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Original article

ENHANCING THE MECHANICAL STRENGTH OF KLUCEL E/CNC COMPOSITES FOR THE CONSERVATION OF WOODEN ARTIFACTS

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Abstract:
In this research, cellulose nanocrystals (CNC) were prepared with a concentration of 2% through a hydrolysis procedure using concentrated sulfuric acid (H_2SO_4). This was followed by the preparation of CNC/Klucel E nanocomposites films with various concentrations (10, 15, 20, 30, 50 %). The obtained films were investigated and evaluated using different techniques such as X-ray diffraction (XRD), Scanning Electron Microscopy (SEM), and tensile strength testing. Based on the results obtained, films with concentrations of 20 and 30 %, which gave the best results, were applied on aged wood specimens in order to study the influence of this nanocomposite on different properties of wood. The influence was studied in terms of mechanical properties, change in chemical composition, and change in physical appearance through tensile tests, Fourier Transform Infrared Spectroscopy (FT-IR) and color change measurements (CIE lab), respectively. The tensile strength of the films was improved after addition of CNC to Klucel E by almost 70% at 30% concentration, and compression strength for wooden samples significantly increased after using the same concentration of the nanocomposite, which reached 35 MPa.

1. Introduction

Wooden artifacts are often affected through different degradation factors such as microbiological infestation or exposure to weather. Several studies have been conducted to investigate the chemical changes that occur in previously identified softwoods [1] and hardwoods [2,3], that were commonly used in manufacturing ancient Egyptian artifacts. Results showed that depending on the decay factors, main components of wood such as cellulose, hemicellulose and lignin are affected. This effect results in a change in the physical appearance of wood and a decrease in mechanical properties. Therefore, there is a continuous search for suitable polymers that could be used for wood treatment. Recently, many natural and synthetic polymers, including cellulose ethers, have been applied on decayed wooden artifacts [4,5] Consequently, interest in studying polysaccharides as wood consolidant increased because of their compatibility to natural cellulose [6]. Cellulose derivatives particularly cellulose ethers such as CMC, HPC, and MC [7] have been reported to be used in the conservation field in consolidation of cellulosic materials such as wood, either in consolidation [8-10] or gap filling [11-13]. Furthermore, such materials have been used in the treatment of textiles and manuscripts [14]

for the enhancement of mechanical properties. However, different types of cellulose derivatives used in the conservation process possess poor mechanical properties [15], therefore it was necessary to figure out a way to improve their mechanical properties [16]. Fortunately, nanotechnology offers very promising solutions to synthesize nanocomposites through the addition of nanomaterials as fillers, which have the ability to improve the mechanical properties [17, 18]. Nanocomposites present very promising solutions to improve mechanical and optical properties depending on the interactions between the filler and the matrix [19]. The formation of network within the matrix between CNC particles and polymeric chains depends on a percolation threshold that relies on the aspect ratio of the nano fillers and strength of the filler/filler interactions [20], therefore the main challenge in the preparation of CNC-reinforced nanocomposites is to obtain good CNC dispersion in the matrix. Cellulose nanocrystals (CNC), which represent nanoparticles extracted from lingocellulose fibers via hydrolysis with strong acids [21] have drawn considerable attention over the recent decades [22,23] because of their interesting mechanical properties [24]. Similar to many nanomaterials, CNC exhibit distinguishable mechanical properties due to their nanoscale structure and crystallinity, they also have high aspect ratio alongside with high specific crystal modulus of cellulose, which is about 100-150 GPa [25,26]. Moreover, they possess abundant hydroxyl groups, which imply the availability of further chemical modifications. This led to the use of CNC as filler in nanocomposites in order to enhance the mechanical properties of various polymeric materials in previously published research [27]. Polymeric matrices exhibit an increase in the mechanical properties to a large degree with slight addition of CNC contents [28,29]. Li et al., investigated the influence of the incorporation of CNC in poly vinyl alcohol; and it was noted that the incorporation increased the crystallinity at all investigated relative humidity. In addition, it made positive influence on the tensile strength [30]. CNCs have been used as nano fillers in various natural polymer matrixes such as chitosan, gelatin, starch, and cellulose derivatives such as carboxymethyl cellulose, hydroxypropylcellulose, and cellulose diacetate [31]. It has been noted, that by the addition of CNC to chitosan the mechanical strength was improved until it reached 13.2% higher than that of the CNC-free membrane [32]. Although the reinforcement of adhesives with the addition of nanocellulose has been already reported, as well as the use of CNC as a reinforcement material for wood, where Gindl-Altmutter reported that the addition of CNC improved both the physical and mechanical properties of the wooden panels [33], the incorporation of CNC into nanocomposites is rarely reported in the scope of archaeological conservation as a possible consolidant material for wood artifacts due to the complexity of requirements for substances involved with archaeological materials. For example, consolidating materials should have the ability to penetrate the wood and last for a very long time, as a nanocomposite matrix and filler it should be compatible with the artifacts substrate and the composite should have a suitable structure to penetrate the wood providing reinforcement to the wooden structure [9]. Hamed et al [8] investigated the influence of the incorporation of different concentrations of nanocellulose as a filler to improve the efficiency of hydroxypropyl cellulose (HPC). The results showed that using nanocellulose as a filler has a high potential to be used in the consolidation of wood without undesired effects on its properties. Antonelli et al [34]. investigated the penetration of CNC on waterlogged archaeological wood samples of different species. The results obtained showed that

