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A Review on Properties of Nano Biocomposite Film for Packaging Applications from Cellulose Nano Fiber

Kritika Singh Somvanshi*, Prakash Chandra Gope**, Saurabh Tiwari***
**(Department of Mechanical Engineering, College of Technology, G.B. Pant University of Agriculture and Technology, Pantnagar-263145, Uttarakhand, India*
***(Department of Mechanical Engineering, College of Technology, G.B. Pant University of Agriculture and Technology, Pantnagar-263145, Uttarakhand, India*
****(Department of Mechanical Engineering, College of Technology, G.B. Pant University of Agriculture and Technology, Pantnagar-263145, Uttarakhand, India*

ABSTRACT
Today, food packaging is a significant sector in the production area, which is expanding day-by-day. Traditional plastics are well accepted in packaging applications. The major drawback of conventional plastics is their non-biodegradability. The intensive use of oil-based plastics has contributed to the fabrication of nano-biocomposites reinforced with Cellulose Nano Fibers (CNF), which is the most versatile biomaterial on earth. CNF is the most profuse, recyclable, and eco-friendly natural fiber in the world which entirely disintegrated by the appropriate action of microorganisms. This review article evaluates the application and various fabrication processes of CNF reinforced nano-biocomposite films in the food packaging industry. The distinct characteristics like mechanical, thermal, and biodegradable properties of CNF based biopolymers with some current research works were studied in this article. Moreover, an important area of concern in packaging films is food-shelf life and antibacterial growth is reviewed. The development of nano-biocomposites utilizing Cellulose Nano Fiber offers eco-friendly material in packaging industries with enhanced mechanical, physical, thermal, barrier and antibacterial properties.

Keywords: Cellulose Nano Fibers, Nano-Biocomposites Films, Food Packaging Application, Mechanical Property Characterization, Antimicrobial Nano-Biocomposite Films, CNF Biodegradation

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I. INTRODUCTION

In recent years, due to increased awareness and push towards more environmentally sustainable technology, composite materials obtained from natural and renewable resources have received significant interest from researchers. Now-a-days water and air pollution is major threat to humanity and development of such kind of materials can overcome this problem up to certain limit. One of the most promising issues in developing countries of Asia is seemed to be plastic pollution, where waste collection systems are often ineffective or nonexistent. Around 400 million tons of human-made plastics derived from fossil fuels are produced each year and 10-13% of plastics are recycled. 9.4 Million tons per annum plastic waste generated and out of these 3.8 Million tons per annum synthetic garbage is not collected. In India, around 43% of manufactured plastics are used for packaging purpose and most are of single use. Around 70% of Plastics packaging products are converted into plastic waste in a short span. Many of these items, like poly bags and food wrappers, have a lifespan of mere minutes to hours, but can last for hundreds of years in the environment.

These kinds of materials are typically extracted from petrochemicals and create waste management issues [1]. In order to satisfy the growing demand for sustainable development and environmental protection, a variety of research activities are aimed at creating food packaging films which could easily disintegrate and effectively mineralized in the environment [2, 3]. The framework of composites is matrix and biopolymers have received tremendous interest as unconventional replacement for petroleum-based polymers due to its biodegradability [4]. This is because of its distinctive properties, such as ecological qualities, alleviating processing and biocompatibility, etc. However, after their desired usage, biopolymers breakdown into nontoxic constituent rudiments by injection molding, film processing, etc. The ecological profit of biopolymer resources incorporate low material energy, CO_2 sequestration, reduced degradation of fossil-based resources with positive impact on agriculture.

Evidently, in contrast with traditional non-biodegradable plastics, the application of biopolymers as food packaging films has impediments like inferior thermal, mechanical and physical characteristics. This gave the emergence of nano-biocomposite theory [5, 6]. Based on different work conducted, nano-biocomposite was recognized as a potential approach to augment barrier and mechanical properties of biopolymers. Nano-biocomposite is a multiphase polymer consisting of two or more components which are consistent phase or matrix mainly biopolymer and inconsistent nano-biofiber phase (<100 nm). The nano-fibers as reinforcement perform a functional role in enhancing the mechanical and surface properties of the continuous biopolymers. The biopolymer tension is conveyed to the nano-fibers from the boundary linking them. The amalgamation of nano-fibers to matrix not only enhances the biopolymers surface and mechanical properties but also provide other features and purposes in food packaging films like antibacterial and antioxidant agent [6, 7, 8, 9, 10, 11 and 12].

