

Original Research Article

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Biodegradable Sheet from Chitosan and Arrowroot Starch by Compression Moulding and its Properties

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ABSTRACT

Keywords

Thermoplastic Starch, Thermoplastic chitosan, biodegradability

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This research work entitled preparation and properties evaluation of biodegradable sheet from arrowroot starch and chitosan. It was carried out through compression moulding method by using Arrowroot starch, Chitosan, Glycerol, Citric acid, water. Functional properties like Tensile strength, Elongation at break (%), biodegradability were evaluated. There were two treatments T1 and T2 with varying composition of arrowroot starch and chitosan. Tensile strength was observed as 4.80MPa (T1) and 3.74 MPa (T2). Elongation at break % was 155.6% (T1) and 203.3% (T2). Soil burial method was used for evaluation of biodegradability of sheet. T2 had better percentage of decomposition in 30 days. The biodegradable sheet can be used for various packaging applications and safety to environment as they are biodegradable.

Introduction

It is widely believed that plastics do not biodegrade due to its synthetic polymers which is not easily and apt degraded; eventually which affects environment and living beings. This creates an urge for a better alternative which is easily degradable by organism, and thus emerged the concept of

biodegradable plastics and polymers which was first introduced in the 1980s. Biodegradable polymer decomposes through the action of living organisms, usually bacteria. There are two types of biodegradable plastics: bio plastics, plastics that are derived from renewable raw materials such as poly-3-hydroxybutyrate (PHB) and polyhydroxyvalerate (PHV) and plastics that contain

biodegradable additives. The latter are derived from petrochemicals and contain additives that enhance biodegradation. Biodegradable polymers are a specific type of polymer that results in by-products such as CO₂, N₂, H₂O, biomass and inorganic salts when they break down.

These plastics are found both naturally and synthetically and consist largely of ester, amide and ether groups. How they breakdown is determined by their structure and is often synthesized by condensation reactions as well as ring opening polymerization. Biodegradable plastics are made from starch, cellulose, chitin, and protein extracted from renewable biomass (Azahari *et al.*, 2011). The development of most bio plastic is assumed to reduce fossil fuel usage, and plastic waste, as well as carbon dioxide emissions. The biodegradability characteristics of these plastics create a positive impact in society, and awareness of biodegradable packaging also attracts researchers and industries (Siakeng *et al.*, 2019). Bio plastics may be openly taken out from natural resources like lignins, proteins, lipids, and polysaccharides (e.g., starch, chitin, and cellulose) (Johansson *et al.*, 2012). Approximately 50% of the bio plastics used commercially are prepared from starch.

Chitosan is obtained by deacetylation of chitin. Chitin is present in the exoskeleton of many marine invertebrates, insects, some algae and mucoraceous fungi. However, despite the wide range of sources for chitin extraction; it is mainly obtained from crustacean shells, a major waste in various food and fish industries (Leceta *et al.*, 2014). Furthermore, with the objective of developing a more sustainable way of waste management, waste valorization techniques have received significant attention in the last years with an emerging interest to use crustacean shell wastes as an attractive source of chitosan (Leceta *et al.*, 2015).

Starch is a biodegradable and thermoplastic polymer formed mainly by two polysaccharides: amylose and amylopectin. In recent research, this polymer is considered to be a promising agent for applications such as food packaging due to its biodegradability, flexibility and its hypoallergenic quality, which makes it an appropriate material for food industry utilization. Starch from arrowroot was used in this research, which is a less studied starch source. Arrowroot starch is cheaper and easily available in anywhere. Arrowroot powder or starch extracted from the root of a tropical plant known as *Maranta arundinacea*.

Justification

Every year, millions of tons of plastic are discarded into landfills, where they will take hundreds of years to break down. In addition to being land filled, some plastic is disposed of improperly, leading to plastic pollution on land and in lakes and oceans where they persist. Once in the environment, animals can accidentally eat the plastic but are incapable of digesting it, so the plastic just sits in their stomachs. Over time, the plastic can block off their digestive system, and the animal will starve. Biodegradable plastics offer a potential solution to this problem. One of the main advantages of using biodegradable polymers to make films or sheets is the significant reduction in the carbon emissions that happen during the manufacturing process as compared to that of regular plastic. Avoiding migration of polymer compound into packaged food is also a big issue. Motivation of this research is to synthesize of biodegradable and biocompatible packaging sheet to replace the petroleum based polymer for packaging of foods. The ultimate aim was to make the biodegradable sheet by compression moulding successfully and to achieve the maximum degradability, sustainability by combining or fusing arrowroot starch with chitosan.