CNC penetrated only about a millimeter inside the treated wood. Furthermore, Cataldia et al, studied composites of methacrylic-siloxane-cellulose, which are composite coatings for wood protection, to assess the potential protective performances. Nanocellulose increased glass transition temperature, in addition to improving thermal properties, dimensional stability and stiffness of the neat photo resin [35]. As discussed in previous research, CNC is considered a promising material for the field of conservation because of its properties, such as high mechanical strength, stiffness, and durability [36,37]. In this paper an attempt to generate composites based on nanocellulose was carried out. Films of Klucel E/CNC nanocomposites in different concentrations were synthesized according to previously published methods to determine the best concentration for the enhancement of the mechanical properties of Klucel E (hydroxypropyl cellulose) for its application in the conservation of wooden antiquities. After interpretation of results of tested films, the best concentrations were applied on wood samples to study how the properties of wood specimens were enhanced and to investigate the influence of composites on chemical and mechanical properties of wood components. *Ficus sycomorus* wood was chosen for this experimental study, because it is a wood species very commonly found in many ancient Egyptian artifacts such as wooden furniture, coffins and ethnographic utensils.

2. Material and Methods

2.1. *Materials*

Two main materials were chosen for the preparation of Klucel E/CNC nanocomposites films. Klucel E (hydroxypropyl cellulose), with commercial name (Pulver Beutel), and MCC (pure Micro-Crystalline Cellulose) were obtained from Deffner & Johann Company, Germany. Sulfuric acid used in the hydrolysis process, which was 95-98%, was purchased from Scharlau Co., Germany. The precursor of CNC was MCC with a particle size of 50µm and pH value ranging from 5 to 7. Wood blocks of *Ficus sycomorus* obtained from Egyptian villages in the delta were prepared and cut for compression testing.

2.2. *Preparation methods of nanoparticles, nanocomposite films and wood specimens*

2.2.1. Synthesis of CNC

CNC was synthesized by following a strategy similar to that reported previously [38, 39]. Hydrolysis process for precursor micrcrystalline cellulose MCC was carried out with 64 wt.% sulfuric acid, 9 ml of sulfuric acid/1g of MCC. The hydrolysis process was carried out at room temperature under vigorous stirring for 45 minutes. Immediately after hydrolysis, suspensions were diluted 10-fold and allowed to settle. The suspensions were then centrifuged at 6000 rpm, washed with water, and the step was repeated three times until the supernatant was turbid. The resulting precipitate was dialyzed for several days using dialysis membranes against distilled water until the water pH remained constant. To achieve colloidal cellulose particles, suspensions were sonicated four times for 10 minutes with 5 minutes off in a sonication bath. Following the one-hour of sonication, 30 seconds of sonication was carried out with 30 seconds off for 30 minutes in a water bath to avoid overheating. After the synthesis of CNC particles, the dispersion was monitored for two months, where no precipitation was noticed during this period.

2.2.2. Synthesis of Klucel E/CNC nanocomposite films

The matrix (Klucel E) with a concentration of 3% was dissolved in ethanol (w/v) and CNC dispersion in water with a concentration of 2%. Nanocomposite films of Klucel E/CNC were produced through the solution casting technique. CNC particles were dispersed in water, which was followed by

their addition to the matrix to prepare five samples with five different concentrations (10, 15, 20, 30 & 50%). The samples were stirred for 15 minutes at 100 rpm before being cast into silicon plates with a thickness of 20 ml for each thin film. Films were allowed to air dry at room temperature for several days depending on the concentration.

2.2.3. Preparation of wood samples

Wood blocks with dimensions of 20×20× 60 mm according to ISO 3787 (1976) were prepared for the assessment of mechanical properties. To simulate archaeological degraded wood, accelerated ageing of the wooden specimens was carried out in climatic chambers for 300 hours at 105°C and relative humidity 60 %. Wood samples were then impregnated by their immersion in consolidation materials (Klucel E 3%, Klucel E/CNC 20 % and Klucel E/CNC 30 %) for 15 minutes.

