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# Bionanocomposite with Cellulosic nano fibers derived from Delonix Regia fruits and Poly-Vinyl Alcohol: Properties Evaluation

**Kalpit P. Kaurase [a*], Dalbir Singh [a]**

[a] *Hindustan Institute of Technology & Science, Hindustan University, Chennai, India*

***A B S T R A C T :***

Cellulose has been used for past centuries different kinds of applications and it is one the most abundantly available and widely used material on the Earth. Several researchers have explored different lignocellulosic sources for extraction of cellulose and the author has introduced a new source Delonix Regia fruits or seedpods for extraction of cellulose and cellulosic nano fibers (CNF). Cellulose and CNF are extracted by mechano-chemical route and PVA/CNF composite is fabricated by using solvent casting technique. The morphological, mechanical, biodegradability, moisture absorption properties of pure PVA and PVA/CNF composite with 1, 3 and 5 percentage of CNF has been evaluated by conducting scanning electron microscopy, tensile test, soil burial test and moisture absorption test.

*SEM results showed uniform and homogeneous distribution of nano-fillers at lower percentage of CNF and agglomeration observed at higher concentration. From the results of tensile tests it is clearly evident that the Young's modulus and tensile strength initially increased at lower percentage of CNF rapidly and gradually decreased for higher concentration and percentage elongation initially decreased and then started increasing at higher concentration. Biodegradability test demonstrated the decrease in resistance to degradation with higher percentage of CNF and pure PVA has highest degradation resistance which showed that CNF addition improved the biodegradability of the composite material. The moisture absorption rate decreased with CNF addition and hence PVA/CNF composite material will have longer life and better performance without degradation of material.*

***K E Y W O R D S*** *: Bionanocomposite; CNF; Delonix Regia fruit fibers, Mechanochemical treatment, SEM, Tensile test, Moisture absorption test, Biodegradability test.*

## 1. Introduction

Past few decades has experienced a tremendous growth in the application of biologically degradable materials due to the exhausting petroleum derivatives and limitations of non-decomposability of petroleum and related products [1, 2]. Some of the most significant reasons for the wide popularity and acceptance of biodegradable materials are its easy availability, reproducibility and ability to customize the mechanical and thermal properties as per the need [3, 4, 5]. A variety of naturally available materials like polysaccharides, lipids, proteins, chitosan and cellulose have attracted attention of researchers all over the world and various applications of these materials has been explored [6, 7, 8]. Lignocellulosic biomass is one of the most abundantly available and widely used materials since the ancient times for different applications like clothing, housing, etc [9, 10]. Plants and animal biomasses basically have varying compositions of cellulose, hemicelluloses, lignin, pectin, wax, protein, starch, fat, and ash contents [11, 12]. Cellulose is derived primarily from lignocellulosic biomasses in fibers, particle or crystal forms at micro and nano scale [13, 14]. Cellulose can be obtained by different top down approaches including chemical processes like acid hydrolysis, enzymatic hydrolysis, bleaching, pulping, tempo oxidation, etc, and mechanical processes like high energy ball milling, ultrasonication, high pressure homogenization, etc [15, 16, 17].

Cellulose is the primary element which provides the structural strength to the entire plant due to its excellent mechanical properties and hence it can be utilized for different structural applications [18, 19]. Cellulose at the nano scale has more exposed surface area compared to micro and macro scale and hence it can offer excellent wettability with polymers and biopolymers which in turn provides excellent mechanical properties [20, 21, 22]. Due to the advantages of nano scale, most of the researchers have demonstrated extraction of different forms of cellulose at nano scale like cellulose nanocrystals (CNC)/cellulose nanowhiskers (CNW), cellulose nanofibers (CNF) and bacterial cellulose (BC) [23, 24]. All the forms of nano cellulose have different morphology, physical and chemical properties and can be utilized with different resins to obtain bionanocomposites with improved mechanical, thermal and barrier properties. However, the poor wettability between nanocellulose and resins can lead to bionanocomposites with poor mechanical properties and hence treatment of fibers becomes necessary to obtain final product with high mechanical, thermal and barrier properties [25].