Chitosan is natural polymer found in the exoskeletons of insects, the cell wall of fungi and hard structure of invertebrates, fish, egg shell etc in the form of chitin, which is insoluble in organic solvents, that's why removal of acetylene group is necessary which leads to the conversion in chitosan. Chitosan is biocompatible, biodegradable, and edible and it also has great antimicrobial property too and on other hand starch is also a biodegradable polymer and arrowroot powder is a great source of starch. It can be available anywhere.

Materials and Methods

Method of Sheet Preparation

Preparation of biodegradable packaging sheet by compression moulding was carried out by below mentioned process according to Lopez *et al.*, 2014 with some modifications according to the availability of infrastructure. Complete process is explained with the help of flow diagram.

The biodegradable sheets of two different proportions were obtained by compression moulding method. At the beginning stage, All the chemicals i.e. chitosan, Arrowroot starch, Talc powder, Citric acid and glycerol were weighed as per the required proportions and then mixed with the help of glass rod. After that the mixture was conditioned at 25⁰C of temperature for next 28 h. Then the mixture was heated with the help of hot plate by continuous stirring by glass rod up to 15 minutes and the temperature was maintained in between 135- 140⁰C. At the time of heating and stirring the temperature is also checked simultaneously by digital thermometers so that the temperature not increase or decrease than the given range. After continuous heating and stirring operation the mixture became slightly sticky in nature. Then the mixture was stored again for next one week at 25⁰C temperature. After one week the mixture was taken out for final stage at which it placed in between the

mould in a thin layer and then it placed into the preheated chamber (135⁰-155⁰C) of compression moulding machine. Then the pressure lever was pulled down on the mould and the pressure set at 25 bar and after each 6 minutes of interval the pressure was reduced to 23 bar and lastly 13 bar. Then cold water supply valve of the machine was opened to allow the water flow through the machine so that the system cools down to the room temperature immediately after stopping the heat supply and gradually the pressure was also allows to down to zero and the mould was taken out from the chamber with prepared biodegradable sheet.

Test for Biodegradable Sheet

Thickness of the Sheet

Thickness is the most important parameter which has direct influence on tensile properties and puncture force (Galdeano *et al.*, 2013). The testing was carried out by using thickness gauge. The prepared biodegradable composite sheets by chitosan and arrowroot starch were tested for thickness from three different dimensions to get the mean.

Tensile Strength

Tensile strength refers to the maximum load that a material can support without fracture when being stretched, divided by the original cross-sectional area of the material. Tensile strengths have dimensions of force per unit area and the measurement are expressed in units of pounds per square inch, often abbreviated to psi or MPA. When stresses less than the tensile strength are removed, the material returns to its original shape and size either completely or partially. The biodegradable composite sheet were obtained from chitosan and arrowroot starch and tested for tensile strength using texture profile analyser in which the exact tensile strength was measured according to ASTM D638

which specifically designed for testing.

Plastics upto 14 mm of in thickness, and can be used to test any type of plastic samples within this defined thickness range. ASTM D638 testing is conducted on a Universal Testing Machine at a constant crosshead speed. The measured tensile properties are UTS(MPA), Elongation, Max%, Break (MPA), Elastic modulus (MPA). The above mentioned data were collected during the test of two samples T₁ and T₂.

Test for Biodegradability

Waste Reduction is a big challenge now days. Plastic makes up around 13 percent of the waste stream, representing 32 million tons of waste. Biodegradable plastics also help conserve petroleum supplies. Bioplastics are important in helping consumer goods companies present their brands in a favourable light. Recyclable or compostable packaging made from biological materials can be used to make their products more environmentally friendly in the eyes of consumers. In biodegradation process biodegradable plastics breaks down with the help of microorganisms into carbon dioxide, methane, water and biomass. The biodegradability test was done to know the impact on environment. The main aim was to achieve the degradability of the prepared biodegradable sheet. For this, the samples were buried in the soil. The test was performed with some modifications as per the following paper (Rachamawati *et al.*, 2015), (Azahari *et al.*, 2011) and (Thakore *et al.*, 2001). A large bucket was filled with the soil and the samples were cut into definite sizes of square pieces and buried in the soil at a depth of 10 cm. The bucket was placed in a room under at 25⁰C temperature and 50-60% of humidity for upto one month. The moisture of the soil was maintained by sprinkling water into it at regular intervals of time. The degradation of the samples was determined at

second and fourth week of intervals by carefully removing the samples from the bucket and washing them to remove the soil from it and the samples were dried at 45 – 50⁰C upto 1.5 – 2 hours, so that the samples were dried properly. The weight loss of the sample over the time was used to indicate the degradation rate of soil burial test. And the weight loss % was calculated according to the Eq. 3.1 to show the amount of biodegradation done.