The advances in development of nano-biocomposite provide a solid base to generate new applications and opportunities as an alternative to Single-Use packaging films. They provide a response to sustainable development of economically and environmentally appealing engineering. The development of nano-biocomposite films from cellulose nano fibers is important not only to reduce ecological issues but also to improve the functions of the food packaging films. This review paper highlights cellulose nano fiber reinforced nano-biococomposite materials and also discussed current consequences on its properties and characteristics.

1.1 Cellulose – An Ecological Biocomposite Reinforcement

Cellulose is regarded as one of the most profuse biodegradable substance in nature [13, 14 and 15]. It is the foremost element of organic compound on earth, particularly within bio-fibers [16, 17 and 18]. The cellulose is mainly structured in the secondary cell wall of the plants collectively with hemicelluloses and lignin. In 1838, the Frenchman Anselme Payen first discovered cellulose which was derived from plant material and established its chemical composition [17 and 14]. Cellulose comprise a linear chain of numerous hundreds to thousands of β (1 4) connected D-glucose blocks (Fig1) with a linear syndiotactic polymer and hydroxyl groups in an equatorial arrangement, having chemical formula $(C_6H_{10}O_5)n$. Cellulose is s tasteless, odorless white substance. Cellulose is the finest substance on planet which is recyclable, carbon-neutral and organic. Apart from this Cellulose Nano Fibers (CNFs) have several distinctive characteristics like extraordinary mechanical properties, high aspect ratio, large specific surface area, low coefficient of thermal expansion with low price and can be disposed off through composting [19]. A plant fiber contains approximately 65–70% of the cellulosic compounds, with C, H, and O molecules [20]. Cellulose is insoluble in water due to strong and intermolecular hydrogen connections inside a single chain by linking individual chain [16]. Much of the agricultural by-product is not used economically. Cellulose Nano Fibers can be derived from agricultural waste obtained from bamboo, jute, wood, husk (wheat, rice, and barley), sisal, cotton, kenaf, flax etc [21, 22 and 23]. Moreover, cellulose fiber reinforced bio-plastics reflects farming community a possible non-traditional source of value added income.

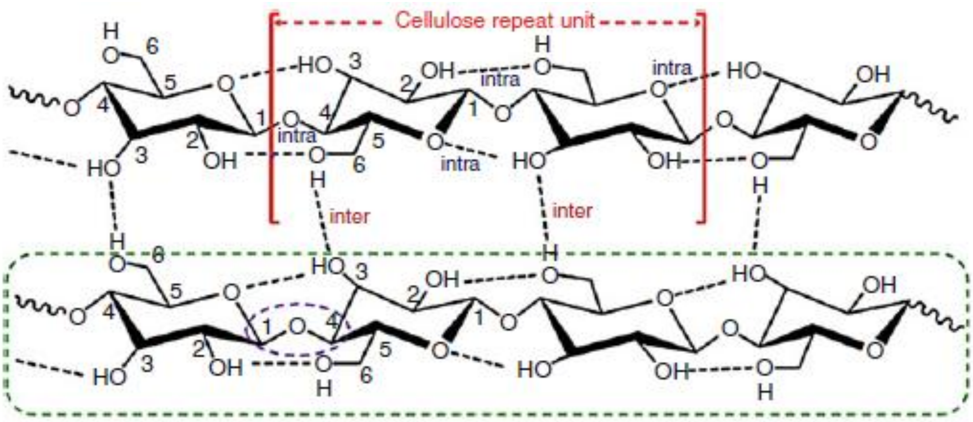

Fig1. Systematic diagram of linear molecular chain, repeating unit and intermolecular hydrogen bonding interactions of cellulose chain - Redrawn from Ref. [14]

Another study described that the compressed arrangement of hydrogen bonding are firmly packed networks in cellulose fibers which provides the stiffness and strength, antimicrobial properties and impermeability to water in the plant cell wall [25]. The constituent of various cellulosic biomasses are briefly described in Table 1 and Fig 2.

