Nanocellulose Based Hydrogel for Cosmetic Application

A dissertation submitted in the partial fulfillment of the requirement for the degree of **Bachelor of Science by Research** in **Chemistry**

> Submitted by: Aarsi Gupta (SAP ID: 500082156)

Under the supervision of Guide- Dr. Bhawna Lamba Co- Guide- Dr. Tridib Kumar Sinha



Department of Chemistry, Applied Science Cluster School of Engineering University of Petroleum and Energy Studies Dehradun, Uttarakhand-248007, India May, 2024

DECLARATION

I declare that the thesis entitled "Nanocellulose Based Hydrogel for Cosmetic Application" has been prepared by me under the supervision of Dr. Bhawna Lamba and Dr. Tridib Kumar Sinha from Department of Chemistry, School of Engineering, University of Petroleum & Energy Studies, Dehradun, India.

Aarsi Gupta

Department of Chemistry School of Engineering University of Petroleum & Energy Studies Dehradun, Uttrakhand-248007, India

CERTIFICATE

I certify that, Aarsi Gupta has prepared her project entitled "Nanocellulose Based Hydrogel for Cosmetic Application" for the award of B.Sc. Chemistry by Research, under our guidance. She has carried out the work at the Department of Chemistry, School of Engineering, University of Petroleum & Energy Studies, Dehradun, India.

Dr. Bhawna Lamba

Department of Chemistry School of Engineering University of Petroleum & Energy Studies Dehradun, Uttrakhand-248007, India

Dr. Tridib Kumar Sinha

Department of Chemistry School of Engineering University of Petroleum & Energy Studies Dehradun, Uttrakhand-248007, India

PLAGIARISM CERTIFICATE

I, **Aarsi Gupta**, herby certify that the research dissertation titled "**Nanocellulose Based Hydrogel for Cosmetic Application**" submitted for the partial fulfillment of a B.Sc. chemistry by research degree from University of Petroleum Energy & Studies, Dehradun, India is an original idea and has not been copied/taken verbatim from anyone or from any other sources.

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Aarsi Gupta Department of Chemistry School of Engineering University of Petroleum & Energy Studies Dehradun, Uttrakhand-248007, India

Approved by:

Dr. Bhawna Lamba Department of Chemistry School of Engineering University of Petroleum & Energy Studies Dehradun, Uttrakhand-248007, India

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Abstract

The use of nanocellulose-based hydrogels in cosmetic products has attracted significant attention due to their impressive characteristics and compatibility with living organisms. In this work, nanocellulose extracted from waste paper using a modified facile technique including the synergistic influence of mechanical, chemical and cryogenic methods. As per the particle size analyser, the nanocellulose was found to have average size within 20 nm. Nanocellulose, because of its large surface area, considerable water absorption and retention

realise, because of its large surface area, considerable water absorption and retention property laid by its abundant polar functional groups, biodegradability, non-toxicity is being considered suitable for diverse healthcare applications e.g. in-vivo, in-vitro drug delivery. Herein, the efficacy of the nano-cellulosic hydrogel has been examined for its controlled release of different ingredient of skin care importance e.g., silver nanoparticle, ascorbic acid or vitamin C to realize its potential future application in skin care industry. Considering the molecular like mildness i.e. Likes-like-Likes, all the important ingredients i.e. silver nano particle, ascorbic acid or vitamin C were found to easily absorbed and well incorporated within the hydrogel matrix. As per the UV-Vis spectroscopic study, copious time was observed for their release from the hydrogel matrix which may be considered advantageous for their controlled release for different skin-care applications. In realm of current scenario of research and development, industrial transformation, this work including the large-scale production of nanocellulose, and its plausible skin care applications may consider praise worthy for both the academia of broader research interest and the industry.