2.3. *Evaluation of CNC particles and Klucel E/CNC nanocomposite films and wood specimens*

2.3.1. Particle size measurements for CNC particles

Zetasizer equipment (Nicomp Nano Z3000) was used to measure particles mobility and charge (zeta potential) and the particle size of CNC using the DLS (dynamic light scattering) technique. Sample of CNC suspension was diluted with water to 0.05% concentration

2.3.2. Scanning electron microscope (SEM)

Scanning Electron Microscope (JEOL JSM 5400LV EDX Link ISIS-Oxford "high vacuum") was used to examine the structural characterization of nanocomposite samples prepared in the form of thin films to study the dispersion of CNC particles within the polymer alongside the degree of agglomeration.

2.3.3. X-ray diffraction (XRD)

The crystalline phases of the synthesized nanocomposite samples were investigated by using X-ray diffraction (XRD) (D8 Discovery-Bruker Company), 40 KV and 40 AM (1600W) at speed scan 0.01 and 2(θ) range from 5° to 60° on samples of thin films for all concentrations. This technique was carried out to study whether the incorporation of CNC particles would influence the crystallinity of the polymer or not. In addition, the structures of both CNC and MCC were investigated using XRD to determine whether the adapted procedure resulted in the formation of crystalline CNC particles or not.

2.3.4. Mechanical testing

The tensile strength, elongation at break and elastic modulus were measured at 23°C and at crosshead speed 5 mm/min. Klucel E/ CNC composite films with five different concentrations were cut in the form of a dumbbell shape and evaluated according to ASTM D638-14.

2.3.5. Viscosity

Viscosity of nanocomposites was measured with a Brookfield viscometer (DV2T) device using spindle 25 at speed rate 120 rpm and temperature 23°C.

2.3.6. Color change

The **L**, **a**, and **b** values were measured three times for each sample with Hunter Lab device; first measurement was before treatment and the sample was considered a control sample to provide standard measurements, the second measurement was taken directly after the immersion and drying of the samples, and the third measurement was after exposure to the aging process for 300 hours. Hence, ΔE for second and third measureements was calculated in comparison with the control sample.

2.3.7. Infra-Red Spectroscopy (FT-IR)

FTIR analysis for wood samples was carried out using Nicolet 380 spectrophotometer (Thermo Fischer Scientific) in the range of 4000–400 cm^{-1} for the characterization of materials and treated wood.

2.3.8. Compression testing of wood

Compression strength was measured with Losenhausenwerk tensile and compression testing machine, with a capacity of 400 KN, expanded uncertainty (±0.4%) and calibrated according to ISO 7500.

3. Results

3.1. *Particle size and concentration of composite*

The synthesis method adapted throughout this paper was followed by a procedure to determine the concentration of CNC. 20 ml of CNC solution was transferred into weighed glass beaker to determine the concentration. The sample was left to dry in the oven at 80°C until it was completely dry. The beaker was weighed again after drying the sample to determine the actual weight of synthesized particles, and according to the dry weight of the particles the w/v concentration was calculated. 20 ml of the prepared solution had a concentration of 0.4g/20ml = 2%. The size and zeta potential for CNC particles were evaluated by DLS techniques. From NICOMP DLS curve, the mean diameter of CNC was approx. 61 nm, fig. (1) with average zeta potential of -41 mv.

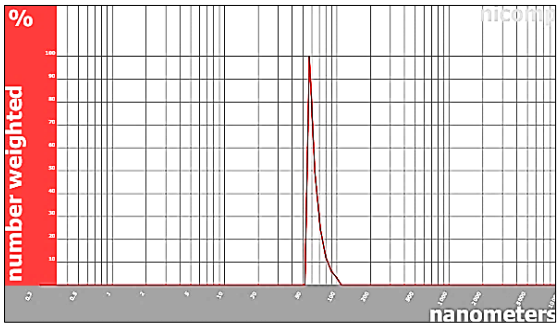


Figure (1) Shows NICOMP DLS curve of synthesized CNC

3.2. *Scanning electron microscope (SEM) of films*

The surfaces of each sample of nanocomposite films were examined by SEM. The surface micrographs demonstrate the purity of pure Klucel E. However, the surfaces became rougher with the addition of CNC, fig. (2-a). The micrographs of samples with 10%, 15%, and 20% concentration displayed the smoothness of the three composites' surface, which did not change significantly after the incorporation of CNC, where no aggregations were observed in the figs. (2-b, c, d). The micrograph of the 30% CNC sample showed slight increase in the CNC particle size and formation of slight aggregates as indicated in fig. (2-e). However, the 50% CNC sample showed a dramatic increase in the particle size resulting from the significant aggregation of CNC particles as seen in fig. (2-f).