Table 1 Chemical composition of cellulosic materials from various sources - Adapted from Ref. [25, 26]

Sources	**Composition**			
	Cellulose	Hemicellulose	Lignin	Extracts, Pectin and Waxes
Hardwood	43–47	25–35	16–24	2–8
Pinecone Biomass	42–46	27	20–23	4–11
Softwood	40–44	25–29	25–31	1–5
Coconut Fiber	31–32	25–26	33–37	5–11
Sugarcane Bagasse	45	30	20–22	3–5
Cotton Stalk	48–52	25–27	24–26	2– 4
Corncob	28–34	39–47	21–29	5–12
Pineapple Leaf	34–40	21–25	25–29	8–10
Jute	60	23	16	1
Wheat Straw	37–43	31–37	18–22	2–14

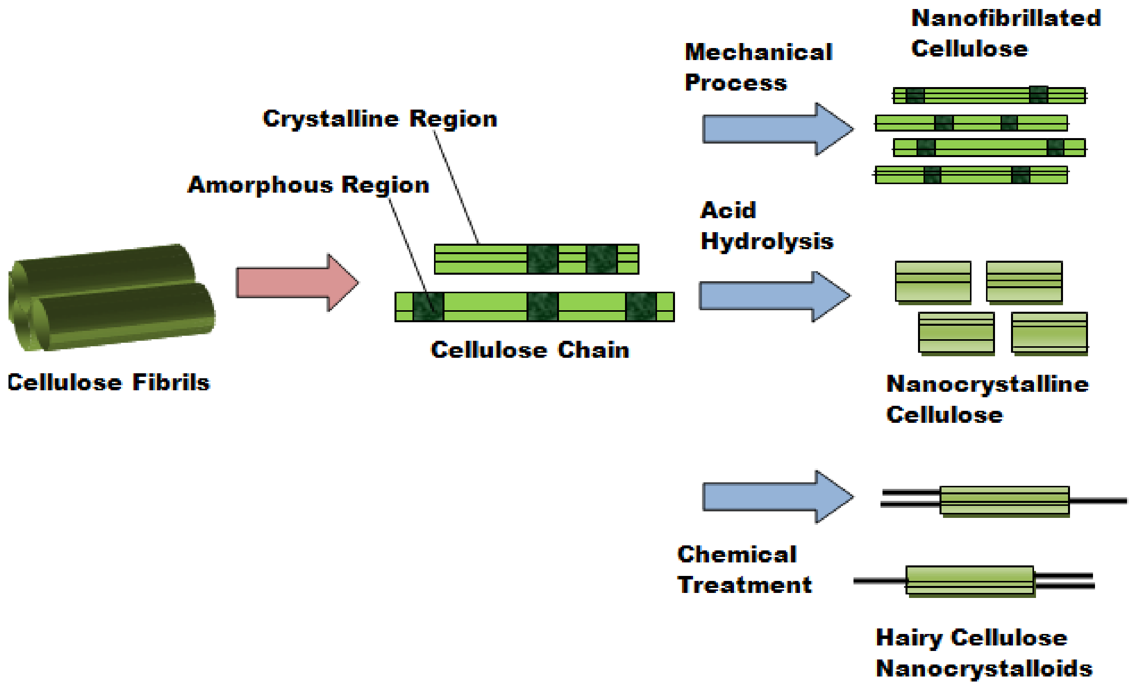

Fig 2. Schematic representation of extraction of nanocellulose (Redrawn from Ref. [25]).

II. NANO-BIOCOMPOSITE FILM

Now-a-days, with growing advancement in polymer technology and widespread exploitation of synthetic substances has led to augmented attention in the fabrication and categorization of biopolymer and its nano biocomposite films [27, 28]. Owing to their outstanding barrier abilities, effortless processing and inexpensiveness oil-based plastics predominate in food packaging applications [29]. Cellulose Nano fiber (CNF) holds certain beneficial compounds, such as antibacterial and antioxidant which prolongs the food shelf life and enhances the food nutrient. The utilization of cellulose nanofibers has huge capability in the development of economical, lightweight, flexible and strong nano-biocomposites for food packaging purpose [30, 31 and 32]. The recyclability of biopolymers empowers the manufacturing of green nano-biocomposites when cellulosic nano fibers are used as reinforcement. Fig 3 depicts an example of flow diagram representing fabrication technique of Polyvinyl Alcohol and Cellulose Nano Fiber biocomposite film by solution casting method.

