

# Valorization Of Domestic Kitchen Wastes Through Formulation Of Ecofriendly Polysaccharide Based Food Packaging Bioplastics

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The growing demand on bioeconomy is associated with immense utilization of agro wastes derived bioplastics as a renewable, biodegradable, environment friendly alternative for petrochemical based food packages. The study aims on valorization of unconventional starch and cellulose sources of domestic kitchen wastes in fabrication of starch pectin-based and cellulose pectin-based food packaging bioplastics. Surface topography of starch pectin-based biofilms is much different from cellulose pectin-based biofilms. The study on physicochemical characteristics reveals that the hygroscopicity of biofilms increases with increasing concentration of starch, cellulose and pectin, while density decreases simultaneously. The control film C1 with 1g starch exhibited maximum moisture content of 32.5% but recorded minimum density of  $0.015 \pm 0.002$  g/cm<sup>3</sup>. The higher water solubility of cellulose pectin-based biofilms ensures applicability as edible food coating material while starch pectin-based biofilms are suitable for improving the self-life of food as packaging films with incorporation of suitable preservatives. Complete biodegradation of cellulose pectin-based biofilms takes place within 3 days while an appreciable soil burial weight loss was reported almost for all starch pectin-based combinations. Among cellulose pectin-based combination biofilms F, C, I was considered best, while among the starch pectin-based combination biofilm C1, G1, E1 was considered best. Thus, this approach is not only a sustainable alternative for food preservation but also a novel initiative to control food bio wastes based environmental pollution.

**Keywords:** Active Packaging; Biodegradability; Biofilms; Environmental Sustainability; Green Chemistry.

High molecular mass and hydrophobicity of conventional synthetic polymers inhibits the natural degradation process thus accumulates in the environment becoming a potential source of pollution by destroying the ecological balance<sup>1</sup>. The debasement techniques of synthetic polymers which mainly includes landfills, substance treatment, incineration, disposal in water bodies leads to greenhouse gas emission, global warming,

release of carcinogenic chemicals, air and marine pollution. According to plastic Europe (2020) about 40.5% of the total synthetic polymers especially plastics are utilized in food packaging applications which has detrimental effect on biodiversity<sup>2</sup>. The process of reengineering the total carbon spine polymers or development of biodegradable plastics from renewable energy sources and to identify possible micro green algae to biodegrade

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the plastics are some of the potential techniques to overcome plastic pollution<sup>3</sup>. Thus, in this forth coming era of bioeconomy and increasing stress on environmental sustainability had led the development of renewable, eco-friendly, bioplastics as an alternative to conventional petrochemical based food packaging materials. The biobased films or coating are semi permeable barriers to gases and water vapor fabricated from renewable natural polymers like polysaccharides, protein, lipids that acts as a sustainable approach for food preservation<sup>4</sup>. Bioplastics also acts as a carrier for various additives and bioactive compounds to reduce microbial spoilage, counteract loss of sensory and nourishing features of the product, improving food quality by increasing the shelf-life<sup>5</sup>.

Among the natural biopolymers, polysaccharides especially starch, cellulose, pectin, gums, carrageenan, alginate are widely utilized in biofilm formulation because of their renewability, ease of availability, affordability, non-toxic nature, ability to blend between them also with other compounds<sup>6</sup>. The molecular weight, type of molecular linkage, shape and degree of polymerization of the biodegradable compound influences the physicochemical properties of the film matrix<sup>5</sup>. Generally unconventional or underutilized sources of hydrocolloids extracted from agro industrial wastes and food processing wastes are widely utilized in bioplastic preparation. This approach seeks to promote valorization of agro industrial by products and reduce environmental pollution<sup>7</sup>. Previous studies included preparation of edible and non-edible biofilms from non-conventional sources like cranberry extract, fruit and vegetable peels, banana peels, soyabean waste, cassava bagasse, fish skin gelatin etc<sup>8</sup>. Domestic kitchen wastes mainly include vegetable and fruit peels, husks, residue, pomaces, bagasse, seeds and pits which are rich sources of biopolymers like starch, cellulose, pectin, hemicellulose etc that can be utilized in product development<sup>4</sup>.