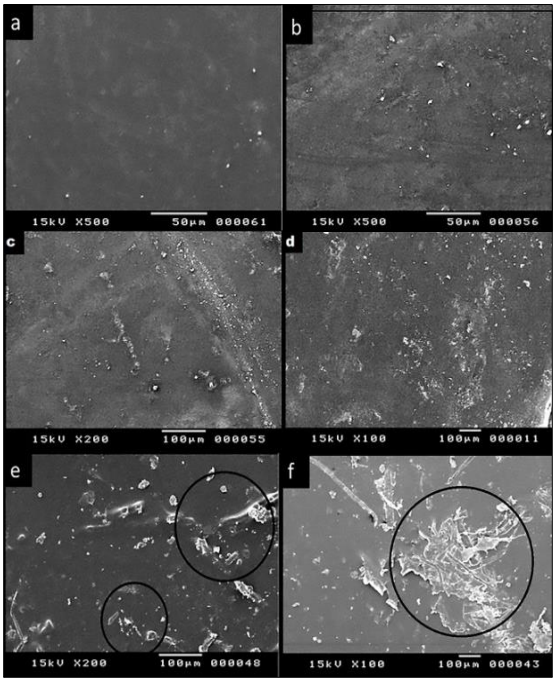


Figure (2) Shows SEM micrographs; **a.** Klucel E, **b.** 10 wt. %, **c.** 15 wt. %, **d.** 20 wt. %, **e.** 30 wt. %, **f.** 50 wt. % of CNC

3.3. *X Ray Diffraction (XRD*

The XRD pattern of all samples showed an obvious increase in peak intensity with the addition of CNC particles. Samples with 20% and 30% had the highest intensity of an extra peak at $2\theta=9^{o}$ demonstrating the increasing effect of the addition of CNC particles on the crystallinity, fig. (3) [40,41].

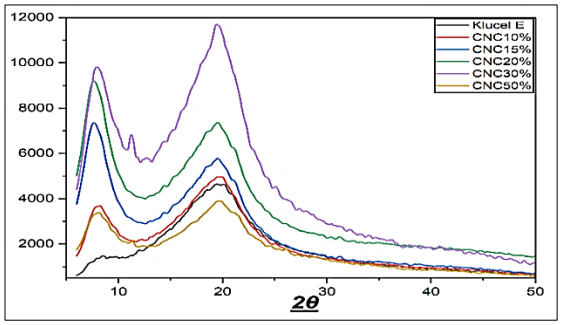


Figure (3) Shows XRD patterns of pure Klucel E, CNC 10%, 15%, 20%, 30% and 50%.

3.4. *Mechanical properties of thin films*

The mechanical properties of the synthesized composite films containing different CNC content (10, 15, 20, 30 & 50 %) are depicted in fig. (4). It can be seen that the tensile strength, fig. (4-a) was improved after addition of CNC to Klucel E matrix by almost 70% at 30% concentration, while at 50% concentration tensile strength showed a decrease. Obtained data shows the change in elastic modulus of the composite as a function of CNC content, fig. (4-b). It appears that the elasticity gradually increases with the increase in the concentration of filler (CNC); the elasticity rises from 300 in pure Klucel to 1160 Mpa after addition of CNC at a con-centration of 10% and then rises to 1500 MPa and 1700 MPa in 20% and 30% concentrations respectively, but it decreases slightly at 50% concentration. Hardness of the composite increased gradually from 65.12 for Klucel E to 80.22 at 50% concentration on "shore D" scale, fig. (4-c); the hardness did not decrease abruptly as in the case of the tensile strength at concentrations higher than 30%. Elongation at break parameter showed a significant difference according to the nature of the tested material. Klucel E recorded the highest percentage reaching 28%, which dramatically decreased with addition of CNC. It reached an average of 15-16 in each of CNC 10%, 15% and 20% concentrations, while it decreased to 11.2% in 30% concentration, and declined completely at 50% concentration, where it reached 2.7%, fig. (4-d).

