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Development of Biodegradable Film from Chitosan and Lemongrass Essential Oil for Food Packaging Applications

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Abstract: Biodegradable films developed from chitosan and lemongrass essential oil to reduce the use of synthetic polymers in food industry. A biopolymer chitosan was dissolved in acetic acid to form chitosan solution and lemongrass essential oil was added to that solution to improve antimicrobial properties of film. This solution was casted and dried in polypropylene petri dish and polypropylene sheet and the dried film was peeled off. Solution was prepared in different quantities (50, 70, 100, 130, 150, and 200 ml) and dried on same sized (27cm×19 cm) sheets in order to obtain required thickness of the film. An attempt was made to fabricate films with combination of chitosan and glycerol. Coloured films also developed by adding edible colours into film forming solution. Highest value of thickness was observed for, 100 ml solution in 176 cm^2 petri dish and for 300 ml solution on 513 cm^2 polypropylene sheets as 0.05 mm. L*a*b values in determination of colour of films were reasonably less indicating its transparency. Highest values of mechanical strength obtained as peak load of 73.25 N and deformation at peak load of 12.45 mm for tensile force and average peak load of 20.153 N for puncture force, was for 150 ml solution casted in 513 cm^2 polypropylene sheet whose thickness is 0.03 mm. Films formed from chitosan and lemongrass essential oil were degraded in 4 months.

Keywords: Chitosan, Lemongrass essential oil, Biodegradable film, Antimicrobial

Synthetic plastic films used in food packaging industries causing severe environmental hazards. To encounter this problem, bio-based degradable films are growing importance these days. Chitin is a biopolymer found in the exoskeletons of crustaceans (shrimps, oysters, krill, crabs, squid, and lobsters) (Priyadarshi and Rhim 2020). Chitosan is obtained by N-deacetylation of chitin in an alkaline environment (Sahib-ul-Islam et al 2017). It is a copolymer made up of β-(1-4)-2-acetamido-D-glucose and β-(1-4)-2-amino-D-glucose (Mollah et al 2016). Chitosan is a nontoxic polymer consisting of antibacterial (Jiwook et al 2018),antifungal, anti-allergenic, antimicrobial, and anti-tumor properties and due to its properties, such as selective gas permeability (only for CO2 and O2), good mechanical properties, biocompatibility, and biodegradability, is environmental friendly and is considered a good film-forming material (Chaudhary et al 2020). Chitosan has limitation on application caused by low solubility in water and thus acetic acid acts as a solvent to dissolve the chitosan.

Lemongrass essential oil extracted from lemongrass, which belongs to *Poaceae* Family (Aashima et al 2020). It exhibits Antibacterial, antifungal, antiprotozoal, anti-inflammatory, antioxidant, anti-tussive, antiseptic, anti-carcinogenic, cardio protective and anti-rheumatic properties (Naik et al 2010) and thus, addition of lemongrass essential oil helps in increasing shelf life of food products packed in these biodegradable films (Riquelme et al 2017). Glycerol is a colourless, odourless, viscous liquid that is sweet-tasting and non-toxic and have high hydrogen bond ability that leads to a good interaction with chitosan macro molecules (Maria et al 2016). Addition of glycerol made bioplastic films flexible (higher elongation at break) (Hidayati et al 2021) and easier to degrade under wet and dry soil.Artificial edible food colours or natural colours extracted from food materials like vegetables, fruits, flowers, can be used in the biodegradable films (Mollah et al 2016). These colours provide pleasing appearance and help in degrading process. This project was undertaken to develop a biodegradable film from chitosan and lemongrass essential oil to reduce the use of synthetic polymers in food industry in order to reduce environmental hazards causing by synthetic plastics.

MATERIAL AND METHODS

Chitosan-from shrimp shells 75% (deacetylated) purchased from Elegance Scientifics, Dattasai Commercial complex, RTC X Road, Hyderabad, Telangana, 500020. Lemongrass oil purchased through authorized online marketing app from R V Essential, India. Acetic acid and glycerol procured from Saphire Scientific Co. 5-4-147, Ranigunj, M.G. Road, Secunderabad, 500003. Polypropylene petri dish and polypropylene sheets were used for casting of films.

Preparation of biodegradable films from chitosan and lemongrass essential oil: By using solvent casting method, biodegradable film was developed by forming the chitosan solution. Chitosan powder was added in the diluted acetic acid (Marina et al 2019) in certain ratio (1% w/w CH_3COOH). The solution was heated at 50°C and stirred (500rpm) on hot plate magnetic stirrer until complete dissolution of chitosan in acetic acid. Then double filtration process was carried out with the help of muslin cloth. Lemongrass essential oil (Ludmila et al 2021) was added in filtered solution to get film forming solution and it was heated and stirred at 50 °C for 10 to 15 min. Coloring agents were added in some samples at this stage and again filtered to form a clear solution. Casting of the filtered solution was carried out on a petri dish or polypropylene sheet. It was dried under electrically operated blower for 24hours. Experiment was carried out in room temperature varied from 26°C to 28°C.