Key words: Nanocellulose, Hydrogel, Cosmetics, Waste-to-Wealth

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CHAPTER 1

1 INTRODUCTION

As our society progresses towards a technology-driven paradigm, the significance of environmental concerns amplifies amidst the pursuit of convenience, comfort, and safety. The arrival of modern technologies, spanning information technology, biotechnology, and nanotechnology, has profoundly shaped various dimension of our society. Nevertheless, the trajectory of future technological advancements must pivot towards prioritizing symbiosis with our natural environment, thereby embracing sustainability as a foundation of innovation. Several studies regarding the forecast and production of nanocellulose have been published by various organizations and companies such as Allied Market Research, Future Market Insights, and TAPPI [1]. As nanocellulose is one of the renewable, eco - friendly and sustainable material which do not impact on the environment and substitute the use of many toxic materials. Assessing market size and value propositions is crucial for maximizing economic potential in cellulose-based production. In the context of commercialization, around 1500 tons of bacterial nanocellulose (BNC) are sourced from nata de coco farms in the Philippines and Indonesia. Ten organizations were identified as capable of manufacturing cellulose nanofibrils on a commercial or pre-commercial level. These entities include the Forest Products Laboratory (FPL) in collaboration with the University of Maine, Paper logic, Borregaard from Norway, Innventia in Sweden, American Process in the USA, Nippon Paper, Oji Paper from Japan, Holmen Paper in Sweden, CTP/FCBA in France, among several others [2] Conducting life cycle assessments (LCA) and techno-economic assessments (TEA) during the initial stages of production route development could effectively tackle these hurdles. These assessment studies aid in pinpointing opportunities for process enhancement to boost material and energy efficiency. While some life cycle assessments studies are accessible in the public domain, techno-economic assessments investigations for cellulose nanocrystals and especially cellulose nanofibres remain relatively rare [3], [4] There is high demand of cellulose as it is used in so many applications such as packaging material, membranes, biomedicine, continuous fibres and textiles, polymer reinforcement, superabsorbent material, flexible displays, paper based diagnostic device utilizing nanocellulose, printed electronics and energy storage device. This work provides an overview of diverse nanocellulose sources and types, highlighting various production methods including pre-treatment, mechanical, chemical approaches, and hybrid

method supplemented with recent literature findings. The aim is to elucidate the process by which cellulose fibres break down into their constituent nanocellulose components [5] Furthermore, this study delves into the distinctive attributes of nanocellulose and underscores the growing interest in eco-friendly products, which has spurred significant research and industrial focus on its applications. However, the widespread adoption of nanocellulose, encompassing both cellulose nanocrystals (CNCs) and nanofibrils (CNFs), remains predominantly limited to laboratory settings. This limitation stems from challenges related to scalability, energy requirements, time-intensive processes, and elevated production expenses[6]. This study examines the available literature and outlines the principal findings while identifying areas for further investigation and research.

2 Nanocellulose- Over the past decade, nanocellulose has garnered significant attention, evident in the proliferation of patents and scientific literature exploring its properties, manufacturing techniques, and prospective applications [7][8][9]. As nanocellulose is the part of cellulose, it is a naturally abundant and environmentally friendly material that holds significant potential for mitigating the detrimental effects of petroleum-based synthetic polymers. Employing cellulose as a reinforcing agent in the production of synthetic matrices or indigenous starch matrices presents a persuasive approach to reduce the environmental impact stemming from hazardous emissions at every stage of polymer goods' life cycle [10]. Cellulose functions as the predominant structural element found in the cell walls of plants, providing structural support for plants and aiding in maintaining their rigidity and upright posture. Consequently, cellulose can be obtained through extraction from various sources, including plants, agricultural residues, animal byproducts, and bacterial pellicles [11]. Additionally, nanocelluloses are derived from plentiful and sustainable biomass sources [12] It characterized by cellulose particles having at least one dimension below 100 nm and it collectively gathered significant attention as an intriguing biopolymer. This is evidenced by the extensive body of scientific literature and substantial industrial investments dedicated to its exploration and development [13]. Extensive research on nanocellulose has indicated significant potential in various application areas such as packaging material, membranes, biomedicine, continuous fibres and textiles, polymer reinforcement, superabsorbent material, flexible displays, paper based diagnostic device utilizing nanocellulose, printed electronics and energy storage device [14].

2.1 **Characteristics of Nanocellulose-** Nanocellulose is distinguished by its exceptional material properties, including remarkable mechanical strength, a high surface area of 250 m/g, thermal stability, a high aspect ratio, intriguing optical characteristics, versatile rheological properties, and the ease of surface modifications [15]. The evaluation of nanocellulose's surface charge can be conducted by analysing its zeta potential value. The presence of negatively charged hydroxyl groups (-OH) on the surface of nanocellulose is indicated by negative zeta potential values. Furthermore, insight into dispersion stability can be derived from these zeta potential values. Zeta potential values below 15 mV typically suggest the beginning of agglomeration, while values above 30 mV result in a stable suspension of nanocellulose [16], [17]. The robust interaction capability of the hydroxyl groups present on the surface of nanocellulose facilitates self-association. This attribute renders nanocellulose an excellent reinforcing agent within polymer composite matrices, resulting in a remarkable enhancement in mechanical strength, typically ranging from ten-fold to hundred-fold, alongside improved optical transparency compared to polymer matrices devoid of nanocellulose integration [18], [19].