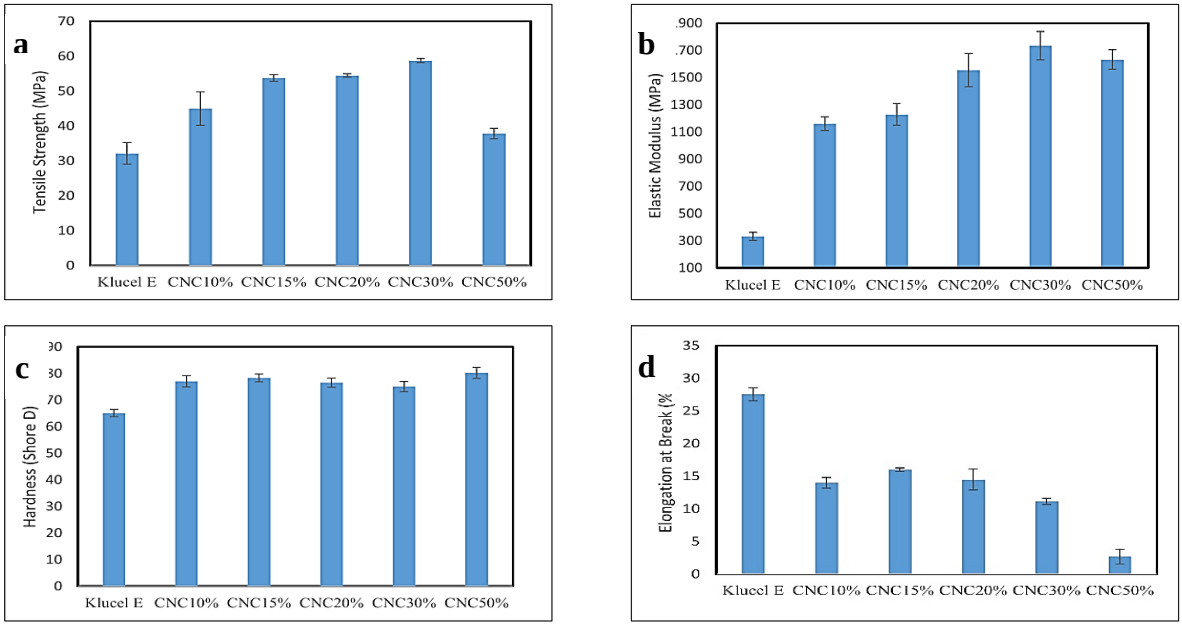


Figure (4) Shows mechanical properties of Klucel E/CNC nanocomposite

3.5. *Viscosity measurement*

The data obtained from the aforementioned tests on nanocomposites with different concentrations of CNC has shown that concentrations of 20 and 30% had the best influence on the mechanical properties. But prior to the application on the wood samples, the viscosity of the consolidant of nanocomposites was compared to that of CNC 2% and Klucel E 3%. The chosen concentration of Klucel E, is commonly used in the reinforcement process due to its intermediate viscosity that allows its penetration in the wood. The low viscosity of CNC suspension lowered the viscosity of CNC 20% composite from 16.7 cP for pure Klucel E 3% to 13.42 cP and to 10.70 cP in CNC 20 and 30% composite, respectively, fig. (5).

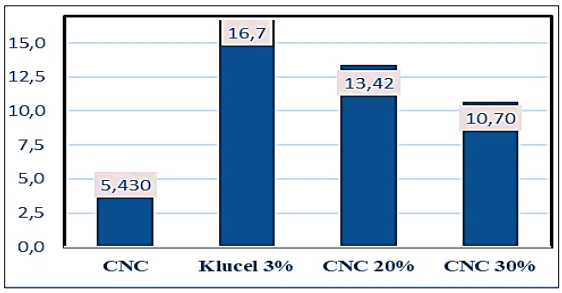


Figure (5) Shows viscosity measurements for CNC suspension, Klucel E 3% and CNC composites with 20% and 30%

3.6. *Weight gain for wood specimens treated with nanocomposites*

The two chosen concentrations were applied on wood samples in order to study the changes that occur on the sample in terms of color, physical appearance, chemical composition, and mechanical properties. Investigation of treated wood samples was carried out after exposure to an accelerated aging process in order to determine the compatibility of this type of nanocomposite, which is to be applied in the reinforcement of wooden artifacts. Wood samples were weighed before and after immersion in order to calculate the gained weight of wood sample after the impregnation process. From the calculated gain of weight in wood samples treated with pure Klucel E compared to samples treated with Klucel E/CNC nanocomposites it can be observed that samples treated with consolidant show gradual gain in their weight; sample treated with Klucel E gained 0.25 g, Klucel E/ CNC 20% samples gained approximately the same weight and Klucel E/CNC 30% samples gained 0.3g, fig. (6).

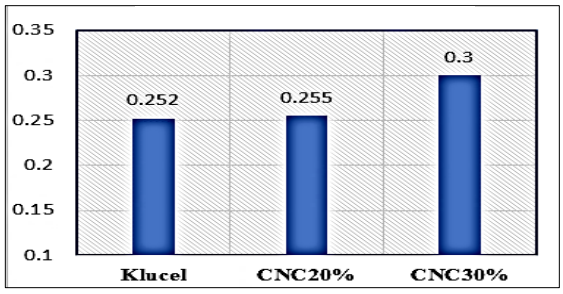
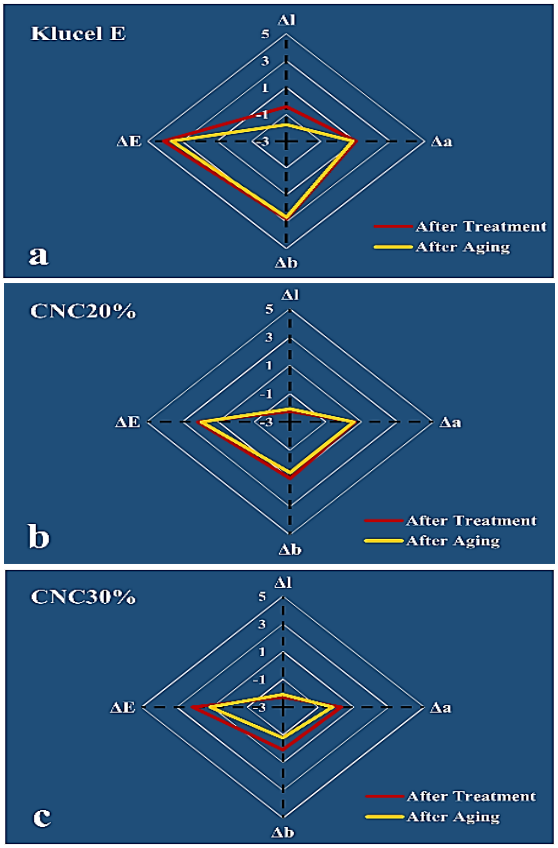