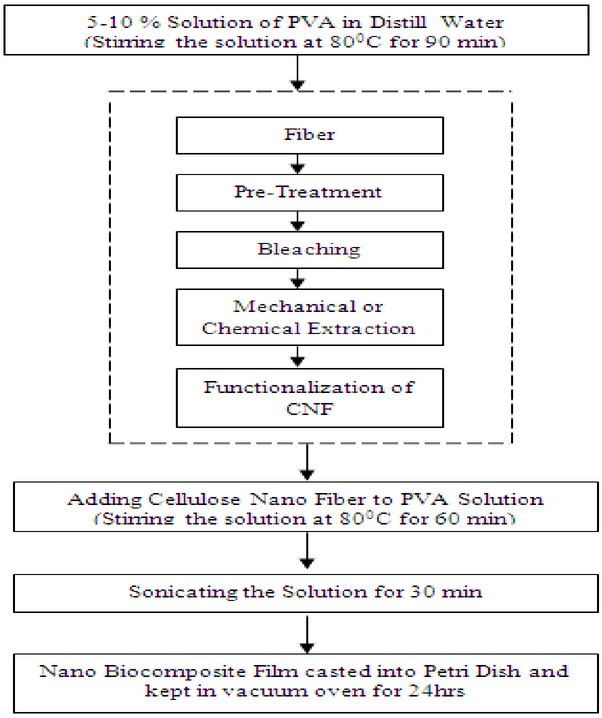

Fig.3 Preparation of Cellulose Nano Fiber reinforced Polyvinyl Alcohol Nano Biocomposite Film through Solution Casting Method

2.1 Mechanical Properties of CNF Film

Cellulose Nano Fiber reinforced biocomposites typically possess excellent mechanical properties relative to other commonly used natural fiber additives. The composite's mechanical performance is strongly associated to the dispersal of the fiber in the polymer and the interfacial relation among them [33 and 34]. Without weakening molecular framework, Cellulose Nano Fibers can be functionalized that inhibits consistent distribution and tough polymer interaction in biocomposite materials [33]. The key characteristic of CNFs as reinforcing agent typically involve tremendously high specific rigidity and elevated aspect ratio, that uphold the proficient load transmission from continuous polymer phase to discontinuous fiber phase [35]. In conjunction with high mechanical properties, CNFs were therefore demonstrated as biodegradable reinforcement resources which convert poor biopolymers matrix to well-built nano biocomposites [36]. Spagnol *et.al.* fabricated nano-biocomposite film from PVA and Cellulose nano whiskers (CWs) obtained from cotton fibers. A considerable raise in mechanical characteristics viz. tensile strength, elongation at break and elastic modulus revealed a strong connection with weight percentage and functionalized CWs in PVA films [37]. Various past studies revealed that the augmentation in tensile strength with incorporation of nano cellulose fillers are a consequence of effectual stress transfer at the fiber-matrix interface related with a strong interaction between them [38, 39 and 40]. Because of the fiber size and fiber-polymer behavior at the interface an enhanced mechanical property were obtained in comparison to the neat biopolymer films when chemically improved cellulose fibers were incorporated [41, 42].

2.2 Thermal Properties of CNF Film

Thermal degradation of natural resources like bamboo, wheat straw, rice straw, sugarcane bagasse and cellulose fibers obtained from different agro-waste exhibit several stages [43]. The first stage linked with low degradation rate associated to lignin, hemicelluloses and pectin at temperatures around 210^{0}C, and a prevalent decomposition stage with weight loss at about 350^{0}C. Thermal degradation of cellulose fibers obtained from wood, bamboo, flax and wheat straw were found to be 332, 331, 347, and 332^{0}C, respectively. The application area of cellulose nano fibers were extended due to their thermal characteristics [44, 45, 46, 47]. A study revealed that because of comparatively high crystallinity (69%), CNF exhibited improved thermal constancy at about 335^{0}C [48]. Yuwawech *et. al.* prepared treated PVA/CNF and PVA/BC (bacterial cellulose) films for food packaging application. The first and second stages occurred at

90-130^{0}C and 240-330^{0}C respectively, in which water is evaporated and 65% weight loss of samples were reported. After reaching third stage at 420^{0}C, due to chemical degradation solid residue or char is formed. The degradation temperature (Td) is marginally higher in PVA/BC and PVA/CNF nano-biocomposites than plain PVA film because CNF act as a nucleating agent, thereby growing the rate of crystallization. Researchers like Lu *et al.*, Frone *et al.* performed the similar study with micro fibrillated cellulose/PVA and found a trivial enhancement in PVA crystallinity when 5 % reinforcement was supplemented [49, 50].