3 Preparation of Nanocellulose- In this section, an overview of diverse techniques employed for the preparation of nanocellulose is given.



Figure-1 Schematic representation of different techniques to produce nanocellulose

3.1 **Pre- Treatment Method-** There are two types of pre- treatment method namely-Enzymatic Hydrolysis and Alkaline- acid hydrolysis [20]. These methods are employed to modify the structure or properties of the cellulose material, making it more suitable for subsequent processing into nanocellulose. The specific pre-treatment method choice depends on factors such as the desired properties of the nanocellulose and the characteristics of the starting cellulose material. Utilizing pre-treatment methods on cellulose aims to minimize energy consumption during the nano-fibrillation process [21]. Here summarize the different pre- treatment methods in brief.

3.1.1 **Enzyme Hydrolysis-** Enzymatic hydrolysis is a biochemical process in which enzymes break down complex molecules into smaller units through the addition of water. In enzymatic hydrolysis, enzymes specifically target certain chemical bonds within the substrate

molecule and cleave them, resulting in the formation of two or more smaller molecules. This process is widely used in various industries, including food processing, biofuel production, pharmaceuticals, and waste treatment [22], [23]. Paakko et al illustrated a technique for synthesizing nano fibrillated cellulose (NFC) from bleached softwood pulp by employing a blend of gentle enzymes, namely Cellulases C and D, along with consecutive refining and homogenization procedures. This study presents a sustainable and effective approach to acquiring NFC, a versatile nanomaterial with diverse potential uses [24].

3.1.2 Alkaline- acid- (Bei Wang et al 2006) elucidate that one of the widely adopted methods for the isolation of cellulose nanofibrils (CNF) involves an alkaline-acid pre-treatment process and this method is renowned for its effectiveness in solubilizing lignin, hemicellulose, and pectin, thereby facilitating the subsequent mechanical process for the isolation of CNF [25]. This method involves three steps as follows:

3.1.2.1-Acid Treatment- An acid pre- treatment is employed to further delignify and remove any remaining non-cellulosic materials. Acidic conditions effectively hydrolyze the residual lignin and hemicellulose, resulting in a purified cellulose substrate primed for the isolation of CNF [26]. (Lei Wang et al 2022) summarize that Nanocellulose extraction was effectively achieved using a combination of phytic acid pretreatment followed by ultrasonication post-treatment from bleached chemical pulp and it shows 22.0 ± 12.3 nm in width, 5.5 ± 2.9 nm in height with a length ranging from 200 nm to 1 µm [26].

3.1.2.2- Sodium Hydroxide- In the pre-treatment process, the material undergoes treatment with a 2 wt% alkali solution for a duration of 2 hours at a temperature range of 60–80°C. This treatment serves to break down the linkages existing between the carbohydrate and lignin components [27].

3.1.2.3- Alkali treatment- Alkali pre- treatment not only aids in the purification of the cellulose material but also helps to improve its overall properties by reducing lignin content and enhancing cellulose purity. (Emma Mauren Moko et al) used alkali pretreatment to eliminate hemicellulose from the Giant Swamp Taro (GST) starch processing waste [28].

(Ayse Alemdar et al 2008) showed the alkaline-acid pre-treatment method has proven to be highly efficient, substantially increasing the cellulose yield from an initial rate of 43% to an impressive 84%[29].

Overall, the utilization of alkaline-acid pre-treatment represents a significant advancement in cellulose extraction techniques, leading to substantial improvements in yield and purity.

3.2 Mechanical Method of Extraction-Mechanical extraction of nanocellulose involves the physical breakdown of cellulose fibres from natural sources into nanoscale dimensions using mechanical forces. This process typically starts with the isolation of cellulose-rich materials, such as wood pulp, cotton, or agricultural residues like kapok husk, sugarcane bagasse etc. This method includes high pressure homogenization, ball milling, crycrushing, probe sonication, ultrasonication etc [30]. (Vern Yi Tan et al 2020) found that increasing ultrasonication time results in a reduction in the size of nanocellulose samples, accompanied by a concurrent elevation in the onset thermal degradation temperature. Conversely, nanocellulose obtained through crycrushing, particularly from samples subjected to a 48-hour freeze-drying process, exhibits a lower degradation temperature relative to those prepared from samples frozen for 24 hours. In terms of mechanical properties, the ultrasonication method demonstrates superior performance compared to crycrushing [31].