Figure (6) Shows gained weight graph after impregnation with consolidant materials, Klucel and composites with CNC 20 % and 30%

3.7. *Color change measurements*

By comparing changes in parameters (ΔL, Δa, Δb, ΔE) for the samples immersed in Klucel E, and nanocomposites with CNC 20 and 30% concentrations, it was observed that Klucel E 3% causes a change in color more than the composites; the changes decreased with the increase of CNC percentage. The specimen consolidated with composite of CNC 30% shows the lowest differences in color, fig. (7-a). Samples treated with only Klucel E showed a high change in Δb and ΔE which reached a value of (4) after immersion process. There was no noticeable change when measurements "after treatment" and "after aging" were compared with the exception of ΔL, which decreased, fig. (7-b). The ΔE value decreased with the addition of CNC to the matrix; this reduction increased with the raise of CNC concentrations. It was noted that ΔE decreased to 2.14 in CNC 20 % samples and to 2.11 in CNC 30% samples, figs. (7-c & d).



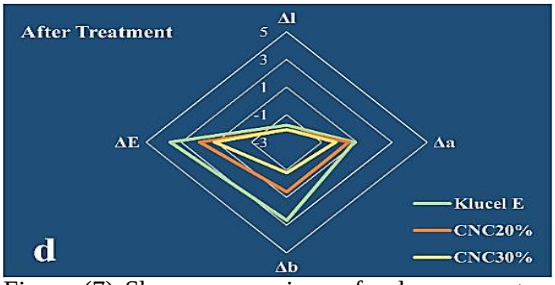


Figure (7) Shows comparison of color parameters of treated wood samples before and after aging; **a.** Klucel E, **b.** nanocomposite with CNC 20%, **c.** nanocomposite with CNC 30%, **d.** all materials after all impregnation process.

3.8. *FTIR analysis*

Figure (8) shows the spectra of the filler, Klucel E and wood using FTIR, and the main bands are mentioned in tab. (1). Cellulose is the main component in the three materials and most bands appear in the three samples. A strong band is attributed to hydrogen bond at (3350-3450 cm^{-1}), and the absorption band at (2925-2923 cm^{-1}) is due to CH_2 and CH stretching vibration [42]. The (O-H) bending at 1640 cm^{-1} is attributed to water absorption [43]. The band at 1431 cm^{-1}, which is assigned to amorphous and crystalline cellulose, appears at 1429 cm^{-1} in wood, 1457 cm^{-1} in Klucel E and at 1435 cm^{-1} in CNC. The shift to a higher wavelength indicated that the amorphous area of the cellulosic component formed new and larger crystalline areas or crystals [44]. The observed bands at 1321 cm^{-1} and 1371 cm^{-1} are due to C-H (bending) in cellulose, hemicellulose and lignin [45] and the bands around 1059 and 1035 cm^{-1} were assigned to C-O stretching vibration. Three bands observed in the wood spectra, fig. (9) are related to the presence of hemicellulose and lignin; first absorption band at 1741 cm^{-1} is due to the carbonyl group of acetyl ester in hemicellulose and carbonyl aldehyde in lignin [46], the second band is at 1511 cm^{-1}, and arises purely due to aromatic skeletal vibration (C═C) in lignin, while the third band is at 1272 cm^{-1} for C–O stretching in lignin and C–O linkage in guaiacyl aromatic methoxyl groups [47, 48].