2.3 Antimicrobial Properties of CNF Film

Increasing popularity of nano cellulose biocomposite films among researchers and industries is not only because of environmentally sustainable qualities but also because of its ability as carriers of antibacterial and antioxidant agents which enhance the food shelf-life, quality and security in packaging [51]. Nano-biocomposites provide tremendous possibility to develop inexpensive and compact film for food packaging applications [52 and 53]. Costa *et al.* fabricated Cassava starch/CNF (Licuri Leaves) nano-biocomposite film blended with glycerol and red propolis. The investigators analyzed film's antioxidant and antibacterial effect on various edible products. They found that with incorporation of CNFs water and gas absorption was reduced, bacterial activity is minimized in cheese, and oxidation is delayed in butter without undermining the system's biodegradability [54]. Another work (CNF/Ag Film) showed reduction in bacterial growth and enhancement of food shelf-life by activity of silver particles and cellulose respectively [55]. Owing to different cell walls the effect of silver nano particle on antibacterial activity of *E.coli* bacteria was more than *S.aureus*, irrespective of incubation temperature and time [56]. A different study on Starch blended chitosan and CNF from oil palm biocomposite film demonstrated that when 2% CNF was added, a significant antibacterial growth against Bacillus subtilis (Gram-positive bacteria) was observed. They also found that cellulose nano fiber empowered the chitosin dispersion in biopolymer which also enhanced the antibacterial activity [57]. Niu *et.al.* fabricated cellulose nano fiber and chitosin coated polylactic acid two-layer nano-biocomposite film treated with rosin for food packaging application. They found an admirable antibacterial activity against *E. coli and B. subtilis*, due to collegial effect of R-CNF (Rosin cellulose nano fiber) and CHT (chitosin). Fig 4 show a sustainable reduction of the bacteria in RCNF/PLA/CHT films (5.81 ± 0.27 log CFU of *E.coli* and 5.80 ± 0.24 log CFU of *B. subitilis*) and no antibacterial activity in neat PLA after 1hr of inoculation. This is due to efficient interaction between OeH groups of PLA and NeH groups of Chitosin and homogeneous dispersion of functionalization (Rosin) CNF in PLA. Moreover, the bacterial membrane is damaged due to electrostatic forces between anionic groups of Gram-negative bacteria and cationic groups on the surface of chitosan.

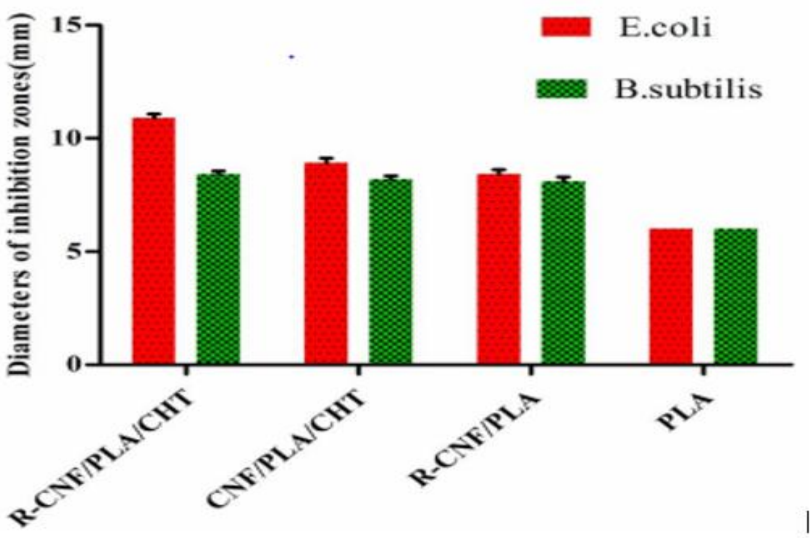

Fig 4. Antibacterial activity of neat PLA, R-CNF/PLA/CHT, CNF/PLA/CHT, and R-CNF/PLA, PLA/CHT films on E.coli and B. subitilis with average diameters of inhibition zones (Redrawn from Ref [58]).