In the current research work, Delonix Regia (Gulmohar) seedpods/ fruits were selected as a novel source for obtaining the cellulosic nanofibers (CNF) and further it was utilized for the production of bionanocomposites with biopolymer Poly-Vinyl Alcohol (PVA) by using solvent casting method of fabrication. The most significant reason for selection of Delonix Regia

(Gulmohar) seedpods/ fruits is that Delonix Regia is grown naturally all over India widely and its seedpods have wide spread applications and hence billions of tons of biomass will be available for production with almost negligible cost [10, 26]. Main aim of this research is to assess the morphological, mechanical, thermal and water barrier properties of the obtained bionanocomposite material to determine the area of application and evaluate biodegradability [27].

## 2. Experimental Procedures

### 2.1 Materials

Raw Delonix Regia fruits were collected from the local fields and all the chemicals used for the treatments were of analytical grades and obtained from Navakar Bio Chemicals, Chennai [5, 10].

### 2.2 Cellulosic Nano-Fiber (CNF) extraction

Delonix Regia fruits were size reduced and given different chemical treatments for isolating cellulose [5, 10]. Further, the extracted cellulose fibers were dried in hot air oven at 1050C for 4 hrs and subjected to mechanical treatment, by using a planetary ball milling process followed by sieving through 200 mesh to obtain nano sized cellulose [26, 28, 29]. To prevent the agglomeration, nano fibers were further treated with 5% ethanol and homogenized with Ultra-Turrax T-25 high speed homogenizer (IKA, Germany) at 15000 rpm for 2 min, again dried at 800C for 4 hrs and stored in desiccator [26, 30].

### 2.3 Preparation of Bionanocomposite

Bionanocomposite was prepared by using ingenuous solvent casting method with extracted cellulosic nano fibers (CNF) and Poly Vinyl Alcohol (PVA) [31, 32, 33]. The PVA solution of 10 wt% was prepared by dissolving PVA granules in distilled water by initially keeping temperature at 350C and then gradually increasing the temperature to 800C with continuous magnetic stirring at 700 rpm for 1 hour with Remi 1 MLH magnetic stirrer with hot plate to make homogeneous polymeric solution [34, 35]. After 1 hour of mixing 5 wt% of Glycerol (GL) was added as a plasticizer to the polymeric solution and continuously stirred for 1/2 hour till the viscous polymeric solution was formed [36, 37]. Once the homogeneous mixture of PVA was prepared 1, 3 and 5 wt% of extracted CNF was added to the polymeric solution with continuous magnetic stirring at 700 rpm for 3 hours to form PVA/CNF composite followed ultrasonication for 10- 20 minutes to avoid agglomeration [34, 37]. These homogeneous composite solutions were poured on petri dishes for slow evaporation of the solvent for 2 hours and finally the solution was kept in a hot air oven for 3 hours for drying [38, 39]. Obtained pure PVA and PVA/CNF composite films were named as PVA, PVA/CNF1, PVA/CNF3 and PVA/CNF5 and kept in a desiccator after drying for further analysis.


**Fig. 1:** Procedure for preparation of PVA/CNF Composite

2.5 Scanning Electron Microscopy (SEM)

Scanning electron microscopy (SEM) of the PVA/CNF1, PVA/CNF3 and PVA/CNF5 were obtained using JSM-6360, JEOL Ltd., Japan at 18 kV [39, 40]. The sample used for the imaging was coated with gold before being measured.

2.6 Tensile Test

Mechanical properties of bionanocomposites were determined by performing the tensile tests at room temperature according to ASTM D638-03 standards using Universal Testing Machine (TKG-EC 500 N) with the speed of 10 mm/min and grip separation of 25 mm [36, 41, 42]. Test specimens were cut to 25 mm × 3 mm size and kept in a desiccator for 2 days for conditioning before testing [41, 42, 43]. All the tests were repeated five times to ensure the repeatability of test methods and the results were obtained by averaging.