$$\text{Weight loss} = \frac{(\text{Initial weight} - \text{Final weight})}{\text{Initial weight}} \times 100 \quad \dots \text{Eq. 3.1}$$

Results and Discussion

Various results obtained for this study is summarized in the below headings.

Evaluation of Properties of Biodegradable Sheet

Sheet Thickness and Tensile properties

Uniform thickness is preferable for any plastic film or sheet. The thickness was measured at three different points of each sample. The mean thickness was then calculated using gauge and mean thickness of T₁ is 1.171 mm and T₂ is 1.206 mm.

All the readings are shown in the Table 4.2 and are Graphically plotted in Graph 4.3. Approximate results were obtained in the studies conducted by Sadhu *et al.*, (2014), whose work was on preparation of Starch-chitosan blend sheet and study of its mechanical properties, according to Siddaramaiah *et al.*, (2003). From the ANOVA analysis of Table 4.2, it can be concluded that significant effect of treatments on thickness of samples were observed. Tensile properties are shown in the Table 4.3.

Table.1 Experimental Plan

VARIABLES	LEVEL	DESCRIPTION
Product	1	Biodegradable Sheet
Method	1	Compression Moulding Methods
Constituents	4	Chitosan, Arrowroot Starch, Citric Acid & Glycerol
Treatments	2	T ₁ , T ₂
Mechanical Properties	2	Thickness, Tensile Strength
Degradability Test	1	Soil Degradability Test
Replications	3	Three replications

Table.2 Different Composition of Biodegradable Sheet

TREATMENT	CHITOSAN (%)	STARCH (%)	GLYCEROL (g)	CITRICACID (g)	WATER (ml)
T ₁	50	50	35	3	45
T ₂	30	70	35	3	45

F- test	S	S	S	S	S
S. Ed. (±)	0.833	0.705	0.804	0.397	0.255
C. D. (P = 0.05)	1.765	1.495	1.706	0.842	0.540

S= Significant

Table.3 Tensile Strength Properties

Treatment	Batch	Dimension(mm)	Thickness(mm)	Tensile (MPa)	Elongation at Break(%)
T₁	1	30x80	1.18	5.62	143
	2	30x80	1.15	4.32	168
	3	30x80	1.20	4.48	156
Mean		30x80	1.17	4.80	155.6
T₂	1	30x80	1.19	3.46	218
	2	30x80	1.21	4.19	189
	3	30x80	1.22	3.58	203
Mean		30x80	1.20	3.74	203.3

