Development of Neem Based Bioplastic for Food Packaging Application

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Abstract: Neem (Azadirachta indica) belongs to mahogany family Meliaceae. It is a traditional medicine used in India. Each part of the tree is used in development of many products such as antifungal, anthelmintic, anti diabetic, antiviral, antibacterial, sedative, contraceptive and others. Present work unveils biopolymer properties of Neem leaf extract (NLE) and their tensile properties for the food packaging applications. In this study, Neem leaves were collected and poured in boiling water (1:10) and crude extract of Neem obtained was then cooled and reconciled for 24hrs, constituents such as glycerol, glucose, agar and Gelatin were added and further, heated for about 20 minutes at 60°C of temperature. A biopolymer based plastic formation was observed after 48 hrs of incubation at room temperature. The biopolymer was found to be green in colour and there was no fungal or bacterial growth was observed on the biopolymeric film, indicated its medicinal and aseptic properties. Since, micro tensile test was carried out for the developed polymeric film. However, its strength was relatively weaker than bioplastic material, but requires significantly less energy for its development. Hence, further rigorous research work has to be carried out to improve its strength properties to use them as a suitable packaging material and could be potential biomaterials to replace the conventional plastic.

1. Introduction

Azadirachta indica (Neem) belongs to the Meliaceae family which is found throughout the world but indigenous to Southeast Asia [1] most popular in India and Burma [2]. In Arabic language Al Shurisha and in English Neem is the name of Azadirachta indica and popularly known as Neem [3]. Since last 4000 years it is a valuable versatile tree with religious, medicinal and social uses [1]. Neem is classified under tree with appropriate height of 25meters with a semi-straight trunk. After 3-5 years of growth fruiting transpires and the prolificacy of the tree is distinguished after 10 year. Approximately 30 centimetres is length of the leaf and each leaf has 10-12 saw-toothed leaflets of 2.5cm wide and 7 cm long [3].
From earlier reports Neem trees found in Asian countries like Burma, India, Thailand, Cambodia, Bangladesh, Indonesia, Iran, Malaysia, Nepal, Sri Lanka and Vietnam. In India wide range of Neem grows and Uttar Pradesh shares the majority. About 18 million of Neem trees found in India [4].

Crude extracts of Neem with different fractions from leaf, bark, root, seed and oil describe biological activity with various diseases [5]. Traditionally, allergic skin reactions, smallpox and chicken pox are treated with the leaves and their paste. Numerous medicinal uses of Neem and their products comprise treatment for cancer, skin diseases, digestive disorders and AIDS. Several active chemical compounds are present in the selected plant, including glycosides, dihydrochalcone, coumarin, tannins, zadirachtin, nimbin, nimbidine, diterpenoids, triterpenoids, proteins [5, 3].

![Figure 1. Map showing worldwide Distribution of Azadirachta indica in different countries [6]](image)

The term Bioplastics was coined by European Bioplastics e.V and stated that bioplastics are either biodegradable, biobased or features both properties [7, 8]. Plastics which are degraded by accessible micro-organisms in nature (bacteria, fungi and algae) are regarded as biodegradable plastics. Degradation may be aerobic or anaerobic in nature results in conversion of bioplastics into water, carbon dioxide and biomass. Biobased bioplastics are the plastics fabricated with biomass as raw materials in place of oil [7].

Owing to the many advantages of natural polymer it is feasible to be used in regular basis, as integration of natural polymer in the environment reduces the rate of pollution. As suggested above Neem has many constructive properties in medicine as well as other areas. Thus, the main aim of our work is to prepare Neem-starch blend for synthesis of Neem bioplastics and evaluate the tensile strength along with its degradation.