Starch is the most versatile and abundantly available polysaccharide that plays pivotal role in bioplastic preparation. Starch is a polymeric carbohydrate comprising of  $\alpha$  1,4 linked D-glucose units present principally in two major macromolecular structure: amylose (linear) and amylopectin (branched) arranged in semi crystalline

and amorphous form<sup>9</sup>. While cellulose is a linear homopolysaccharide of high molecular weight composed of  $\beta$  D- glucopyranose unit linked by a 1,4 bond, that barely mixed freely with common solvent. Often cellulose microfibers and nanofibers are incorporated into starch films to improve its tensile strength and water barrier properties<sup>10</sup>. Pectin is a complex heteropolysaccharide comprising linear backbone of  $\alpha$  (1,4)-D-galacturonic acid partially esterified with methanol as a periodic interruption to L rhamnose forming an irregular structure with neutral sugars. Pectin is well known for its gelling property thus forming a continuous matrix for biofilm formation<sup>11</sup>. The materialistic approach of bioplastic fabrication involves gel formation from biopolymers and simultaneously conversion of gel to thin films in association with the incorporation of secondary component such as plasticizers<sup>12</sup>. Plasticizers of low molecular weight like polyols plays significant role in increasing flexibility with decreasing the strength of hydrogen bonds between the polymers by sliding in between the polymeric chains of the molecule<sup>13</sup>.

To now, although there have been detailed studies on bioplastics developed from conventional polysaccharide sources, but unconventional polysaccharide sources still now remained underexplored in formulation of bio packages. This was the first time when randomly collected easily accessible domestic kitchen wastes was utilized as the source of biopolymers like starch and cellulose for development of biodegradable packaging materials. The main aim of the study is valorization of domestic kitchen wastes in fabrication of starch pectin-based and cellulose pectin-based food packaging bioplastics as a novel initiative to control environmental pollution caused by petrochemical-based packaging materials.

## MATERIALS AND METHODS

Plant materials: Domestic kitchen wastes rich in starch sources (potato peels, carrot peels, pumpkin peels, tapioca peels, colocasia peels, ripe papaya peels), cellulose sources (banana stem wastes, drumstick peels, ridge gourd peels, ivy gourd peels, discarded leafy vegetables, unripe jackfruit wastes, discarded beans etc).

Other materials: commercial pectin (putrix), sodium hydroxide, sorbitol, mannitol, glycerol, sodium bicarbonate all purchased from Sigma- Aldrich (USA).

#### Methods

##### Extraction of Starch from Kitchen Wastes

Starch from fruits and vegetable husk was extracted according to the method reported by Cuellar E.A. Initially, the husks were selected, manually chopped into small pieces, cleaned and disinfected with 2% sodium bicarbonate solution. Further the husks were macerated in blender along with distilled water (1:2 v/v) for 3 min and the homogenate was washed with distilled water while passing through thin cotton mesh cloth. The starch containing filtrate was resuspended in distilled water (1:4 v/v) and left to combine for sometimes. The settled starch at the bottom of the beaker was separated from the supernatant and further redistributed in distilled water. This washing process was repeated several times until complete decolorization of the starch has taken place. After which the collected starch sediments were subjected to drying in hot air oven at 70-80°C for 12 h<sup>4</sup>.

##### Extraction of Cellulose from Kitchen Wastes

Domestic kitchen wastes rich in cellulose were selected in extraction of cellulose according to the method demonstrated by Gupta. with certain modifications. The process of cellulose isolation was classified into 4 different steps:

##### Moisture Removal and Milling

Initially the vegetable and fruit husks were subjected to removal of all extractives by soaking in distilled water for 3 h followed by drying in hot air oven at 70°C for 2 days. The dried biomass was grounded into fine powder by mixer grinder and passed through 60 mesh sieves.

##### Alkali Treatment

30g of powdered husk was treated with 2% NaOH solution on hot plate at 80! for 2-2.5 h followed by washing and filtering in distilled water with the utilization of cotton mesh cloth. The supernatant was discarded and the filtered residue was dried in hot air oven overnight at 70°C

##### Acid Hydrolysis

After the removal of lignin and other impurities through alkali treatment the residual biomass was subjected to acid hydrolysis with utilization of 4% H<sub>2</sub>SO<sub>4</sub> (v/v) at 85°C for 2 h.

Further the treated fiber was washed and filtered in aquadest to attain pH of 7 followed by oven drying at 70°C.

##### Bleaching

Decolorization of the residual biomass fiber was carried out by the treatment with 4% glacial acetic acid at 80°C for 2 h. During washing process, the pH was maintained in between 3-4 followed by drying of the decolorized cellulose at 70°C for 24 h<sup>14</sup>.

##### Fabrication of Starch and Cellulose based biofilms

The biofilms are prepared with combination of commercial pectin by solvent casting technique as the method demonstrated by Akshaya R. with certain modifications<sup>15</sup>. Separate series of starch and cellulose biofilms were fabricated by mixing pectin, extracted starch and plasticizers at different concentration. The concentration of cellulose, pectin and plasticizers was determined by as per data and trials. For starch pectin-based biofilms the control film was prepared by dissolving 1g starch, 0.4g pectin, 1g sorbitol as plasticizer in 20 ml distilled water. while, in case of cellulose pectin-based biofilms comprised of 0.7g pectin, 0.5g cellulose and 1g sorbitol. The film forming solution was heated at 90°C for 15 min on hot plate followed by continuous agitation. Finally, the solution was casted on glass plate and subjected to drying at ambient temperature for 30 h. The dried films were peeled off from the plate and stored in desiccator for further tests<sup>16</sup>.