3.2.1 **Ball Milling Process-** The process of ball milling consists of immersing the cellulose suspension within a cylindrical vessel containing milling balls crafted from substances such as ceramic, zirconia, or metal. Rotation of this vessel occurs in both clockwise and counterclockwise directions, resulting in collisions between the balls and the cellulose fibers, thereby initiating the defibrillation process of the fibers [32]. Rafaela R. Ferreira et al reported that the nanocellulose (NC) was effectively extracted from eucalyptus sawdust through a process which involved both ball milling and high-intensity ultrasonication techniques and the use of ball milling, durations (ranging from 1 to 4 hours) consistently yielded cellulose particles at the nanoscale [33]. Jinyi Zhao et al asserted that the conversion of bamboo pulp to soluble saccharides and nanocellulose was effectively achieved via enzymatic hydrolysis, aided by ball milling. The maximum yield of Xylo oligosaccharides (XOS) was recorded at 21.6% when employing 90 minutes of ball milling and a xylanase loading of 300 U/g xylan. Furthermore,

the use of ball milling resulted in a significant decrease in the crystallinity index (CrI) of the material, with reductions of up to 26.0% observed [34].

3.2.2 **Crycrushing-** Ultrafine friction grinding utilizing a custom-designed disk has been employed by numerous researchers for the production of cellulose nanofibers. This process involves subjecting raw cellulose to both static and rotating grinding at 1500 rpm. As the grinding proceeds, the cell walls break down, leading to the fragmentation of hydrogen bonds by shear forces generated during the grinding process. This results in the production of individualized nanofibers from the cellulose pulp [35].

3.2.3 High Pressure Homogenization- It offers exceptional efficiency coupled with straightforward implementation, all without the need for organic solvents [36]. In this method, pre-treated cellulose pulp undergoes dilution with distilled water before being propelled through a micron-scale aperture within the impacting and homogenizing valve, utilizing elevated pressure (ranging from 69 to 207 MPa) and high velocity, all at ambient temperature [37]. Yihong Wang and colleagues reached a conclusion indicating that the application of the HPH process instigated alterations in the physical and morphological composition of cellulose, consequently resulting in the attainment of specific properties. Moreover, a notable reduction was observed in the weight average molecular weight, thermal stability of nanocellulose, and viscosity of cellulose/[Bmim]Cl solution following treatment via HPH [38]. Changling Wu et al. also produced nanocellulose components from okara with enhanced functional performance, including thickening, gelling, and heat stability, using the high-pressure homogenization process. However, there are drawbacks to High Pressure Homogenization (HPH), such as high energy consumption and fiber blockage. To address these drawbacks, pulp might go through pre-treatment or chemical purification. Furthermore, there's a chance that the crystalline structure of cellulose nanofibers (CNF) will sustain severe mechanical damage [39] [40].

3.2.4 **Micro fluidization-** Micro fluidization is a method used in fluid dynamics and microfluidics, mostly in biotechnology and nanotechnology. It deals with the small-scale manipulation and control of fluids, usually at the micro- or nano meter-scale.

In micro fluidization, fluids are passed through microchannels or small channels within microfluidic devices. These devices can precisely control the flow rate, pressure, and mixing of fluids, allowing for various processes such as emulsification, particle size reduction, mixing,

and chemical reactions to be carried out with high precision and efficiency [41]. Ana Ferrer et al reported that after undergoing chemical and mechanical treatments, cellulose nanofibrils were isolated from Empty Palm Fruit Bunch Fibers (EPFBF), this method included pulping soda, milox, and formosol, followed by mechanical micro fluidization, which allowed suspensions containing unique, long nanofibers to be produced. The breakdown of EPFBF into nano fibrillated cellulose (NFC) regularly resulted in the resultant films absorbing less water, which is an essential property for many applications [42].

3.2.5 **Grinding-** Another method for converting cellulose into nano-form involves grinding. In this process, the pulp is subjected to grinding between two stones. One of the stones remains stationary while the other rotates, generating shear force that effectively disrupts the hydrogen bonds within the fibres, leading to the breakdown of cellulose into nano-scale particles [43], [44].