2. Literature Review
Natural polymers are the product of plants and trees. Products such as proteins, starch, plant oils and cellulose are extracted which are essential for developing renewable and biodegradable polymer materials [9]. Many researchers have implicated the idea of utilizing natural polymer for development of bioplastic such as starch from oxidized corn starch, [10] potato, wheat, rice etc. There was report on development of banana peel based bioplastic by Jayachandra et al. [11].
3. Methodology

3.1 Sample collection:
Dark green, matured leaves of Azadirachta indica were plucked from the campus of KLE Technological University, Hubballi. The plant species was authenticated by a botanist and verified by morphological features. Care was taken to take the leaves of similar size and shape for the extract preparation [12].

3.2 Leaf extract:
5g of Neem (Azadirachta indica) leaves were weighed and washed with water. It was then crushed using a mortar and pestle to make it into paste. 100ml of distilled water was added to the paste and boiled. It was then filtered using a kitchen strainer to remove the debris. The extract was stored at room temperature for 24 hours [13].

3.3 Blend Preparation and casting for Neem bioplastic:
To the extract 0.75g of Glucose, 1.125g of Gelatin, 0.565g of Agar and 1.8ml of Glycerol were added and kept on the magnetic stirrer with 60°C temperature for 2 hours. All the chemicals were procured from SD fine of AR grade. The blend was then cast on a clean dry acetate sheet and left for drying for 48 hours. It was then peeled.

3.4 Swelling and solubility studies:
To evaluate the sustainability and retention of the bioplastic, the swelling test was conducted. Pre-weighed samples were immersed in solvents such as water, chloroform and methanol for 2 hours. The weight difference was calculated. Then the bioplastic developed were subjected to different solvents to examine their solubility. Samples of the size 2 cm by 1 cm were taken. Solvents such as ammonia, acetic acid, chloroform, acetone, methanol, sulphuric acid, ortho-phosphoric acid, and ethanol were utilized. Solvents were diluted to 20% and 40% for the comparative study. The bioplastic were immersed in these solvents for about 2 hours and results were tabulated.

3.5 Degradation by Soil Burial method:
The peeled biopolymer was cut into small circles of 12mm diameter and buried in soil. The condition for the degradation is as shown in Table 1. The internal controls used were plastic along with a paper piece of the same dimensions as our sample taken. The bioplastic degradation was observed.

| Sample name | Condition          | Water component |
|-------------|--------------------|-----------------|
| S1          | Plant+ Fertile soil| Yes             |
| S2          | Plant+ Infertile soil| Yes           |
| S3          | Fertile soil       | Yes             |
| S4          | Infertile soil     | Yes             |
| S5          | Infertile soil     | No              |

3.6 Tensile Test:
The tensile properties of bioplastic developed were characterized for their tensile strength, approximately sample size of 50x0.2x10 mm3 (LxWxH) was used for Universal Testing Machine (Instron 3345, UK) equipped with a 500 N load-cell. The ends of the rectangular specimens were mounted vertically on two mechanical gripping units of the tensile tester, leaving a 40-mm gauge
length for mechanical loading at an extension rate of 1 mm/ min. The reported tensile moduli and maximum tensile strengths represented average results of six tests.

4. Results and Discussion

4.1 Blend formation:
Figure 2 depicts the bioplastic produced and the natural colour was brownish in nature and it was in the form of a thin sheet.

![Figure 2 The blend prepared by using Neem leaf extract](image)

4.2 Swelling Test:
After 2 hours of immersion, there was a weight gain in two of the three samples. The sample ST1 showed significant weight gain, ST2 showed slight weight gain while ST3 pointed out loss of weight compared to the initial weight.

| Sample No. | Solvent     | Quantity (ml) | Initial Weight (g) | Final Weight (g) | Difference (g) |
|------------|-------------|---------------|--------------------|-----------------|----------------|
| ST1        | Water       | 5             | 0.0571             | 0.0690          | 0.0119         |
| ST2        | Chloroform  | 5             | 0.0615             | 0.0630          | 0.0015         |
| ST3        | Methanol    | 5             | 0.0517             | 0.0198          | -0.0319        |