##### Characterizations of the Fabricated Biofilms

##### Physical Characteristics

##### Visual Inspection

Visual observation was utilized in determining the apparent characteristics of the biofilms as demonstrated by Judawisastra. Each sample biofilms were folded several times. The colour and texture of the biofilms were noted<sup>17</sup>.

##### Moisture Content

The moisture content of the biofilms was determined by AOAC method by utilizing oven drying technique. Generally, three replicate of each sample was taken and initial weight  $W_i$  was recorded by using digital weighing balance. Then final weight  $W_f$  was determined after drying the samples at 105°C until a constant weight was obtained<sup>18</sup>. The moisture content was calculated by Eq. 1,

$$\text{Percentage of moisture} = (W_i - W_f) / W_i \times 100 \quad \dots(1)$$

Where,  $W_i$  is the initial weight (in g) and  $W_f$  is the final weight (in g)

#### Density

The ASTM D-792-00 was utilized in determination of the density of the prepared biofilms. The films were weighted (m) for taking in account the preliminary dry matter of the biofilms then immersed into solvent of volume (v). The density (d) was determined from the equation given below. The test was carried out in triplicate<sup>18</sup>.

$$d = m/v \quad \dots(2)$$

#### Water Barrier Property

##### Water Absorption Capacity

The water absorption capacity of the biofilms was determined according to Muhammad as per the standard method for determining water absorption capacity of plastics, ASTM D570. The biofilms were subjected to oven drying at 50°C for 5 h and the initial weight was recorded after cooling at room temperature. The biofilm specimens were immersed in beaker containing distilled water and left undisturbed for 24 h at 23°C. After which the specimens were replaced from water one at a time, wiped off carefully with tissue paper and weighted immediately. The test was performed in triplicate and the mean value was recorded. The water absorption capacity and loss of soluble matters were calculated according to the equations given below<sup>19</sup>,

$$\text{Water absorption \%} = (\text{wet weight (g)} - \text{initial dry weight (g)}) / (\text{wet weight (g)}) \times 100 \quad \dots(4)$$

$$\text{Loss of soluble materials \%} = (\text{initial dry weight (g)} - \text{final dry weight (g)}) / (\text{initial dry weight (g)}) \times 100 \quad \dots(5)$$

#### Thermal Property

##### Burn Test

The burn test was carried out according to the ASTM D3801. The biofilm specimens were subjected to burning on Bunsen burner and the

odour, colour of flame, speed of burning of the fabricated bioplastics was observed and recorded accordingly. Each specimen was tested thrice<sup>19</sup>.

##### Melt Test and Wire Test

The melt and wire test are two criteria considered for determining the thermal property of the biofilms. In melt test lightened and immediately extinguished match stick was brought in contact with the film specimen. The observation was recorded accordingly. Each sample was tested in triplicate.

In wire test red hot copper wire was brought in contact with film specimens and the observations were recorded accordingly.

#### Environmental Analysis

##### Biodegradation Study

The biodegradation of the fabricated bioplastics was studied through soil burial test as demonstrated by Tarique. The test was carried out in triplicate, where pre weighted each sample films of 10 mm × 10 mm was buried separately 100 cm underneath the garden soil of the university campus under restrained humid condition. The degraded biofilm sample was taken out from soil carefully at an interval of 48, 72, 96 h weighted accordingly to determine the weight loss during biodegradation process<sup>20</sup>. The percent of weight loss is calculated by the formulae given below,

$$\text{Percentage of weight loss} = (W_o - W_t) / W_o \times 100 \quad \dots(6)$$

Where,  $W_o$  is the initial weight of the biofilms prior burring and  $W_t$  is the final weight of the biofilms after burring.

##### Food Packaging Application

The application of the biofilms as food packaging material was studied as per the method demonstrated by Che Hamzah. For this fresh apple of optimum maturity were selected washed with tap water and sanitized using 3% acetic acid solution for 3 min, drained and dried properly using clean muslin cloth. Then freshly cut slice of apple were wrapped using the prepared biofilms and sealed properly from all sides by using thread to avoid coming in contact with air. The biofilm wrapped fruit slices along with unwrapped fruit slice as control was kept at ambient temperature for 72 h.

After 3 days, the fruit slices were unwrapped and visually analyzed in comparison with the control slice. The test was carried out in replicate by selecting best film from each series<sup>21</sup>.