2.7 Biodegradability Test

The biodegradability of PVA, PVA/CNF1, PVA/CNF3 and PVA/CNF5 samples was obtained by a soil burial test. Initially all the samples were cut into the size of 3 × 3 cm2 and initial weight (Wi) was recorded with digital weighing balance. These samples were buried in the closed plastic container to a depth of 10 cm and kept at room temperature. Humidity level inside the container was maintained at 40–50% relative humidity (RH) by sprinkling water with uniform flow rate during the entire test. During the total test duration was 20 weeks, the samples were examined in the gap of 2 weeks. After completion of test duration, samples were cleaned with distilled water and dried in a hot air oven at 700C for 24 hours and final weight (Wf) is recorded with digital weighing balance [38]. Three samples of each material were tested to confirm the repeatability of test procedure. Biodegradation rate was calculated as per the equation given below:

$$BDR(\%) = \frac{W_i - W_f}{W_i} \times 100\%$$

where BDR = Biodegradation rate, $W_i$ = Initial weight of the sample and $W_f$ = Final weight of the sample.

2.8 Moisture Absorption Test

The moisture absorption rate of PVA, PVA/CNF1, PVA/CNF3 and PVA/CNF5 samples was obtained by a moisture absorption test. Initially all the samples were cut into the size of 1 × 1 cm2, dried in a hot air oven at 600C for 48 hours and initial weight (Wi) was recorded with digital weighing balance. These samples were kept in a closed chamber at room temperature and 75% relative humidity (RH) during the entire test. During the total test duration was 6 hours, the samples were examined in the gap of every 30 minutes. The samples were weighed (Wf) after every 30 minutes with digital weighing balance [44, 45]. Three samples of each material were tested to confirm the repeatability of test procedure. Moisture absorption rate was calculated as per the equation given below:

$$MAR(\%) = \frac{W_i - W_f}{W_i} \times 100\%$$

where MAR = Moisture absorption rate, $W_i$ = Initial weight of the sample and $W_f$ = Final weight of the sample.

## 3. Result and Discussion

3.1 Chemical Composition

Chemical composition of the Delonix Regia fruit fibers is obtained as per the previous work and shown in Table 1 [5, 10, 26]. Procedures were repeated three times to ascertain the reproducibility of the method employed.

**Table 1:** Chemical composition of Delonix Regia fruit fiber (%w/w) [5, 10, 26]

| Cellulose | Hemi-cellulose | Lignin | Extractives | Moisture | Ash |
|---|---|---|---|---|---|
| 66.9% | 11.6% | 6.5% | 4.5% | 8.3% | 2.2% |

3.2 Scanning Electron Microscopy (SEM)

The SEM micrographs of PVA/CNF1, PVA/CNF3 and PVA/CNF5 are shown in Figure 5. Smooth surfaces and homogeneous distribution of nano-filler was observed in PVA/CNF1 and PVA/CNF3 films without any signs of cracking, whereas for PVA/CNF5 agglomeration of nano-filler was observed at different locations and the distribution does not remain homogeneous. At higher concentration of nano-fillers, higher interaction between nano particles occurs instead of interaction between filler and matrix which can cause agglomeration and non uniform distribution of nano-fillers [39, 40]. Hence, it is recommended to limit the addition of nano-fillers well below 5% to obtain homogenous distribution without agglomerations.