3.3 Chemical Method of Extraction- In order to remove lignin from fibers, cellulose extraction uses a variety of chemical processes, including alkali retting, acid retting, chemical retting, chemical-assisted natural (CAN), and degumming. These techniques also affect non-cellulosic elements like as pectin and hemicellulose, which are additional components of the fiber microstructure [45], [46], [47], [48]. For instance, alkali or acid retting is a chemical extraction method known for causing minimal fibre damage [49]. Van Nang et al were used chemical treatments which were able to successfully extract cellulose nanocrystals (CNCs) from three distinct byproduct sources: rice husk (RH), coconut husk fiber (CHF), and the trunk of Nypa fruticans (NFT). For NFT, CHF, and RH, the measured CNCs showed high levels of crystallinity at 76.6%, 79.3%, and 82.8%, respectively. They also discovered that the yield of nanocellulose synthesis was about 30–40% by weight, with variations based on the parent material (NFT: 38–40%; CHF: 30–32%; RH: 35–37%) [50].

4 Application of Nanocellulose



Figure-2 Application of nanocellulose

Nanocellulose's remarkable thermal, mechanical, and optical qualities make it a promising material with a wide range of potential applications. Because of its high tensile strength and barrier qualities, which set it apart from typical plastics, nanocellulose has been thoroughly studied in research for its potential in sustainable packaging materials [51]. For example, El-Wakil et al assessed the antibacterial activity of coated sheets against Saccharomyces cerevisiae, Staphylococcus aureus, and Escherichia coli and they also concluded that the coated kraft paper with WG/CNC 7.5%/0.6% TiO2 showed outstanding antibacterial activities for three layers of paper after two hours of exposure to UVA light illumination. They did not, however, confirm the paper's antimicrobial efficacy with food systems in real-world storage situations.[52]. Furthermore, because nanocellulose is easily functionalized for targeted drug release and is biocompatible [53], it has demonstrated considerable promise in biomedical applications, spanning from drug delivery systems to scaffolds for tissue engineering. Additionally, in the field of environmental remediation, sensors, materials based on

nanocellulose have been studied for their ability to absorb contaminants from soil and water, offering a possible way to reduce pollution [54] [55]. Muhammad Zubair et al conclude in their paper that materials generated from nanocellulose have exhibited exceptional efficacy in eliminating a wide range of contaminants, including organic and inorganic pollutants from wastewater. When it comes to materials used for water and wastewater cleanup, nanocellulose is an affordable, sustainable, and renewable option. The remarkable adsorption capabilities of nanocellulose were demonstrated in the elimination of salts, dyes, and heavy metals [56].

CHAPTER 2

EXPERIMENTAL DETAILS

Research Gap: From the literature survey, these research gaps were identified.

- 1- Nanocellulose is hard to process in bulk
- 2- High-cost production of nanocellulose

Objective: Focusing the existing research gaps, following objectives are framed for the current research works for providing a cost-effective solution:

- 1- Extraction of nanocellulose from waste material (sugarcane bagasse & wastepaper)
- 2- Formation of nano cellulosic hydrogel
- Inclusion of nanoparticles (e.g., Ag°, TiO₂, etc.) and other chemicals (e.g., vitamin C) especially in cosmetic applications

Material & Methods:

Locally accessible lignocellulosic resources i.e. Sugarcane bagasse was gathered from the local vender then organized and purified it. Subsequently, it was undergoing manual cutting into small fragments after dried in the sunlight. Then dried it into the oven at 65 °C followed by grinding. The waste paper was collected from the office of the UPES and the other chemicals used in this investigation were sourced from SRL Chemicals and Molychem Chemicals, USA which include Sodium Hydroxide, Sodium Sulphate, Urea, Sodium Chloride, Ammonium per sulfate, Bis acryl amide, acrylic acid, Silver Nitrate, Ascorbic Acid. The vitamin C tablet was purchased from the local shop. The water used in this investigation was DI water.

Method

Delignification of cellulose from bagasse- The dried and ground bagasse (1gm) was first dipped into the 1.5 wt% and 2 wt% NaOH solution (100ml) and autoclaved it for 1 hour at 120 °C for removal of lignin. The residue was filtered and subsequently washed with distilled water several times until neutralization. This neutral residue was then dried into the oven at 100 °C to remove the lignin completely and hemicellulose partially.