F- test	S	S	S
S. Ed. (±)	0.006	0.168	4.035
C. D. (P = 0.05)	0.013	0.357	8.555

Fig.1

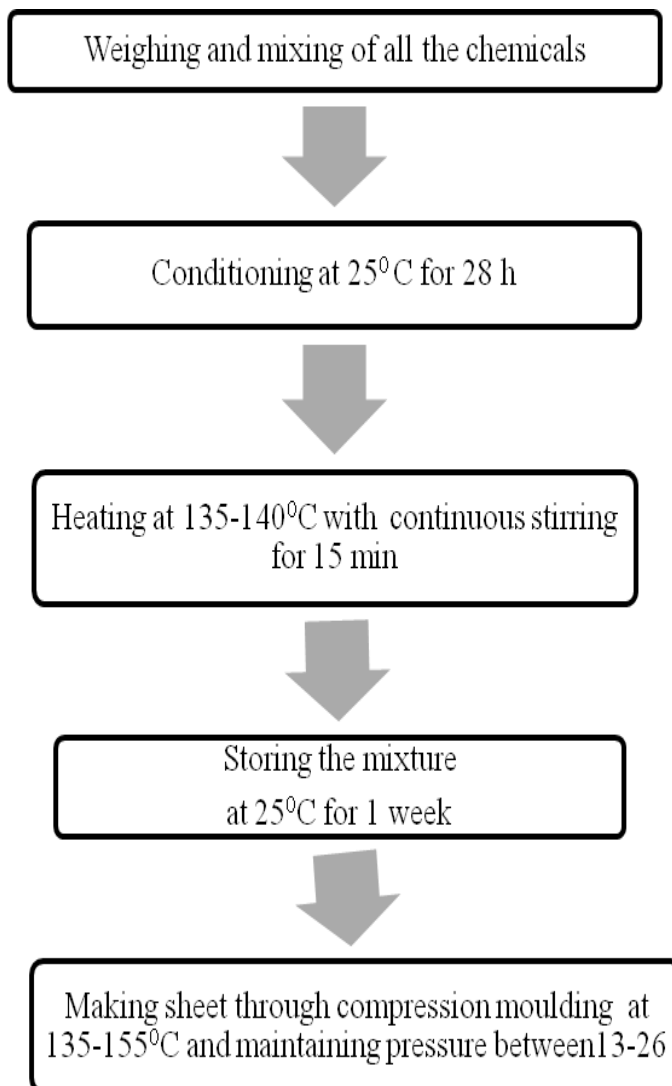
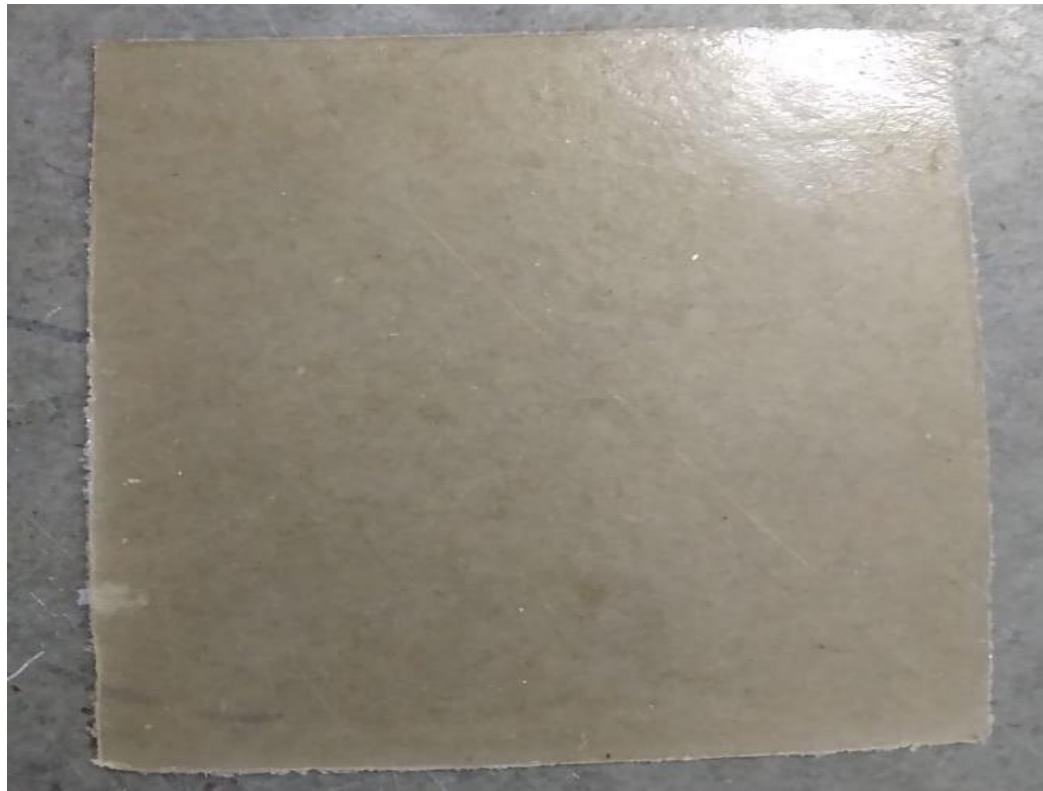


Fig.2 Prepared biodegradable sheet



From ANOVA analysis of tensile strength Table 4.3, it is evident that the calculated value of F due to treatment is smaller than the tabulated value at 5% probability level.

Therefore it can be concluded that significant effect of treatment on tensile strength of samples were observed. ANOVA analysis of Elongation at break (%) (Table 4.3) also showed that there were significant effect of treatment on Elongation at break (%) of samples.

Test for Biodegradability

After preparation of the sheet, Biodegradability test was carried out to check the biodegradability property of the sheet which was obtained by compression moulding.

