70(10A): 108-115; 2022. DOI: 10.18311/jmmf/2022/31060

Print ISSN : 0022-2755

Journal of Mines, Metals and Fuels

Contents available at: www.informaticsjournals.com/index.php/jmmf

Biodegradable Nano-Cellulose and its Composite Materials for Food Packaging Applications: A Review

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Abstract

Bagasse is a fibrous material obtained after crushing sugarcane to extract its juice. this sugarcane bagasse can be utilized to produce cellulose nano-crystals for various applications. Researchers have been studying the manufacturing of nano-cellulose-based products and food packaging films. the evolution of environmentally friendly and ecologically balanced food packaging materials has gotten a lot of interest as a potential solution to partially replace the perishable fossil fuel-derived plastic. Nano-cellulose and so its uses have lately received considerable attention across both research and application areas due to their attractive characteristics such as exceptional mechanical properties, larger surface area, rich hydroxyl for alteration, and biological properties with 100 per cent environmental protection. It is widely produced around the world in big quantities. It is a sugar industry waste product. It is most widely employed in the paper industry, although researchers have proposed that various mechanical and chemical treatments can aid in the extraction of cellulosic fibres, pure cellulose, cellulose nano-fibers (CNF), and cellulose nano-crystals (CNC). These extracted components have a wide range of uses in the manufacturing of regenerated cellulosic fibre and composite materials. The extraction processes for these extracted components in food packaging are discussed in this review study, as well as their usual application in composite industries.

Keywords: *Sugarcane Bagasse, Cellulose nano-crystals (CNC), Cellulose nano-fibers (CNF), Food packaging, chemical treatments*

1.0 Introduction

SCB (sugarcane bagasse) is a widely available agro-waste that has been employed in a variety of applications, with its use as a source of cellulose drawing interest in biomedical and other fields. About half of the bagasse produced is used as a fuel to generate hotness and capability to run sugar factories, ethanol plants, and distillery plants, with the remainder being hoarded, posing an environmental risk due to the possibility of spontaneous combustion. Cellulose was, and continues to be, the most abundant biopolymer produced from all plant fibres, including agricultural waste. Despite its widespread use in the fibre, paper, film, and polymer sectors, the use of this natural biomass for the processing of novel material applications has recently piqued interest due to its ecological and renewable qualities. As a result, it is important to convert this waste into value-added products in addition to using it to produce fuel, chemicals, papers, and newspapers. It is a hot topic right now to use this garbage as a beginning material for new products. Nano-cellulose materials are produced from various plants consisting of cellulose [1]. Conventional synthetic polymers are now being replaced by environment-friendly materials like nano-cellulose. And all these have happened due to the

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growing interest in sustainable development. Nano-composites derived from cellulose nano-crystals generally tend to have good mechanical, barrier and thermal properties as compared to the traditional synthetic polymers. As a reinforcing agent, cellulose nano-crystals influence their own performance due to the effect of morphology and properties [2]. Nano-crystalline cellulose, nano-fibrillated cellulose, and bacterial nano-cellulose are the three kinds of nano-cellulose [1][3]. The majority of commercially available nano-cellulose is made from high-quality fibre sources such as wood pulp, dissolving pulp, and cotton. However, an increasing number of publications and attempts have demonstrated that it is also possible to use biological waste as a source to produce commercial-grade nano-cellulose [4]. Sugarcane bagasse is a fibrous substance that is mostly composed of cellulose. It is manufactured in vast quantities around the world. It is a type of waste material produced by the sugar mills. It is most widely utilised in the paper industry, although researchers have proposed that various mechanical or chemical procedures can aid in the extraction of cellulosic fibres, pure cellulose, cellulose nano-fibers, and cellulose nano-crystals [4]. Plants, animals, and microbes are the main producers of nano-cellulose, which is a cellulosic substance with one aspect in the nano-metre range. The production of nano-cellulose from different biomass origins for uses in a range of industries is receiving more interest. 3 tonnes of bagasse is produced for every 10 tonnes of sugarcane. So, and around nearly 54 million tonnes of dried sugarcane bagasse are produced per year in the entire world. This dried bagasse usually contains a quite large amount of cellulose which is utilized to produce cellulose derived products [5].

