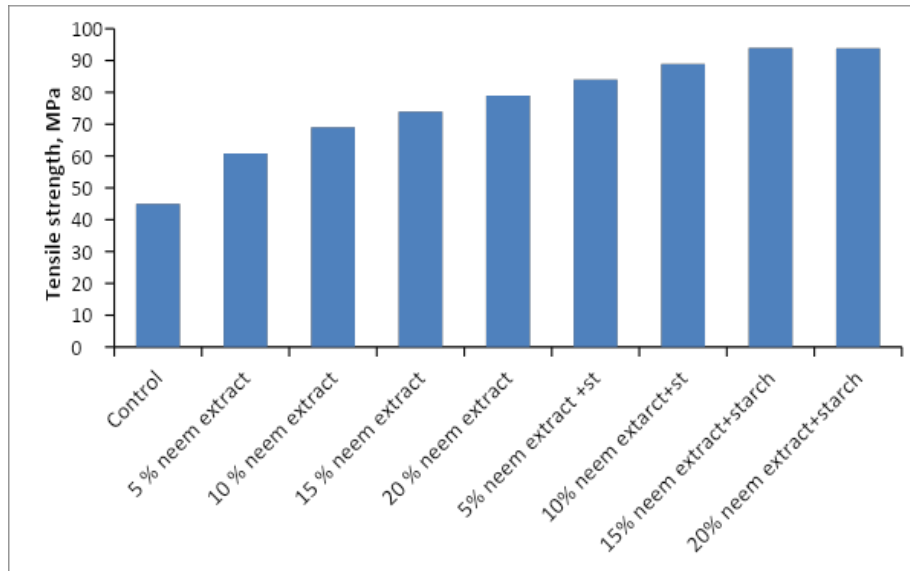


Fig. (5) Tensile strength of jute bags treated first with chitosan nano particles (20, 67, 93, 130 nm) and then different concentrations of neem extract and the blend of neem extract /starch.

5. CONCLUSION

Jute bags were treated with nano chitosan suspension with the addition of Neem leaves extract. The rheological properties of various samples were investigated, and all samples showed non-Newtonian pseudoplastic behavior. The thixotropic effect was assessed for all samples to select the optimal coating solution that provides the best adhesion to jute bags. The findings demonstrate that solutions with a higher thixotropic behavior adhere to jute bags better. The thixotropic effect was seen in samples of Neem extract (5 and 20%) and (20nm, 93nm nano chitosan suspensions). The results reveal that treated jute bags affect the permeability, water absorption, and tensile strength of the material.

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