To check the biodegradability, Soil burial test was carried out in which initial weight was taken before putting into the soil and then weight was taken after 2nd and 4th week after preparation of the sheet and the weight loss calculated by the below mentioned formula

$$\begin{aligned} & \text{Weight loss} \\ & = \frac{(\text{Initial weight} - \text{Final weight})}{\text{Initial weight}} \times 100 \end{aligned} \quad \dots \text{Eq. 4.1}$$

All the readings of the degradation are shown in the Table 4.4. Result showed that sample T₂ showed the higher percentage of weight loss, i.e. 19 % and T₁ has the lower percentage of weight loss, i.e.13% after 4weeks. The values were represented graphically in the Fig 4.6 shows degradability after four week of observation. From ANOVA analysis of biodegradability study Table 4.4, it can be concluded that there is significant effects of treatments and days on biodegradability, However replication of each treatment have no significant effects on biodegradability.

Biodegradable polymers will have a greater role in the packaging sector. Biodegradable plastics and other biodegradable or compostable materials like paper, food and garden waste are generally unsuitable for landfill due to their potential to release methane under anaerobic conditions. Biodegradable bio plastics are more suitable for biological waste treatment through industrial and/or domestic composting and, subject to further demonstration, potentially in anaerobic digestion systems. Petroleum polymer should be replaced with biopolymer to save the environment and to maintain the ecosystem properly. However, biodegradable polymers can-not be solely used and must be blended with other biodegradable additives to enhance their mechanical properties and shape stability. Although the properties of chitosan and arrowroot starch blend films containing different plasticizers. However new additives should be introduced to support bio based packaging material production. In this research, compression moulding technique was used to produce the biodegradable sheet by chitosan and arrowroot starch. Generally casting method is practised mostly to produced biodegradable packaging film or sheet but in terms of bulk production we should focus on bulk amount of production by extrusion method for continued and bulk amount of production but in that case more research is needed to increase the stretchability. Looking onto the situation compression moulding technique had been used which is also a important production technique for plastic and it also can be used for commercial production as like extrusion technique.

Chitosan and arrowroot starch biodegradable polymer sheet was obtained with glycerol and citric acid and water through compression moulding method.

All the prepared sheets were evaluated for thickness, tensile strength and

biodegradability.

Thicknesses of the both sheet were 1.17 and 1.20 mm.

Tensile strengths of the both sheet were 3.74 MPA and 4.80MPA. Sheet denoted as T₁ has the higher tensile strength and compared as T₂. Elongation at break % of the both sheet were 155.6% and 203.3%. Sheet denoted T₁ has the lowest elongation at break % and sheet labelled T₂ has the highest.

Performance evaluation of biodegradable films was done by soil burial method for 30 days at an interval of 15 days. Sheet denoted T₂ has the highest percentage of decomposition, i.e. 19% after 30 days.

Chitosan and arrowroot starch biodegradable composite sheet was obtained in two proportions T₁ and T₂ with three replications of each through compression moulding method.'

Mechanical properties of T₁ shown better result as compared to the T₂ in which chitosan and starch was taken in equal proportions

Elastic modulus of T₂ was greater than T₁.

Biodegradability of the sample T₂ also showed better result compared to the sample T₁.

There are a large number of sectors where biodegradable plastics may find use. All sectors require environmentally friendly polymers. Because the level of biodegradation may be tailored to specific needs, each industry is able to create its own ideal material. The Environmental responsibility is constantly increasing in importance to both consumers and producers. For the producers and researchers biodegradable plastic materials, this is a key advantage. Biopolymers limit carbon dioxide emissions

during creation, and degrade to organic matter after disposal. Although synthetic plastics are a more economically feasible choice than biodegradable ones, an increased availability of biodegradable plastics will allow many consumers to choose them on the basis of their environmentally responsible disposal. Biodegradable plastics containing starch and/or cellulose fibres or chitosan appear to be the most likely to experience continual growth in terms of demand. Microbiologically grown plastics are scientifically sound, and a novel idea, but the infrastructure needed to commercially expand their use is still costly, and inconvenient to develop. Time is of the essence for biodegradable polymer development, as society's current views on environmental responsibility make this an ideal time for further growth of biopolymers. From this research it can be concluded that chitosan and arrowroot starch can be a good combination for making biodegradable plastics as arrowroot starch is less expensive and easily available in anywhere. It is also felt that more research also needed to increase its tensile properties to make it more versatile.

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