Hydrolysis of acid which is one of the production processes for nano-cellulose removes amorphous regions of the cellulose, leaving the crystalline regions in the colloidal solution. The stability of the suspension is due to the negative sulphate groups on the surface of the nano-fibers which occurs due to electrostatic repulsion [6]. Depending on the source of celluloses, cellulose chains cluster form micro fibrils with cross dimensions ranging from 2 to 20nm during production. Vander Waals forces and intra molecular and inter molecular hydrogen bonding are the primary mechanisms for aggregation [7]. The disadvantages of employing cellulose are that the nano-structures must be separated, and insertion of the crystallites in a matrix typically results in difficulties regulating the distribution level. A very well method for removing unstructured areas is acid hydrolysis of cellulose [8]. Plants generate roughly 75 million tonnes of cellulose every year, making cellulose biopolymers limitless. The usual formula for cellulose is $(C_6H_{10}O_5)n$, and cellulose is perhaps the most abundant in vegetable tissues. In the environment, cellulose is rarely found in its purest forms [9].

Although biopolymers are seen to be viable substitutes for traditional plastic materials, several of their qualities need

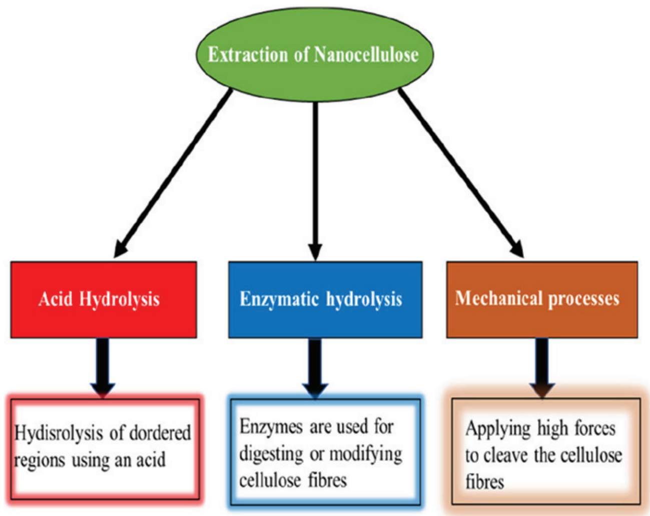

Figure 2: Extraction processes of nano-cellulose

to be enhanced before they can compete directly with fossil counterparts. Combining biodegradable polymers and/or introducing nano-fillers is an excellent technique to improve biodegradable polymer characteristics and, as a result, expand their fields of use [10]. Unfortunately, the majority of substances being used in food packaging are effectively non-biodegradable, posing a severe major ecological challenge. Novel bio-based polymers are often used to create beneficial and ecologically friendly films in an effort to extend shelf life and improve food quality while reducing waste generation [11].

Producing bio-nano-composite products with low filler particle loading has earlier been shown to be a viable technique for producing novel nano-structures with unique characteristics and excellent results for the packaging industry [12]. In the world of nano-materials, elongated nano-crystalline cellulose for bio-nano composite creation has sparked an interest.

Migration and porosity are essential issues in food packaging since no substance is completely resistant to atmospheric gases, liquid water, or natural substances present inside the packaged food or possibly even the packing materials themselves [13]. High barriers to movement or gaseous diffusion are undesirable in some situations, such as fruit and vegetable containers,where life span is critical is subject to the availability of a constant amount of air for continuous cellular respiration [13].

A superior mechanical properties nano-cellulose generated from acid hydrolysis of cellulose-fibres is known as nano-crystals or cellulose nano-whiskers [14]. It features a small rod-shaped or whisker-shaped form with a size of 2-20 nano-meters and a length of 100-500 nano-meters. It also has a chemical composition of 100 per cent cellulose, mostly in crystalline regions. Nano-fibrillated cellulose is a nano-