Bagasse after drying in the oven at 100°C

Preparation of Nanocellulose via base hydrolysis (Sugarcane bagasse)- Firstly, the solution of Sodium sulphate and Sodium Hydroxide 1:4 ratio has been prepared, then chopped pieces of raw bagasse dipped into that solution and processed the mechanical stirring overnight. After that vacuum filtration and neutralization with distilled water has been done. The filtrate was deep feezed at -20 °C for lipolyzing then ball mill (1:10) for 1 hour. The sample which is obtained after ball milling that is nanocellulose which is further dialyse for sugar removal.



Bagasse after Filtration & deep freezing at -20 °C



Dialysis for sugar removal

Preparation of Nanocellulose (WastePaper)- Cellulose was isolated from wastepaper for this investigation. Wastepaper was cut into small pieces and dip into the water solution of urea and sodium chloride in the ratio [Urea (0.25): NaCl (0.125): Waste paper (1)] followed by overnight mechanical stirring and then processed probe sonication less than $-10 \circ C$. After that lyophilize it for 5 hours then ball milling at the ratio (1:10) for 1 hour. Nanocellulose was formed which is characterized by particle size analyzer.



Nanocellulose formed by wastepaper

Preparation of Hydrogel- The prepared wastepaper nanocellulose was dipped 500mg into 50 ml of water. Following that, 100 mg of ammonium per sulphate, 100 mg of acrylic acid, and one bead of sodium hydroxide were added to it. Next, 100 mg of bis acryl amide was added, and it was stirred continuously for 30 minutes at 65 °C while ammonium per sulphate was added four or five times during the stirring and a nitrogen atmosphere was present for the entire operation.

The same procedure was repeated with the nanocellulose formed by sugarcane bagasse but the hydrogel was not formed.



Hydrogel formed by NC of waste paper



Hydrogel not formed by bagasse

Incorporation of Silver nano particles, Vitamin C and Ascorbic Acid into Hydrogel- Silver nitrate solution (1 mg/ml) was added to the mixture during the hydrogel production, causing an in-situ reaction that resulted in the synthesis of Ag NPS hydrogel. The dried hydrogel was immersed in the vitamin C and ascorbic acid solution for an overnight period to incorporate these acids.



Hydrogel immersed in ascorbic acid solution



Hydrogel immersed in Vitamin C solution



Hydrogel with silver nanoparticles





Figure-3 Schematic Representation of processing hydrogel

CHAPTER 3

Results and Discussion

Results:

In Figure 3, the FTIR analysis of raw bagasse reveals a prominent lignin peak at 1605, a peak that is notably absent in the FTIR spectrum of pretreated bagasse. This observation suggests that delignification occurred during the pretreatment process; however, the formation of hydrogel did not take place, possibly attributed to the existence of sugar. The size of nanocellulose nanoparticles derived from wastepaper was confirmed using a particle size analyser showed in fig.4 and the hydrogel that had been dried and combined with silver nanoparticles, ascorbic acid, and vitamin C was dipped into a 50ml of water. The hydrogel's delivery time was then repeatedly monitored using UV-Visible Spectroscopy over a period of 10 minutes. This showed that the absorbance increased over time, indicating that the concentration of these acids and silver nanoparticles increased over time. However, after a while, there was a saturation that indicated the maximum concentration of acids and nanoparticles were released into the water, leading to the occurrence of equilibrium. Subsequently, the hydrogel was removed from the water and placed in fresh water, and the delivery time of the hydrogel was once more observed using the UV-Visible Spectroscopy which showed the less absorbance of vitamin C, ascorbic acid and silver nano particles with the previous one which indicates that the maximum amount of the acids and NPs were released in the previous water and only certain amount of these acids and NPs were left which were released later in the fresh water.



Figure-4 FTIR of raw bagasse and pre-treated bagasse

Size Distribution by Intensity



Figure-5 The particle size of nanocellulose is 13 nm



Figure 6- UV-Visible spectroscopy peak of Ascorbic Acid



Figure 7- UV-Visible spectroscopy peak of Vitamin C



Figure 8- UV-Visible spectroscopy peak of silver nano particles

Conclusion

Hydrogel may have failed to be derived from sugarcane bagasse through a mechano-chemical approach possibly because of the existence of sugar. The synthesis of nano cellulosic hydrogel was successfully carried out utilizing cellulose sourced from wastepaper via a refined and standardized mechano-chemical process, capable of being expanded for large-scale manufacturing. Investigation was conducted on the development of nano cellulosic hydrogel incorporating vitamin C and silver nanoparticles.

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