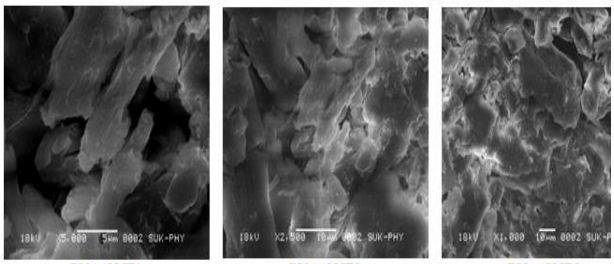


**Fig. 2**: SEM images of PVA/CNF1, PVA/CNF3 and PVA/CNF5

3.3 Tensile Test

Young's modulus, tensile strength and percentage of elongation at break of pure PVA specimen and PVA/CNF1, PVA/CNF3 and PVA/CNF5 composite specimens with addition of varying amount of nano-filler composites were determined by tensile tests and depicted in Table 2 [35, 40, 48]. Young's Modulus of pure PVA specimen was found to be 1108 MPa and with the increase in the percentage of nano-filler from 1% to 5% the value of Young's Modulus was observed to increase. The values Young's Modulus of PVA/CNF1, PVA/CNF3 and PVA/CNF5 specimens are observed to increase by 52.08%, 68.23% and 83.48% respectively compared to modulus of pure PVA. Tensile strength of pure PVA specimen was observed as 53.1 MPa and gradual decrement in the value of tensile strength was observed with increase in nano-filler loading [41, 42, 44]. The value of tensile strength of PVA/CNF1 increased by 62.7% compared to pure PVA whereas tensile strength of PVA/CNF3 and PVA/CNF5 specimens are observed to decrease by 14.81% and 24.42% respectively compared to tensile strength of PVA/CNF1 [41, 46, 47]. Percentage elongation at breaking pure PVA specimen was observed as 223.5% and the value of percentage elongation was observed to decrease with increase in the amount of nano-filler. The value of percentage elongation of PVA/CNF1 specimens by decreased by 12.8% compared to pure PVA, whereas percentage elongation value of PVA/CNF3 and PVA/CNF5 specimens was observed to increase by 11.44% and 25.87% respectively compared to the value of percentage elongation of PVA/CNF1 specimen. It is advisable to select the material composition as per the need of structural applications.

**Table 2:** Mechanical Properties PVA/CNF Composite

| | Young's Modulus (MPa) | Tensile Strength (MPa) | Elongation (%) |
|---|---|---|---|
| PVA | 1108±57 | 53.1±1.2 | 223.5±11 |
| PVA/CNF1 | 1685±65 | 86.4±1.2 | 194.8±11 |
| PVA/CNF3 | 1864±82 | 73.6±0.9 | 217.1±24 |
| PVA/CNF5 | 2033±91 | 65.3±1.4 | 245.2±38 |

3.4 Biodegradability Test

Biodegradability of the material is entirely dependent on the water absorption rate which helps in the subsequent growth of fungicides and bacteria especially in contact with the soil. Hence, to understand the biodegradability of any material, generally soil burial test is carried out. Pure PVA, PVA/CNF1, PVA/CNF3 and PVA/CNF5 samples were subjected to soil burial test as per procedure mentioned earlier [38, 39]. Degradation of the material was clearly visible in almost all the samples and the entire 20 weeks time was divided into two stages. In the first stage of initial 3 weeks, the rate of degradation is generally very rapid compared to the second stage of remaining subsequent period in which the rate degradation is relatively slow. At the end of 20 weeks, minimum value of biodegradation rate of 7.25% was observed for pure PVA sample and the value of biodegradation rate was observed to increase with the increase in CNF percentage. Hence, the resistance to degradation decreases with addition of higher amount of CNF and pure PVA has highest degradation resistance.