## RESULTS

### Physical Characteristics

#### Visual Test

The study on surface topography reveals that the fabricated starch-based biofilms were

flexible, homogenous, uniform, smooth, translucent and slight yellowish to whitish in colour. While cellulose based biofilms were brownish in colour with irregular, rough, non-uniform texture. Generally, the dark brownish colour of the cellulose-based biofilms lightens under unpolarized whit light with gradual decreasing the concentration of cellulose and increase in concentration of pectin. This was similar to the findings of A. González Moreno, who observed that the sparkly orange colour of the biofilms faded with increase in

**Table 1.** Different Combinations of Cellulose-Pectin Based Biofilms

Combination	Code	Cellulose (g)	Pectin (g)	Sorbitol (plasticizer) (g)	Mannitol (plasticizer) (g)	Glycerol (plasticizer) (ml)
Pectinmodification	A	0.5	0.4	1	-	-
	B	0.5	0.5	1	-	-
Plasticizer modification	E	0.5	0.7	-	1	-
	F	0.5	0.7	-	-	1
Cellulose modification	H	0.6	0.7	1	-	-
	I	0.7	0.7	1	-	-
Control	C	0.5	0.7	1	-	-

**Table 2.** Different Combinations of Starch-Pectin Based Biofilms

Combination	Code	Starch (g)	Pectin (g)	Sorbitol (plasticizer) (g)	Mannitol (plasticizer) (g)	Glycerol (plasticizer) (ml)
Starch modification	A1	0.75	0.4	1	-	-
	B1	0.5	0.4	1	-	-
Plasticizer modification	D1	1	0.4	-	1	-
	E1	1	0.4	-	-	1
Pectin modification	F1	1	0.3	1	-	-
	G1	1	0.5	1	-	-
Control	C1	1	0.4	1	-	-

**Table 3.** Physical Characteristics of Starch Pectin and Cellulose Pectin-Based Biofilms

Plasticizers	Starch-pectin based biofilms	Cellulose-pectin based biofilms
Glycerol	Homogeneous, glossy, uniform, translucent, flexible, whitish to pale yellowish colour	Non uniform, irregular, flexible, glossy, translucent, heterogenous, brownish colour
Sorbitol	Homogeneous, non-glossy, uniform, flexible, translucent, whitish to pale yellowish colour	Non glossy, flexible, uniform, translucent, brownish colour
Mannitol	Broken, rigid, wavy, non-flexible, brittle	Rigid, stiff, broken, flaky, brittle.

pectin content in pectin cellulose nanocrystal bio composites<sup>17</sup>. The variation of colour between starch and cellulose biofilms existed due to the difference in colour of the starch and cellulose extracted. While the textural difference might be due to the variation in the nature of bonding between the

extracted biopolymers, pectin and the plasticizers. The starch-based films were homogeneous, free from projections and wrinkles since the plasticizers might have improved the crosslinking between starch molecules and pectin. This is similar with the findings of Wu Z, who prepared cornstarch-based

**Table 4.** Study on Density of Starch Pectin and Cellulose Pectin-Based Biofilms

Cellulose-pectin based biofilms		Starch -pectin based biofilms	
Sample code	Density (g/cm <sup>3</sup> ) mean $\pm$ SD	Sample code	Density (g/cm <sup>3</sup> ) mean $\pm$ SD
A	0.110 $\pm$ 0.008	A1	0.059 $\pm$ 0.009
B	0.064 $\pm$ 0.004	B1	0.078 $\pm$ 0.007
C	0.047 $\pm$ 0.011	C1	0.015 $\pm$ 0.002
E	0.091 $\pm$ 0.008	D1	0.111 $\pm$ 0.010
F	0.123 $\pm$ 0.008	E1	0.063 $\pm$ 0.014
H	0.035 $\pm$ 0.009	F1	0.033 $\pm$ 0.012
I	0.022 $\pm$ 0.011	G1	0.012 $\pm$ 0.004

Data expressed as mean  $\pm$  SD of three independent experiments

**Table 5.** Study on Water Absorption Capacity of Starch Pectin-Based Biofilms

Combination	Code	Water absorption capacity (%) (Mean $\pm$ SD)	Percentage loss of soluble matters (Mean $\pm$ SD)
Starch modification	A1	148.53 $\pm$ 4.39	64.9 $\pm$ 4.49
	B2	130.97 $\pm$ 4.12	59.57 $\pm$ 2.67
Plasticizer modification	D1	124.73 $\pm$ 4.20	73.63 $\pm$ 3.90
	E1	102.26 $\pm$ 4.10	75.67 $\pm$ 3.09
Pectin modification	F1	162.03 $\pm$ 3.39	63 $\pm$ 1.00
	G1	142.13 $\pm$ 4.11	67 $\pm$ 1.52
Control	C1	193.67 $\pm$ 2.51	85.7 $\pm$ 2.20