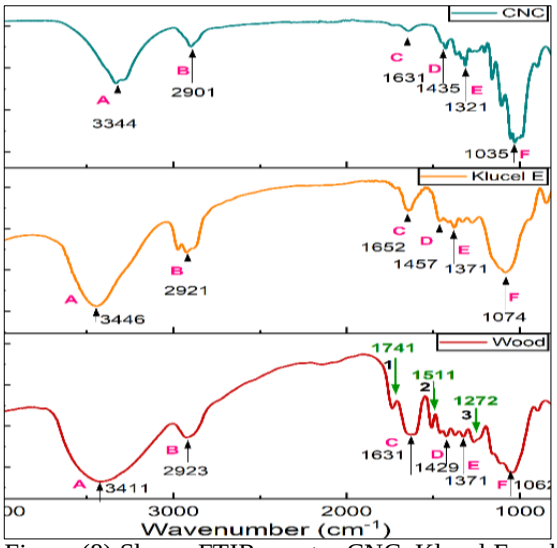


Figure (8) Shows FTIR spectra CNC, Klucel E and wood

Table (1) Assignments of characteristic absorption bands of wood, Klucel E and CNC

	cm^{-1}	Assignment	Reference
A	3347	A strong hydrogen bonded (O–H) stretching absorption	[47]
B	2925	C- H stretching vibration	[49], [50]
C	1640	Water absorption (O-H) bending and C=O stretching mode	[43]
D	1426	CH_2 bending in cellulose	[51], [44]
E	1375	C-H (bending) in cellulose, hemicellulose and lignin	[45]
F	1052	Cellulose, hemicellulose (C_3-O_3H stretching)	[52]
1	1734-1738	C=O stretching in unconjugated ketones aldehydes and carboxyl in wood, C-H bending in CNF	[45], [52]
2	1506-1510	C=C stretching of the aromatic ring (lignin)	[48], [53]
3	1268- 1272	C–O stretch (lignin) and C–O linkage in guaiacyl aromatic methoxyl groups in wood O-H bending and C-H wagging in Klucel E	[48], [50]

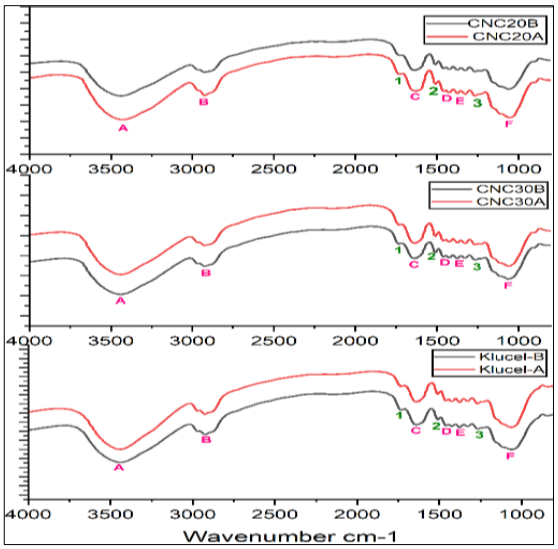


Figure (9) Shows FTIR spectra for consolidated specimens with composites CNC 20%, CNC 30% and pure Klucel E (*black line = before aging* & *red line = after aging*)

3.9. *Compression strength test for wood samples*

Compression strength was measured for wood samples after their exposure to accelerated aging for 300 hours. The compression strength of samples treated with Klucel E did not show significant increase in strength compared to standard wood specimen as it reached 30.2 MPa. However, compression strength significantly increased after treating wood with Klucel E/CNC. An increase was recorded after using nanocomposite with CNC 20 % which reached 31.42 (MPa), while increase was more observable in samples treated with CNC 30% nanocomposites reaching up to 35 MPa, fig. (10).

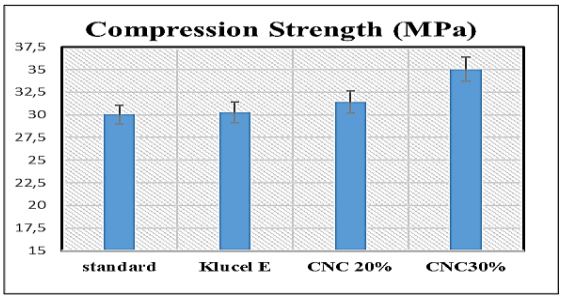


Figure (10) Compression strength for wood specimens consolidated with Klucel E, composites with CNC 20 and 30%