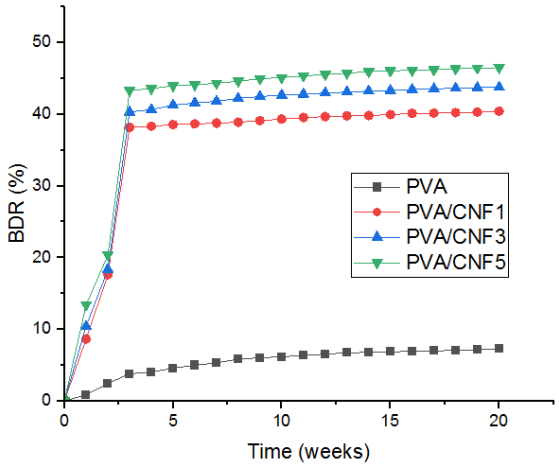


**Fig. 3:** Biodegradability rate of Pure PVA and PVA/CNF Composites

3.5 Moisture Absorption Test

Moisture absorption is a very common phenomenon in polymeric resin based composite materials which degrades the mechanical, thermal and chemical properties of the material and significantly reduces the performance and life of the material. Hence, it is very much essential to determine the moisture absorption rate of material to assure the estimated performance and life of the material [44, 45]. Moisture absorption test was conducted for pure PVA, PVA/CNF1, PVA/CNF3 and PVA/CNF5 samples. Highest moisture absorption rate was observed for pure PVA samples, whereas the moisture absorption rate was observed to decrease with increase in CNF percentage. During initial 90 minutes moisture absorption rate was increasing very rapidly in all the samples and after 90 minutes it was observed that increase in the moisture absorption rate slowed down and it almost became uniform in all the samples.

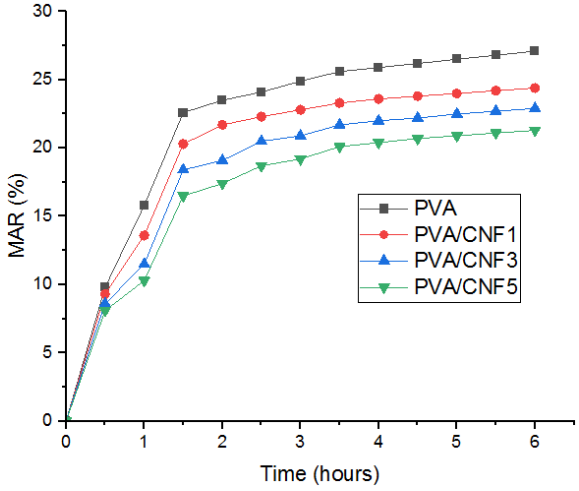


**Fig. 4:** Moisture absorption rate of pure PVA and PVA/CNF composites

## 4. Conclusions

Results of Scanning Electron Microscopy indicated uniform and homogeneous distribution of nano-fillers for lower percentage of CNF whereas signs of agglomeration are visible in higher concentration of CNF in PVA/CNF composite. Hence, it can be concluded that for uniformity of the concentration and hence better mechanical properties lower percentage of CNF is very much suitable. Results of the tensile tests indicated that the Young's modulus initially increased at lower percentage of CNF very rapidly and gradually decreased for higher concentration, similar trend was observed for tensile strength whereas percentage elongation initially decreased and then started increasing at higher concentration. Hence, average percentage of CNF concentration is recommended for mechanical applications and CNF concentration can be chosen depending on the area of application. Results of biodegradability test showed that the resistance to degradation decreases with addition of higher amount of CNF and pure PVA has highest degradation resistance. CNF addition improved the biodegradability of the composite material. Also, the moisture absorption rate decreased significantly with addition of CNF and hence PVA/CNF composite material will have longer life and better performance without degradation of material.

**Nomenclature**

PVA- Poly- Vinyl Alcohol

CNF- Cellulosic Nano Fibers

SEM- Scanning Electron Microscopy

MAR - Moisture Absorption Rate

BDR- Biodegradation Rate

ASTM- American Society for Testing and Materials

RH - Relative Humidity

GL – Glycerol

Wf - final weight of the sample

$W_i$ = Initial weight of the sample

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**Conflicts of Interest**

The author declares that there is no conflict of interest regarding the publication of this manuscript. In addition, the authors have entirely observed the ethical issues, including plagiarism, informed consent, misconduct, data fabrication and/or falsification, double publication and/or submission, and redundancy.

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