Data expressed as mean  $\pm$  SD of three independent experiments

**Table 6.** Study on Thermal Property of Starch Pectin-Based Biofilms

Code	Odour	Flame colour	Sparks	Speed of burning	Melt test (on contact with match stick)	Wire test (on contact with red hot copper wire)
A1	Present	Orange	Present	Very fast	Yes	Yes
B2	Present	Yellow	Present	Very fast	No	No
D1	Absent	Yellowish orange	Absent	Rapid	Yes (very rapidly)	Yes (very rapidly)
E1	Present	Yellowish orange	Present	Slow	No	No
F1	Present	Yellowish orange	Present	Fast	Yes	Yes
G1	Present	Yellowish orange	Present	Fast	No	No
C1	Present	Yellowish orange	Present	Very fast	No	No

biofilms in combinations with PVA for packaging applications<sup>22</sup>. While cellulose based biofilms were irregular, heterogenous comprising few holes since cellulose fibres remains suspended in the matrix of pectin and plasticizer instead of completely dissolving in the solution unlike starch molecule. Also, during the oven drying process one particle might have end up on top of the other as a result of self-stratification due to the presence of complex

mixture of dissolved and undissolved polymer which might be responsible for the presence of small pores. This was similar with the findings of Batori, who prepared pectin cellulose-based biofilms from orange wastes by using different drying techniques<sup>23</sup>.

Again, utilization of different plasticizers is associated with producing variations in the textural appearance of the biofilms. During

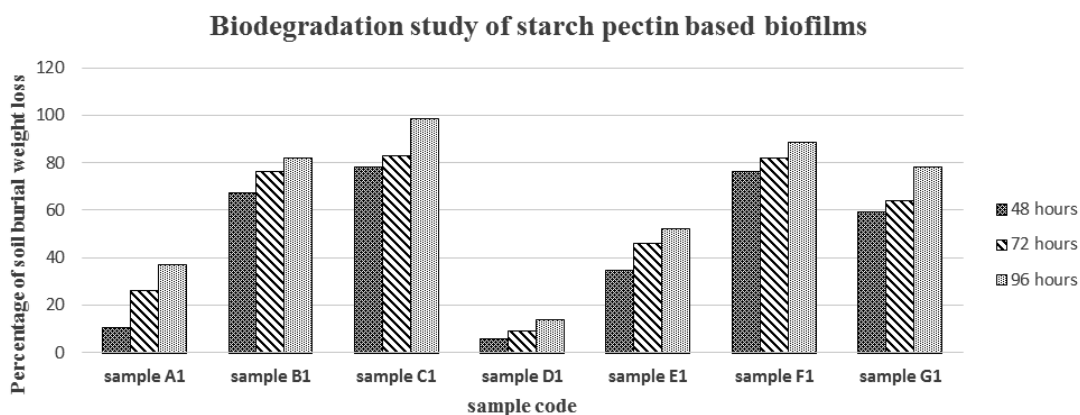


Fig. 1. Soil Burial Weight Loss of Starch Pectin-Based Biofilms

Table 7. Study on Thermal Property of Cellulose Pectin-Based Biofilms

Code	Odour	Flame colour	Sparks	Speed of burning	Melt test (on contact with match stick)	Wire test (on contact with red hot copper wire)
A	Absent	Yellowish orange	Present	Slow	No	No
B	Absent	Yellow	Present	Very slow	No	Yes
E	Absent	Yellowish orange	Absent	Slow	Yes	Yes
F	Absent	Yellowish orange	Present	Rapid	No	Yes
H	Absent	Yellowish orange	Present	Slow	No	No
I	Absent	Yellowish orange	Absent	Very slow	No	No
C	Absent	Yellowish orange	Present	Slow	No	No

Table 8. Shelf-life Analysis of The Wrapped and Unwrapped Food




Sample Code	Color	Texture	Odor Change	Visiblemicrobial Growth
Unwrapped slice	Excessive browning	Dry	No	Yes
I	Slight browning	Soggy	No	No
C	Slight browning	Slight dryness	No	No
F	Slight browning	Soggy	No	No
C1	Slight browning	Dry	No	No
E1	Slight browning	Dry	No	No
G1	Slight browning	Dry	No	No

biofilm preparation plasticizers such as polyols interferes with the intermolecular packaging of the biopolymers resulting in flexibility by boosting the movement of the molecules. The table given below represent the nature of starch and cellulose based biofilms produced with different plasticizers<sup>18</sup>.