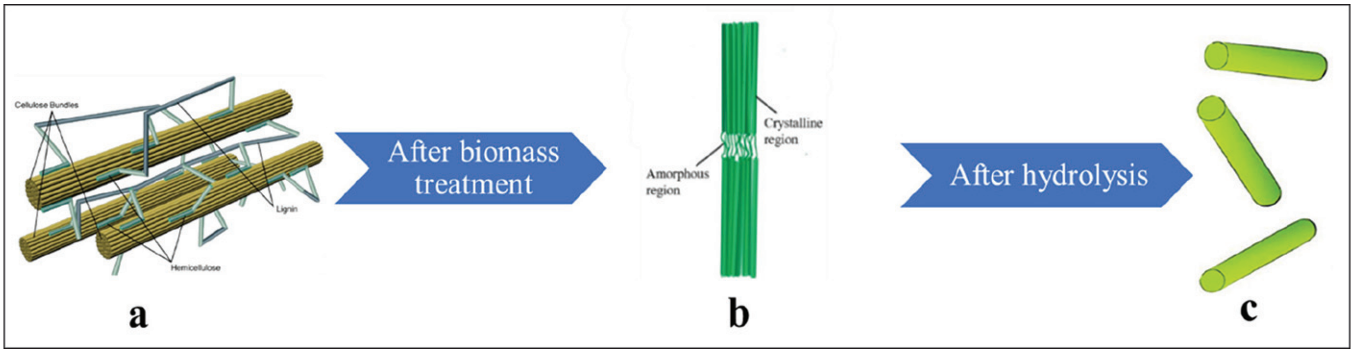

Figure 2: (a) Plant source, (b) Fibrils of cellulose, (c) Nano-crystals

cellulose that is a lengthy, elastic, and entanglement material which may be mechanically separated from cellulose micro fibrils. It is also referred to as cellulose micro fibril, cellulose nano-structured materials, cellulose nano-fibril, or nano-fibrous-cellulose. Long fibre forms have diameters ranging from 1-100 nm and lengths ranging from 500-2000 nm[14].

Ecological health and safety are two critical issues that nano-technology can successfully address. Nano-cellulose (NC) is made from lingo-cellulosic bio-mass by a mechanical or chemical method. The chemical process comprises structural disintegration, alkaline treatment, delignification, and acid hydrolysis, which results in rod-like nano-cellulose (CNCs) diameters range from 3-50nm, while length range from100-500 nano-meters. Compression and increased synthesis are standard methods for producing cellulose nano-fibrils (CNFs) with dimensions ranging from five to fifty nano-meters in diameter and length ranging from 500 to 2000 nanometer [15].

Nano-scale separation and production of cellulose materials have been investigated extensively in the last ten years, and their use as reinforcing filler in the preparation of composites for emerging applications is a hot research field that is gaining some traction among bioengineering researchers [16]. This review attempted to highlight the various nano-cellulose extraction methods, their qualities, and potential uses in food packaging.

Acid hydrolysis, ultrasonic procedure, and enzymatic hydrolysis are some of the ways that can be utilised to create nano-cellulose. Acid hydrolysis is the most extensively used procedure. This technology makes nano-cellulose with better properties that may be made easily and quickly. According to certain studies, the crystalline index of nano-cellulose produced by acid hydrolysis was higher than that of nano-cellulose produced by other methods. The acid hydrolysis-produced nano-cellulose is also smaller in size. The acid hydrolysis approach was chosen to create nano-cellulose for these reasons [17]. The manufacturing of CNCs mediated by strong mineral acids (such as H_2SO_4) is hampered by a number of significant problems, including large amounts of contaminated effluent, substantial corrosion risks to the facility, and low yield [18].

The use of cellulose nanostructures as components of materials for a range of purposes, including food packaging, has been extensively researched. In nano-composites, they are frequently used as are enforcing phase [19]. Food packaging materials sourced from non-renewable fossil fuels confront disposal and recycling challenges. The issue of migration and permeability in food packaging is crucial. No substance is completely impermeable to environmental gases, liquid water, or natural substances present inside the packed food or the wrappings itself [20]. High migratory or gas passage barriers are unfavourable in some uses, also including fresh fruits and veggies boxes whose storage time is reliant on continual oxygen supply for sustaining cellular respiration [20].

Antimicrobial food packaging was developed in response to the rising need for longer shelf lives for fresh foods and the necessity for protection against food borne infections. The mix of organic and inorganic packaging, i.e. metal nano-particles embedded in polymers, is shown as one of the most efficient ways. According to the Centre for Disease Control and prevention (CDC) food borne illnesses sicken 76 million people each year in the United States, 325,000 of whom are admitted to hospitals, and 5000 of whom die [21].