Fig 4 indicate that among all the nano-biocomposite films, RCNF/PLA/CHT sample presented the largest Zone of Inhibition of 10.88/6 and 8.47/ 6 mm against *E. coli and B. subitilis* respectively. The ZOI of CNF/PLA/CHT films (8.91/6 mm againstE. coli, 8.20/6 mm against B. subitilis) were more than those of the R-CNF/PLA films (8.43/6 mm against *E. coli*, 8.09/6 mm against *B. subitilis*). This is because of the incorporation of R-CNF in the PLA than the sole action of chitosin [58].

2.4 Biodegradation of CNF Film

The key benefits of utilizing environmental friendly biopolymers in food packaging technology is because of the fact that they are capable of completing microbial degradability in various environments, reducing incineration of garbage due to plastic [59]. Several hydrophilic additives speed up the cycle of degradation when used with cellulose reinforced nano biocomposites [60]. Luzi *et al*. documented the disintegration of PLA/CNF nano-biocomposite into compost and an enhanced composting condition was recorded when utilizing a commercial surfactant CNF was reinforced [61]. Syafri *et.al.* prepared nano-biocomposite film from starch and cellulose, extracted by water hyacinth (Eichhornia crassipes). The solution casting method was used and solution was vibrated for 0, 15, 30, and 60 min. The soil burial study showed that sonicated nano-biocomposite has a slower pace of degradation, as compared to non-vibrated films. This finding was confirmed by morphological analysis after decomposition, in which a coarse surface and low porosity formation was displayed by 60-minute sonicated specimen. [62]. Fig 5 represents the sonication effect on the decomposition rates of nano-biocomposite films.

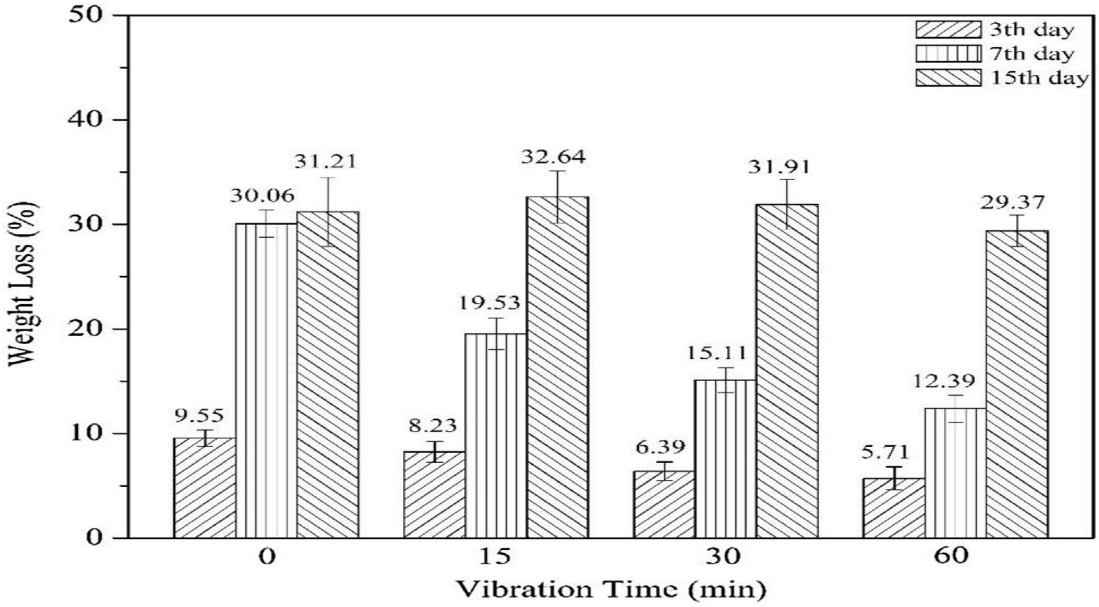

Fig 5. Effect of sonication time on Biodegradation rates of tested samples (Redrawn from Ref. [63]).