Generally, glycerol excels sorbitol and mannitol in its plasticizing ability during the fabrication of biofilms. This was similar with the findings of Mitrea who observed that triols

like glycerol and diols like 1,3-propanediol (PDO) and 2,3-butanediol (BDO) is associated with maintaining good behavioral property like elasticity and flexibility and external features like uniform, soft texture<sup>24</sup>. Unlike glycerol and sorbitol, mannitol forms brittle, rigid, non-flexible films since large molecular size of mannitol might not allow it to completely interpose in between the molecular spaces of the polymers. Also, mannitol plasticized films are prone to premature rupture

**Table 9.** Study on Best Cellulose Pectin based Biofilms

Codes	Cellulose pectin-based biofilms	Description
F		Combination F is best among the plasticizer modification series of cellulose pectin-based biofilms. Heterogeneous, translucent, uniform, free from cracks and pores, glossy
C		Combination C is considered best among the pectin modification series of cellulose pectin-based biofilms. Heterogeneous, translucent, higher moisture content, best in food packaging application
I		Combination I is considered best among the cellulose modification series of cellulose- pectin based biofilms. Smooth textured, uniform, higher strength



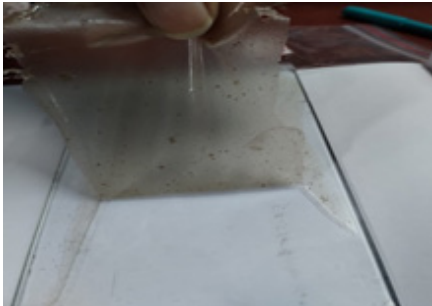
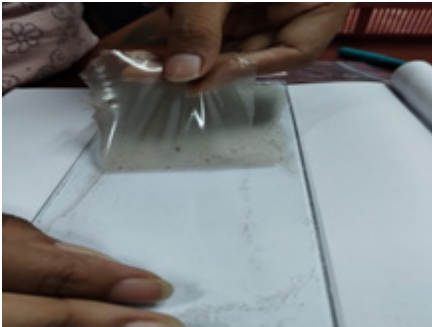

points as an effect of crystal in the structure. Similar observation was recorded by Santana who studied on physical evaluation of biodegradable films of calcium alginate plasticized with polyols<sup>25</sup>.

#### Moisture Content

The study on the moisture content of the starch pectin-based and cellulose pectin biofilms reveals that on varying the concentration of

starch and cellulose the moisture content changes appreciably. The control film C1 with 1g starch showed highest moisture content of 32.5% while biofilm with 0.5 g starch is lowest in moisture content of 3%. Similarly in case of cellulose pectin-based biofilms the control film with 0.5g cellulose possess lower water content of 13.03% than biofilm H and I. The increase in hydrophilicity

**Table 10.** Study on Best Starch Pectin-based biofilms

Codes	Starch pectin-based biofilms	Description
C1		Combination C1 is best among the starch modification series of starch pectin-based biofilms. Homogeneous, transparent, uniform, free from cracks and pores, higher moisture content, high water absorption capacity, best in food packaging applicability
E1		Combination E1 is considered best among the plasticizer modification series of starch pectin-based biofilms. Homogeneous, transparent, uniform, glossy, free from cracks and pores
G1		Combination G1 is considered best among the pectin modification series of cellulose- pectin based biofilms. Pale yellowish in colour, flexible, smooth textured

of the biofilms with increase in starch and cellulose content might be the responsible for high moisture content. Previous study of Datta also showed higher water content of corn starch film in comparison with biofilm prepared from potato starch due to higher hygroscopic nature of corn starch<sup>20</sup>. Again, with increasing pectin content the moisture content of the biofilms increases simultaneously both in case of starch pectin-based and cellulose pectin-based biofilms. The elevation in hygroscopic nature of biofilm with increase in pectin content might be responsible for increase in water content of the biofilms<sup>26</sup>. Both for starch pectin-based and cellulose pectin-based biofilms maximum moisture content was recorded when glycerol was utilized as plasticizer. This might be due to high water affinity of glycerol due to the presence of hydroxyl group that easily forms hydrogen bonds. This finding resembled the studies of Tarique who found that the moisture content of glycerol plasticized arrowroot starch biofilms elevates with increase in glycerol concentration<sup>18</sup>.

#### **Density**

Both for starch pectin-based and cellulose pectin-based biofilms the density gradually decreases with increase in concentration of starch, cellulose and pectin respectively. This might be due to the increase in thickness of the biofilms with increase in biopolymer content<sup>18</sup>. Incomplete gelatinization resulting in change of viscosity and non-uniform casting and drying technique might be responsible for affecting the thickness of the biofilms. This was similar to the findings of Ayquipa Cuellar who found that the density increases with decrease in thickness in biofilm prepared from potato husk in combination with prickly pear mucilage<sup>4</sup>.