In the food sector, synthetic antioxidants are commonly employed to prevent food from oxidising. Butylated hydroxyl anisole (BHA) and butylated hydroxyl toluene (BHT) are examples of synthetic antioxidants (BHT) are commonly utilized in food products [22]. Finding appropriate techniques to increase the barrier qualities it is extremely difficult to recycle plastics utilized in flexible food containers. Metallic coatings, such as aluminium, are used as air or liquid water shields in current research, although they have intrinsic limitations such as breaking and serious health concerns [23].

2.0 Properties

Anuj et.al [1] extracted CNCs from acid hydrolysis process using sugarcane as raw material and studied regarding the morphology, topography, structural, elemental and thermal properties. In their investigations they found that CNC's exhibited lower thermal stability and higher crystalline nearly 72.5% as compared to native cellulose which is 63.5%. They also studied the characterization like morphology and topography of the nano-crystals with the help of FE-SEM, TEM and AFM. Among these studies SEM and TEM studies showed CNC's in nano-scale range. Also CNC's were seen as rod like structures whose sizes ranged from the length is between 250 and 480 nm, while the diameter is between 20 and 60nm.

Hudson Alves Silverin et.al [2] extracted cellulose nano-crystals using corncob as raw material from acid hydrolysis process. They extracted CNC's at different time intervals and then labelled them as CNC90, CNC60, CNC30 based on the time of extraction. The index of crystalline thermal stability, shape of the resultant cellulose nano-crystals of corncob were all measured, and their reinforcing capacity was assessed PVA (polyvinyl alcohol) was used as the polymeric matrix. They even found that the CNC60 had a needle like structure, strong crystalline (83.7%), better thermal stability (about 185°C), and a diameter of 4.15 ±1.08 nm and an average length of 210±44.2, yielding the L/D aspect ratio of roughly 53.4±15.8. During their experimentation they found that when just 9% (wt.%) CNC60 was added, the CNC/PVA composites had dramatically enhanced tensile strength to nearly to 140.2%. It was seen that, there was CNC dispersion in a polymer matrix that is homogeneous by CNC's inclusion into PVA as it had no effect on the polymeric matrix's transparency or homogeneity.

In their review study, Youssef Habibi et al [7] state that nano-cellulose's nano-scale size and amazing mechanical capabilities make it a great choice for increasing the host material's mechanical characteristics. They also claim that cellulose nano-crystals have a Young's modulus that is comparable to Kevlar and potentially stronger than steel. Coming to the thermal properties Habibi et.al reviewed that adding unmodified CNs to semi-crystalline polymers has no effect on the melting temperature (Tm) of the nano-composites.

El Achaby et.al [10] obtained CNC from SCB (Sugarcane bagasse) through acid hydrolysis. They used a solvent evaporation approach to create a carboxy methyl cellulose/ starch polysaccharide matrix strengthened with nano-cellulose bio-nano-composite film. They discovered that the influence of nano-crystals on the transparency, liquid water permeation, and tensile parameters of bio-nano-composite coatings was examined. It has been revealed that bio-nano-composite coatings are transparency because to the dispersal of CNC at the nanometer. The integration of CNC reduced the water vapour permeation significantly, whereas the young's modulus and strength properties both rose progressively.

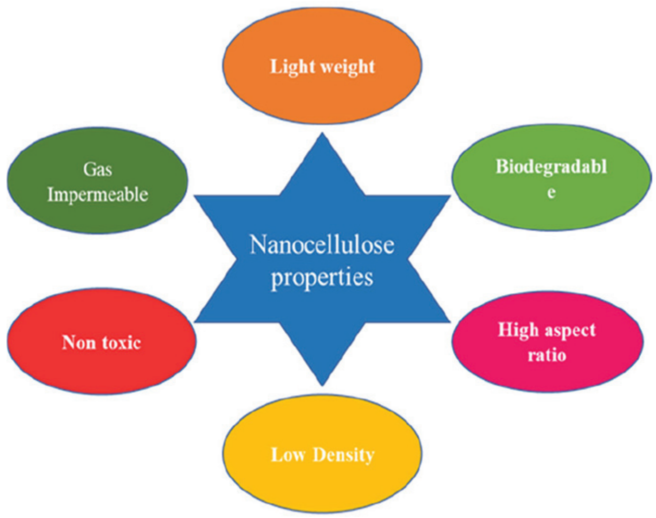

Figure 3: Some Unique characteristics of nano-cellulose

Mounir El Achaby et.al [12] extracted nano-cellulose from SCB (sugarcane bagasse) through acid hydrolysis. Using the casting/evaporation approach, bio-nano-composite films were created. The tensile modulus of the neat PVA film is 865.78MPa, a 49.80MPa ultimate tensile strength, a toughness of 23.44108 J/m^3 and a 64.98% elongation at break. The creation of linkages between the polymer chains of both PVA and CMC might explain these differences in PVA's tensile characteristics after it was blended with CMC.