On 3rd , 7th and 15th day of soil burial test, percentage weight loss of untreated samples were recorded as 9.55%, 30.06% and 31.21%, while for 60 minute sonicated samples the values were 5.71%, 12.39%, and 29.37% respectively showing slower decomposition rate than the untreated specimens. This pattern was ascribable due to the reason of fiber clumping in the biopolymer and kinetic energy generation from the ultrasonicator [64, 65]. This observation was confirmed by the evidence shown in Fig 6, which depicts a deep void with an unusual shape in morphological framework.

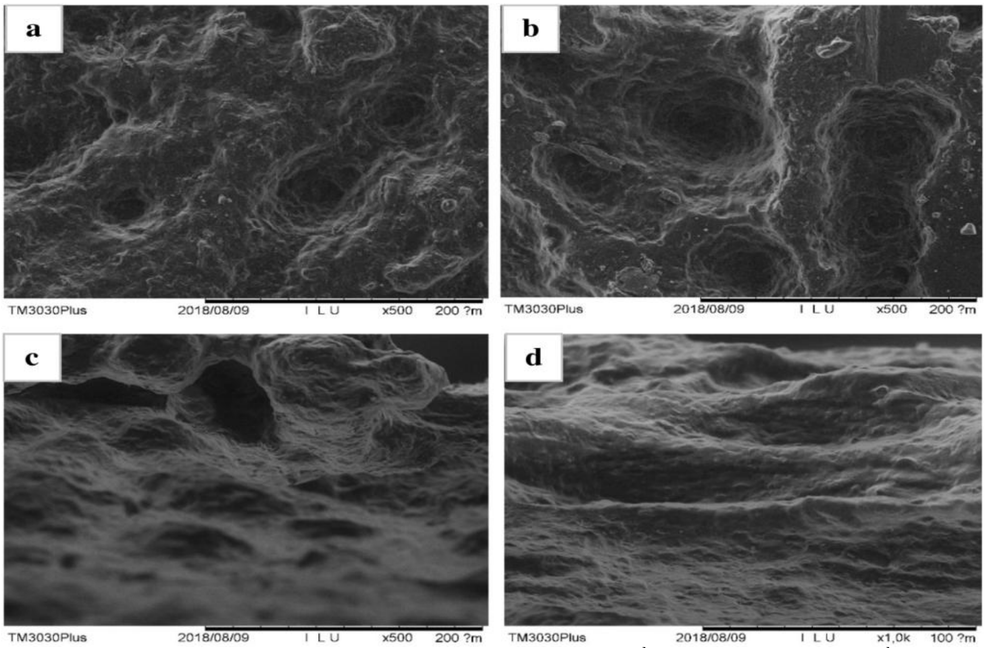

Fig 6. Morphological diagrams of specimen after soil burial test. (a) 3rd day Untreated, (b) 15th day untreated, (c) 60 min sonicated-3rd day, and (d) 60 min sonicated-15th day [65].

III. CONCLUSION

Subsequently, many significant researches has been made to develop promising novel nano-biocomposites in food packaging application with the goal of achieving enhance mechanical, thermal and physical properties [66, 67]. Moreover, with the utilization of abundant and renewable resources like nano-cellulose, improved food shelf-life with superior biodegradation properties were obtained which helps in elimination of waste production from food packaging systems. Nano Cellulose Fibers are carbon free, renewable, recyclable, biodegradable and non-toxic. Because of weak interfacial constancy between fiber and matrix, the major obstacle of improper dispersion was obtained which could be overcome by different techniques like chemical treatment of fiber and use of surfactants. In addition, because of intrinsic compostable behavior of nanocellulose, it is extensively used as reinforcement in biopolymer providing an economic and environmental viability. Further, improved food shelf-life and antibacterial activity was obtained when CNF is used when blended with active fillers. Thus with the help of current fabrication methods compostable, multi-purpose, antibacterial nano-biocomposite films utilizing biopolymer and cellulose nano fibers could be developed, with an emphasis on the green food packaging industry.

CRediT authorship contribution statement

Kritika Singh Somvanshi: Conceptualization, Investigation, Writing - original draft. Prakash Chandra Gope: Supervision, Writing - review & editing.

Conflict of interest

The authors declare that they have no known competing personal relationships that could have appeared to influence the work reported in this manuscript. No conflict of interest exits in the submission of this manuscript.

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