#### **Water Barrier Property**

##### **Water Absorption Capacity**

The study on water absorption capacity and water droplet test reveals that cellulose pectin-based biofilms completely dissociates in water within 5-10 min leaving the insoluble cellulose remanent. This might be due to high solubility of pectin and larger size cellulose fibers resulting the biofilms to lose its structure. This is similar to findings of Efthymious who found that solubility of biofilms prepared from sunflower protein isolate is higher when incorporated with larger size cellulose nano fiber derived from soyabean straw than

biofilms with nanosized cellulose fibers obtained after enzyme hydrolysis<sup>27</sup>. The strong structural bonding of starch pectin-based biofilms does not allow it to dissociate completely when conditioned for 24 h but exhibited high-water absorption capacity<sup>18</sup>. For starch pectin-based biofilms water uptake capacity ranges between 102.26 %- 193.67%. While loss of soluble matters ranges between 59.56% to 87.56%. The water uptake generally increases with increase in immersion period but gradually reaches a state where water absorption capacity becomes constant<sup>19</sup>. The water uptake capacity gradually increases with increase in concentration of starch. The control film C1 with 1 g starch has maximum water absorption capacity of 193.67%. This might be due to increase in free hydroxyl groups in biopolymer matrix that results in water absorption by facilitating hydrogen bonds between water molecule and polymer chain<sup>18</sup>. In case of modifying the pectin concentration the biofilms with 0.3g, 0.5g pectin exhibits 162.03%, 142.13% water uptake while control C1 with 0.4g pectin exhibit 193.67% water absorption. This might be due to the combined effect of area of exposed surface area, thickness, fiber content, orientation, permeability etc influencing water uptake property<sup>20</sup>. Among the plasticizers used biofilms plasticized with sorbitol facilitated high water uptake capacity than glycerol plasticized biofilms. This might be due to the presence of stronger hydrogen bond between glycerol and starch molecule which resists the water molecule to combine with plasticizers and starch molecules. This was similar with the findings of Tarique who reported that with increase in concentration of glycerol the water absorption capacity of arrowroot starch biofilms decreases<sup>18</sup>. Again, loss of soluble matters or solubility of biofilms increases with increase in starch concentration. This might be due to the increase in amylose content of biofilms with increase in starch concentration which elevates hydrophilicity of the biofilms. This was similar to the findings of Basiak who found that the solubility of films in water was about 14.52%, 30.16%, and 44.76% for potato, wheat, and corn starch, respectively depending on the amylose content<sup>28</sup>.

#### **Thermal Property**

##### **Burn Test, Melt and Wire Test**

The table represents the observations of burn test, melt and hot wire test. Generally, four

different criteria like odour released on burning, colour of flame, presence of sparks, speed of burning along with the biofilms behavior towards thermally activated objects are considered for determining the thermal property of the biofilms. Almost all cellulose pectin-based biofilms are free from burning odour and exhibited slow to moderate speed of burning which is considered highly appreciable for being utilized as packaging materials. While starch pectin-based biofilms produce mild odour during burning. This might be due to degradation of plasticizers and starch polymer while burning. This was similar to the findings of Muhammad, who observed that soy waste bioplastic produces faint smell of gluey like substances upon burning. Both cellulose pectin-based biofilms and starch pectin-based biofilms ignites with yellow to yellowish orange flame like plastic materials and more or less melts on coming in contact with red hot copper wire and burned match stick<sup>20</sup>. Initially during thermal degradation process loss of mass is associated with the loss of water molecule, weak bonds and low molecular weighted compound of biofilms then gradually plasticizers molecules are lost and finally residual mass are left after complete vaporization of the plasticizers<sup>29</sup>.

#### **Environmental Analysis Biodegradation Study**

Mineralization is the process of complete decomposition or degradation of complex polymers into smaller compound of low molecular weight by certain metabolic and enzymatic actions of micro-organisms and other biological conditions. This phenomenon of decomposing complex compound into its simpler form is considered highly desirable for maintaining ecological balance<sup>19</sup>.

The present study on biodegradation of fabricated biofilms revealed that cellulose pectin-based biofilms disintegrate completely into the soil within 48 hours of burial. While in case of starch pectin-based biofilms complete disintegration does not occurs within 96 hours but exhibited higher percentage of soil burial degradation. Complete biodegradation of cellulose pectin-based biofilms rapidly within 48 h might be due to high water solubility and weak structural bonding between pectin, cellulose and plasticizers makes it highly prone to microbial degradation and hydrolytic action of soil<sup>20</sup>. With increase in

starch concentration the soil burial weight loss increases simultaneously. Highest soil burial weight loss of 98.04% was observed in case of control film C1 with 1g starch. While minimum weight loss of 13.03% was observed in biofilm D1 with mannitol as plasticizer. This might be due to the strong influence of high moisture content on the elevated microbial action of the soil. Similar feature was recorded by Tarique who observed that the rate of soil burial degradation of arrowroot starch-based biofilms gets elevated with increase in hydrophilicity<sup>19</sup>. The soil is an immense source of starch degrading bacterial and fungal species like *Bacillus amyloliquefaciens*, *Bacillus firmus*, *Aspergillus niger*. Apart from hydrophilicity and solubility index the other factors influencing soil burial degradation are thickness, type of plasticizer used and amylose-amylopectin content of starch-based biofilms<sup>20</sup>.