Nassima El Miri et.al [21] extracted nano-cellulose from acid hydrolysis treatment. Here, the CNC was isolated from SCB to be used as nano-reinforcing agents using alkaline, bleaching, and acid treatments. The goal of this work was to use the solution casting process to create CMC/starch (ST) polysaccharides matrix containing nano-crystals reinforced bio-nano-composite films (CNC). From UV-Vis spectroscopy it was seen that because of its nano-metre-scale dispersion, the inclusion of CNC had no effect on the transparent of the CMC/ST mix material. The water barrier characteristics revealed that even when cellulose nano-crystals was introduced to a nano-composite mixture, the water vapour permeability of the bio-nano-composite coatings created enhanced. In addition, when the nano-crystal content was raised from 0.5 - 5.0 per cent by weight, the young's modulus and strength properties of the CMC/ST-CNC nano-composite film rose continuously in contrast to the CMC/ST blended film.

Julien Bras et.al [24] isolated cellulose whiskers from sugarcane bagasse kraft pulp by acid hydrolysis process.

Here the authors observed that mechanical properties of NR nano-composites revealed that when whiskers were added to rubber, the Young's modulus and strength rose significantly, but the strainat break dropped. From thermogravimetric analysis it was seen that the temperature at which the temperature at which NR/cellulose whisker nano-composite membranes with 10 per-centage cellulose whiskers began to collapse was somewhat less (265C) than those of clean natural rubber. Because bagasse whiskers have a lower start decomposition temperature than rubber, beginning disintegration of rubber/cellulose whisker nano-materials may take much longer. The temperature at which whiskers begin to degrade has been determined to be around 170 degrees celsius. Water vapour permeation (WVP) resistance is a must have feature among composites used in specialized packaging and other applications.

Amita Sharma et.al [25] used freeze-dries CNF from rice straw as a filler material and cellulose acetate as a matrix. Also, DMSO (Dimethyl sulfoxide) is used as a filler and polymer-solvent. A known quantity of CA solution was generated in several organic solvents (dichloromethane DCM, acetone, ethanol, dimethyl form amide DMF, dimethyl sulfoxide DMSO) for the optimization procedure. The optimal circumstances, defined as those that resulted in better transparency and tensile strength, were discovered to be 2 wt% CA matrix and 5 wt% CA-based CNFs. The addition of RSCNFs reduced the water absorption characteristics of CA films significantly. In comparison with clean CA films, CNFs improve the nano-composite films' thermal stability.

Na Yina et.al [26] prepared nano-composites out of bacterial cellulose and lotus root starch using aceto bacterxylinum bacteria. During their study, they found that low concentrations of lotus root starch had an impact on the production of cellulose obtained from bacteria. A comparison between bacterial cellulose and produced nano-composites was made. XRD results revealed that there was not much difference in the structure of the nano-composite as compared to pure cellulose alone. The tensile tests showed that there was an improvement in the mechanical property of the composite in contrast with the unmodified bacterial cellulose.

Rajinipriya Malladi et.al [27] in their work showed that the structure of CNC is dependent on the method, hydrolysis time, temperature, total acidity, and various acids like as hydrochloric, phosphoric acid etc. The author further mentions that it is described somewhere that cellulose is composed of highly crystallized and disordered unstructured parts. It is considered that during extracting, disordered amorphous are as are eliminated, leading to enhanced crystallisation. However, nano-cellulose's thermal stability falls when contrasted to raw material due to the inclusion of sulphate groups.