4. Discussion

The resulting diameter of the CNC was slightly bigger than previously reported values, which were an average of 35 nm [54, 55]. The slight increase in the particle size of CNC in 30% concentration was not significant where the average particle size of the small aggregates formed was still less than 100 nm, and the size of aggregates was proportional to CNC content in the nanocomposite films. Despite of the small aggregates of nanocellulose in the CNC 30% film it still showed the best results, in addition to the 20% concentration. Also SEM micrographs showed that the aggregates did not influence the distribution of CNC particles in Klucel E as the sample with 30% concentration demonstrated almmost even distribution of CNC on Klucel E film surface. Concentration 50% aggregation resulted in ruining the uniform dispersion of CNC particles leading to an unwanted structure. The XRD spectra showed a significant increase in the crystallinity of the nanocomposite and indicated the interactions between CNC particles and Klucel E chains, which influence the structure significantly. The mechanical properties of any material are heavily dependent on its crystallinity [56], and this was clearly noticeable at concentrations from 10% to 30 %. The addition of CNC had a crucial influence on the tensile strength of the films, because nanomaterials can form intermolecular interactions between the filler and polymer matrix resulting in better stress transfer from the polymeric matrix to the filler and consequently improving the tensile strength. The interactions between CNC particles themselves represent another crucial role in enhancing the mechanical properties. Therefore, a network of physical bonds is formed inside the composite because of the CNC-HPC and CNC-CNC interactions; and this network induces a significant improvement in the tensile strength of the polymer because it is formed mainly from hydrogen bonds, which is strong enough to influence the mechanical properties [57]. The agglomeration at 50% CNC resulted in stress concentration in certain parts within the composite and consequently resulting in poor transfer of the stress from the polymer to the filler, causing a decrease in the tensile strength at the end. Increase in elastic modulus until 30% concentration is an indicator for the raise of stiffness. This positive effect of CNC on the stiffness may be attributed to the increasing crystallinity of the composite. Hardness is the resistance of the composite to mechanical indentation or deformation, and the elastic modulus is used as an indication of the stiffness and strengths of the materials; when the stiffness and rigidity of the sample

increases, the elastic modulus increases [58, 59]. Hardness of the tested samples showed the same trend as elastic modulus. Similar to elastic modulus, the incorporation of rigid reinforcing nano-filler in the polymeric matrix increases the hardness of the composite. Hence, the presence of CNC in the polymer matrix increased the hardness of the composite. As indicated from SEM micrographs, CNC showed full dispersion up to 30% CNC, at which the beginning of agglomeration became obvious. Therefore, the elastic modulus of the composites with concentrations up to 30% improved greatly when compared with pure polymeric material. Yet the elongation at break decreased with the increasing content of CNC. Decrease in elongation may be attributed to that the CNC restricts the segmental motion, which causes the polymer chains to break at a lower value. The FTIR spectra of wood sample that was treated by CNC/Klucel E nanocomposites or pure Klucel E after accelerated aging compared to standard wood sample shows that no change in the peaks was observed, which means no change in functional groups due to chemical reaction could be observed between wood components and reinforcing materials. Furthermore, no chemical change was observed after exposing the treated wood to the aging process proving the stability of wood samples that were treated with CNC/Klucel E nanocomposites. This explains the results obtained from the color change measurements and proves that there is no change in chemical composition of wood samples. The tested nanocomposites had no noticeable changes in color on consolidated wood samples, when parameters of "after treatment" and "after aging" are compared, which proves that the composites have almost the same stability of pure Klucel E in different humidity and temperature conditions. The ΔE value decreased with the addition of CNC, because CNC causes the dissolved Klucel E to turn from colorless into pale white which reduces the effect of darkness in Klucel E after application on wood. The low viscosity of CNC resulted in the reduction of the composite viscosity and raised the ability of the polymer to penetrate the wood specimen, increasing the weight of the samples, especially the samples that had been treated with CNC composite 30%; and that was reflected in the compression strength test of the treated wood samples. Samples consolidated with CNC 20 % recorded 31.42 MPa and samples treated with CNC 30% reached up to 35 MPa whereas samples consolidated with Klucel E recorded only 30 (MPa).

5. Conclusion

This work has addressed the ability of CNC particles to enhance the mechanical properties of Klucel E (hydroxypropyl cellulose polymer) and attempted to determine the ideal concentration of CNC in the nanocomposite that result in the best enhancement. The films of five samples of the nanocomposite were prepared, characterized and had their mechanical properties tested. All samples showed an increase in the degree of crystallinity. The samples had good dispersion in the matrix for concentrations less than or equal to 30%, while sample with concentration of 50% had big agglomeration and bad dispersion. Although all samples showed a significant enhancement in the mechanical properties, yet the sample with 30% concentration gave the best results in mechanical strength and samples with 50% showed the least enhancement. The results indicated that the Klucel E/CNC composite with a concentration of 30% is an ideal candidate for use in the conservation of wooden artifacts as it shows the highest mechanical strength enhancement alongside with acceptable crystallinity and dispersion. The above mentioned tests also gave a clear indication that the addition of CNC, as a filler, to Klucel E can have a significant influence of the strength of Klucel E. This influence increases up to a concentration of CNC 30 %. The filler also enhanced the color change caused by Klucel E 3% diluted in alcohol 70%, reduced the viscosity, and improved the penetration into the sample which resulted in enhancing the compression strength of wood samples especially in nanocomposites with CNC 30%. This increase was due to the

interactions between wood and reinforcing materials alongside with easy penetration into wood samples. Finally, no chemical change was observed in wood samples in addition to high stability after exposure to 300 hours ageing. Using cellulose nanocrystals (CNC) as a filler in composites has led to interesting results which will presumably prove successful in archaeological conservation after further testing on different types of wood.

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