#### **Food Packaging Applications of Biofilm**

The study on food packaging application was conducted by selecting the apparently best films from each series of combinations for wrapping freshly cut slice of apple. Also, an unwrapped slice was kept under same storage condition as a control for comparison with wrapped slices. Among cellulose pectin-based combinations biofilm C, F and I was selected from pectin modification, plasticizer modification and cellulose modification series respectively. While in case of starch pectin-based combination biofilms coded C1, E1, G1 was selected from starch modification, plasticizer modification and pectin modification series respectively. The food packaging application of biofilms was recorded by studying various parameters for self-life analysis of wrapped and unwrapped food after 72 h.

Almost in all fruit slices wrapped in starch pectin-based and cellulose based biofilms slight color changes due to browning reaction, slight changes in texture with no unfavorable odor and visible microbial growth was reported after 72 h. The occurrence of browning reaction on fruit slice might be due to absence of antioxidant and anti-microbial in biofilm formulations. While slight dryness of texture in case of fruit slice wrapped in biofilms might be due to hydrophilic nature of the biofilms that absorb excessive moisture from the sample fruit slice. This was similar to the findings of Che Hamzah who recorded that hygroscopic

nature of sago starch and red cabbage-based biofilms causes excessive dryness of tomato and apple slice when kept for 5 days. The hygroscopic nature of the biofilm makes it highly susceptible to environmental condition like temperature, relative humidity etc. Although the biofilms manage to prevent microbial attack during short storage period but for long storage period microbial contamination may occur due to high moisture content of the biofilms. Thus, certain anti-microbial, anti-oxidant and hydrophobic substances must be added in film formulation in order to improve the suitability of biofilms as food packaging materials<sup>21</sup>.

### DISCUSSION

Increasing stress on environmental sustainability had led the development of renewable, eco-friendly, bioplastics as an alternative to conventional petrochemical based food packaging materials. The study aims on valorization of unconventional starch and cellulose sources of domestic kitchen wastes in fabrication of starch pectin-based and cellulose pectin-based food packaging bioplastics. The study on surface topography reveals that the fabricated starch-based biofilms were flexible, homogenous, uniform, smooth, translucent and slight yellowish to whitish in colour. While cellulose based biofilms were brownish in colour with irregular, rough, non-uniform texture. The control film C1 with 1 g starch showed highest moisture content of 32.5% while biofilm with 0.5 g starch is lowest in moisture content of 3%. Similarly in case of cellulose pectin-based biofilms the control film with 0.5 g cellulose possess lower water content of 13.03% than biofilm H and I. Complete biodegradation of cellulose pectin-based biofilms takes place within 3 days while an appreciable soil burial weight loss was reported almost for all starch pectin-based combinations. Thus, this approach of formulating biopolymer-based food packages can be recognized as sustainable initiative or synergistic improvisation in the traditional food packaging and preservation technology.

### CONCLUSION

The study is based on environmental sustainability approach of not only eradicating

pollution caused by non-renewable fossil fuel-based food packaging materials but also valorization of domestic kitchen wastes. The fabricated starch and cellulose based bioplastics differ in their physicochemical and functional properties. High water solubility of cellulose pectin-based biofilms ensures their wide applicability as edible active packaging materials while starch pectin-based bioplastics can effectively be utilized in enhancing the self-life of foods as food wrappers with incorporation of suitable preservatives. Rapid biodegradability of the cellulose pectin and starch pectin-based biofilms validates its utilization as ecofriendly approach for food packaging. Among cellulose pectin-based combination biofilms F, C, I was considered best, while among the starch pectin-based combination biofilm C1, G1, E1 was considered best.

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#### Ethics Statement

This research did not involve human participants, animal subjects, or any material that requires ethical approval.

#### Informed Consent Statement

This study did not involve human participants, and therefore, informed consent was not required.

#### Clinical Trial Registration

This research does not involve any clinical trials.

#### Authors' Contribution

A Chatterjee: designed the research plan, analysed the data; critically revised and

finalized the manuscript; S Laha: performed the experiments, collected the data and prepared the manuscript; Final form of manuscript was approved by both authors.

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