3.0 Nano-Cellulose for packaging

New packaging sectors have been among the most frequent consumers of nano-cellulose to substitute polymer composites derived from petroleum resources. Traditional food packaging materials are made from non-renewable fossil fuels and have a difficult time being disposed of and recycled. The development of environmental-friendly and ecologically balanced food packaging materials has gotten a lot of interest as a potential alternative to partially replace the perishable fossil fuel-derived plastic The use of non-renewable and non-biodegradable materials that cause environmental imbalance is quite widespread in packaging applications. As a result, there is a significant need for renewable and biodegradable packaging materials many scholars have recommended that biomass materials be used in packaging applications. Using several methods such as chemo-mechanical, cryo-crushing, and high-speed grinding, some researchers have effectively recovered the nano-cellulose of various natural fibres. Recycling process has gained popularity as a method of minimising the quantity of plastic rubbish in our ecosystem. This recycling approach, however, does not work on packaging material systems due to adultration of organic compounds in packaging material polymers. As a result, packaging makers must examine not only the factor of food quality preservation, but also environmental consequences of new packaging disposal.

Antioxidants, oxygen scavengers, flavouring, moisture absorbers, UV barriers, and antibacterial are just some of the activities that active food packaging may perform. However, active packaging that emits active biocide compounds into food has gotten a lot of interest because of its capacity to extend food's shelf life and reduce the prevalence of food borne germs. Antimicrobial activity can be achieved by incorporating active biocides into food products or the environment around them. The basic goal of antimicrobial packaging is to reduce and eventually eliminate the growth of spoilage germs. The use of antibacterial nano-particles in food preservation has become more common.

When compared to normal-sized Agparticles, AgNPshave a bigger surface area, which makes it easier to interact with microbial cells and achieves a more effective antibacterial activity[9]. Antimicrobial compounds in food packaging materials have recently attracted a lot of interest. Antimicrobial-active films may aid in the control of pathogenic and spoilage bacteria. Due to the nano-composite matrix's excellent structural integrity and barrier qualities, as well as the antimicrobial capabilities given by the natural antimicrobial agents embedded inside, an antimicrobial nano-composite film is especially appealing. The adsorption of Ag^+ from solution to 1 mg kg 1 level was achieved using nano-structured calcium silicate (NCS). The NCS-Ag composite that results has effective antibacterial activity at low silver levels as low as 10 mgkg1 and might be employed as in food packaging as an antibacterial agent [11]. High barriers to movement or gaseous dispersion are unfavourable in some situations, such as wrapping for fruit and veggies, where storage time is based on continual oxygen transport for continued cellular respiration.

Plastics used in packaging for carbonated beverages must have robust O_2 and CO_2 barriers to prevent oxidizing and decarbonisation of the drinking contents. CO_2 movement is far less of a challenge in other products than O_2 or liquid water transfer. Food items involve complex and unique packing capabilities as a result of this complexity, and the package industry's needs would only rise as food is carried over increasingly larger distances across suppliers and users [13].

Since no one pure polymers provides all of the mechanical and tribological qualities required for every potential food packaging, complicated multilayer film or composites are frequently used. In an application which requires ultra-high oxygenation barriers over a wide temperature range, for example, an increased oxygen barriers, water control material such as EVOH can be made up of two layers of a moderately hydrophobic polymers which includes polyethene [13].

Marilena Carbone et. al [21] reviewed that one of the researchers analysed that in Turkey deli meat preservation, edible pullulan films coated with nano-particles (AgNPs, ZnO NPs) and essential oils (EOs) such as oregano oil or rosemary oil showed antimicrobial activity. AgNPs and oregano oil consumable films were much more effective than Zno Nano-particles and rosemary oil safe to eat films, as per the findings. The conservation and expansion of the prolonged storage of orange juice was investigated using a reduced polyethene (LDPE) polymer matrix incorporating Ag and ZnO nano-particles. When combined with thermal treatment at pasteurised temperatures, this active-nano-composite proved to be an extremely excellent antimicrobial nano-material.

When it comes to fungus, the antibacterial response of LDPE including nano-silver was significantly higher as compared to certain other effective polymer matrices integrating nano-particles (yeast and moulds). Because of the antibacterial activity of AgNPs, the pasteurized temperatures of orange juice may be reduced by 10 degrees celsius [21].