4.3 Solubility Test:
To evaluate the effect of different solvents solubility test was conducted on the synthesized bioplastics. Apart from sulphuric acid none of the solvents seem to have affected it. In 20% ammonia solution partial solubility and colour change was observed. No other physical change (shape or size) was observed.
Table 3. Solubility test using different solvents.

| Sample No. | Solvent       | Concentration | Results        |
|------------|---------------|---------------|----------------|
| 1          | Ammonia       | 20%           | Partially soluble |
|            |               | 40%           | Insoluble       |
| 2          | Acetic acid   | 20%           | Insoluble       |
|            |               | 40%           | Insoluble       |
| 3          | Chloroform    | 20%           | Insoluble       |
|            |               | 40%           | Insoluble       |
| 4          | Acetone       | 20%           | Insoluble       |
|            |               | 40%           | Insoluble       |
| 5          | Methanol      | 20%           | Insoluble       |
|            |               | 40%           | Insoluble       |
| 6          | Sulphuric acid| 20%           | Partially soluble |
|            |               | 40%           | Soluble         |
| 7          | Ortho phosphoric acid | 20%   | Insoluble |
|            |               | 40%           | Insoluble       |
| 8          | Ethanol       | 20%           | Insoluble       |
|            |               | 40%           | Insoluble       |

4.4 Degradation by Soil burial method:
Figure 2A and 2B were taken a week apart. Substantial degradation was observed after a week. Samples S1-S4 showed better degradation than sample 5, the reason being that samples S1-S4 were watered frequently (once in two days). The sample being kept in soil without providing water indicated that water is needed for the degradation process as seen from the figure 2B.

4.5 Tensile strength:
Tensile strength of developed Neem based bioplastic material was found to be 4.9 Mpa which is considerably less than the standard requirement of ASTM D882. However, by considering huge potential of Neem leaves especially in medical products comprise treatment for cancer, skin diseases, digestive disorders and AIDS. Rigorous research work has to be carried out to find the best composite for Neem based blend preparation and could be a potential alternative to conventional plastic in packaging industries.
5. Conclusion
From the above work we have seen that Neem based bioplastic can be used for various applications in medicinal as well as in food packaging. Provided the improvement of tensile strength, further applications can be explored. However, the developed bioplastic has adequate soluble, swelling and degradation properties within acceptable norms and further improving tensile properties can make it feasible for its utilization at industrial scale.

References
[1] Kasarkar A R and Barge A N 2016 2016 J. Med. Plants 4 11-3
[2] National Research Council 1992 Neem: a tree for solving global problems ed N D Vietmeyer and F R Ruskin (Washington, DC: The National Academies Press) chapter 4 pp 31-38
[3] Al-Hashemi Z S and Hossain M A 2016 Pac. Sci. Rev. A 18 128-31
[4] Tinghui X, Wegener M K, O'Shea M and Deling M Conf., 45th World distribution and trade in neem products with reference to their potential in China January 23-25 2001, Adelaide Australian Agri. l and Resource Economics Soc
[5] Al Charchafchi F, Al-Nabhani I, Al-Kharousi H, Al-Quraini F, Al-Hanai A and Al-Khuwair O 2007 Pak. J. Biol. Sci. 10 3885-9
[6] http://eol.org/data_objects/21122306 July 2012
[7] Rujnić-Sokele M and Pilipović A 2017 Waste Manage. Res. 35 132-140
[8] Chen Y J 2014 J. Chem. 6.1 226-231
[9] Tănase E E, Rápá M and Popa O 2014 Sci. Bull. Ser. F 18 188-95
[10] Dang X, Shan Z and Chen H 2016 Biotech. Appl. Biochem. 180 917-29
[11] Yaradoddi J, Patil V, Ganachari S, Banapurmath N, Hunashyal A and Shettar A 2016 Int. J. Pharm. Res. Allied Sci. 5 56-65
[12] Nagappan R 2012 Asian Pac. J. Trop. Biomed. 2 707-11
[13] Parashar V, Parashar R, Sharma B and Pandey A C 2009 Dig. J. Nanomater. Biostruct. 1 1