The physical, barrier, mechanical, and antioxidant characteristics of mangopeel extract (MPE) were tested in fish gelatin films for active food packaging. The solution casting process was used to make films with three different MPE concentrations (1–5%). Water vapour permeability (WVP) and film solubility ($P>0.05$) were both lower edin MPE-containing films. MPE films with a high amoun to fMPE also had a more stiff andless flexible film formation [22]. Physical features of food packaging materials (particularly tensile and permeability properties) are critical for ensuring that packaged goods are adequately protected from external forces and that

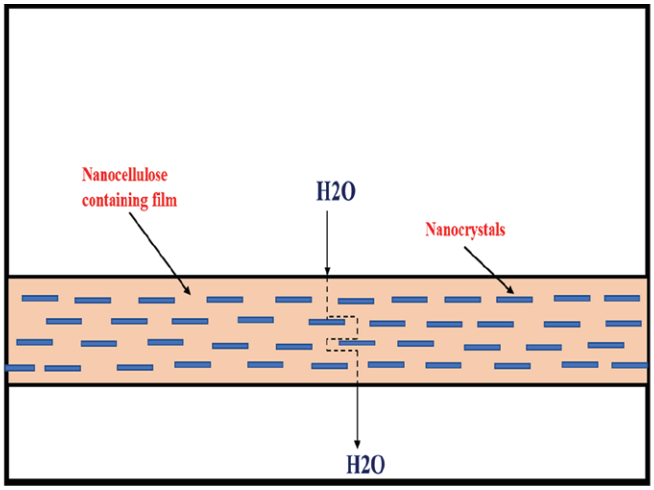

Figure 4: Water barrier property of CNC

deterioration is kept to a minimum. The physical qualities of materials used in foodpackaging are not governed by any legislation [19]. According to several research, packing sheets should have a tensile strength of at least 3.5 MPa.

FeiLiet.al [23] extracted cellulose nano-crystals by acid hydrolysis procedure. Coating dispersions were made by dissolving 8wt% CNC in distilled water and ultrasonically treating the mixture until it was visibly homogenous. Coated films were prepared according to ASTMD823-07. As per the results, the transparency value and haze readings of plain films were 87–92 per cent and 2.1–3.0 per cent, respectively, while coated materials had readings of 88–91 per cent, 2.1–3.0 per cent, including both, along with 3.3–4.0 per cent. Excluding one substrate, the coefficient of friction (COF) results indicate that the castings process generated a completely new cellulose nano-crystals coating layer. In addition to improved COF and outstanding optical qualities, empirical boiling water tests revealed that CN coated films offer excellent anti-fog properties.

Avelina Fernandez et al. [28] illustrate the known implementations of bio-catalytic in packaged food in their journal article and, additionally, inform some of the latest innovations reported in the literature to form continuous polymers and produce nano-structures with potential uses for the advancement of enzyme-based productive packaged food remedies.

4.0 Conclusions

Despite the reality that nano-cellulose has a variety of distinguishing characteristics and is plentiful in environment, obtaining nano-cellulose from biomass resources or cellulosic products still remains a significant challenge. Since hemicellulose, lignin and other challenge. Since hemicellulose, lignin and other components clump up together in the plant cell wall, biomass pre-treatment is required to cellulosic products still remains a significant challenge. Since hemicellulose, lignin and other challenge. Since hemicellulose, lignin and other components clump up together in the plant cell wall, biomass pre-treatment is required to remove any non-cellulosic compounds. Nano-crystalline cellulose is particularly intriguing because of its unique characteristics, such as controllable size, large specific surface area, self-assembling tendency, rod-like shape, and excellent dispersion, which are generated from natural cellulose fibres by controlled acid hydrolysis. The morphological characteristics of nano-structured materials are mostly determined by the cellulose source and extraction method. In the previous few decades, significant progress has been made in establishing methodologies and knowledge of nano-material uses, notably in therapeutic applications. To generate new generations of sophisticated nano-materials, there is a need for sustainable and readily available resources combined with modern technology. A lot of studies have recently been focused on the creation of improved cellulose nano-fibers with various morphologies and functional features. Medical implants, tissue engineering, medication delivery, wound repair, cardiology uses, and other therapeutic diagnoses all use nano cellulose fibres. As a result, nano-cellulose has emerged as a viable replacement for hazardous materials. The effectiveness of nano-cellulose as a packaging material has been examined in this study.

5.0 Acknowledgement

The authors would like to express their gratitude to the MSRIT for support.

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