Chitosan solution was prepared in different compositions by dissolving chitosan in diluted acetic acid (1% w/w) and film forming solution was obtained by mixing of lemongrass essential oil (Table 1) and an attempt was made to cast solution on different types of moulds like, glass petri dish, polypropylene petri dish, iron metal dish and polypropylene sheet. Best films were formed on polypropylene petri dish and polypropylene sheets. Troubles encountered while peeling the film from glass petri dish and iron metal dish. Polypropylene petri dish and polypropylene sheet of 176 cm^2 and 513 cm^2 size respectively were used for casting of films. Film forming solution was also formed by combination of glycerol, coconut oil, lime yellow food colour and natural colouring agents like beetroot juice with chitosan solution (Table 2). Films with lemongrass essential oil were found as best due to its good physical and mechanical properties.

Physical and Mechanical Properties Analysis

Thickness of the film (mm): Thickness of the film was measured by using thickness gauge (Make: Mitutoyo, No.2046S, made in Japan). Thickness values taken at 5 random locations on each film and average value considered.

Colour of the film: The colour characterization was carried out using Spectrophotometer Hunter Lab (Make: ColorFlex EZ) colorimeter with the CIELAB system represented by: L (luminosity 0 to 100), chromaticity a* (-a green, +a red) and chromaticity b* (-b blue, +b yellow) (Santhosh et al 2018). Three readings were taken for each film and average value considered.

Tensile test: The mechanical properties were determined by tensile tests (tensile strength and elasticity) using a Texture Analyzer (BROOKFIELD, MODEL: CT3 50K). (Antonio et al 2020). The specimen used was 25 mm wide and 90 mm long, the space between the claws was adjusted to 60 mm, the velocity was 0.5 mm/s. Probe: TA-DGA and Fixture: TA-DGA

Table 1. Different composition of biodegradable films

Sample	Distilled water (ml)	Acetic acid (ml)	Chitosan (g)	Lemongrass oil (ml)	Mould used for casting
S1	100	1	1	0.05	Polypropylene petri dish
S2	70	0.7	0.7	0.035	Polypropylene sheet
S3	50	0.5	0.5	0.025	Polypropylene sheet
S4	130	1.3	1.3	0.065	Polypropylene sheet
S5	150	1.5	1.5	0.075	Polypropylene sheet
S6	300	3	3	0.15	Polypropylene sheet
S7	75	0.75	0.75	0	Polypropylene sheet

Table 2. Different compositions involved in biodegradable films

	Sample-S8	Sample-S9	Sample-S10	Sample-S11
Distilled water (ml)	100	100	75	200
Acetic acid (ml)	1	1	0.75	2
Chitosan (g)	1	1	0.75	2
Lemongrass oil(ml)	0.05	0.05	0	0.1
Glycerol (ml)	0.05	0	0	0
Lime yellow food colour (g)	0	0.1	0	0
Coconut oil (ml)	0	0	0.037	0
Beetroot juice (ml)	0	0	0	1.3
Mould used for casting	Polypropylene sheet	Polypropylene sheet	Polypropylene sheet	Polypropylene petri dish

were used to determine tensile strength of the film (Landova et al 2014).

Puncture test: The puncture force was determined using Texture Analyzer. The films were fixed to two acrylic plates with a small opening at its centre. Probe**:** TA39 and Fixture**:** TA-JPA were used to determine puncture force (Landova et al 2014).

Biodegradability of film: Biodegradability was tested in soil (Haiquan et al 2019). Biodegradable film was placed in soil and checked at 10 days interval.

RESULTS AND DISCUSSION

Thickness (mm): The highest thickness obtained for S1, S6, S11, with compositions of 100, 300, and 200 ml respectively. S1 sample casted on polypropylene petri dish (176 cm^2) S6 sample casted on polypropylene sheet (513 cm^2) and S11 sample with beetroot juice addition casted on polypropylene sheet (513 cm^2). Thickness of sample increased with the amount of solution poured on polypropylene sheet.

Colour: The L*a*b values of films were reasonably less, indicating all the films were almost transparent. Sample S9 in which lime yellow food colour was added having 100 ml composition, clearly shows yellow colour as the b value is +9.26. Sample S11 in which beetroot juice was added having 200 ml composition, showing slightly higher b value of +1.43, but the colour of this S11 sample is not homogenous and not durable as it lasted for only 1 week.

Tensile test: Highest values of peak load at tensile force and deformation at peak load observed for Sample S5 with 150 ml composition ad 0.03 mm thickness as 73.256 N and 12.45 mm respectively. Peak load at tensile force for samples S6, S1, S9 with 300, 100, and 100 ml compositions respectively were also found good and the values as, 65.21, 51.583, and 50.72 N respectively and for the same samples, deformation at peak load were observed as, 10.14, 2.93, and 2.82 mm respectively.

Puncture test: Highest values of puncture force observed for samples S5, S9 (lime yellow colored), S1, S6 with

Chitosan solution (chitosan + diluted acetic acid)

↓

Heating (50 ^{o}C) & Stirring (500 rpm) until complete dissolution (hot plate magnetic stirrer)

↓

Double filtration using muslin cloth

↓

Lemongrass essential oil → Filtered chitosan solution ← Glycerol / coloring agents

↓

Film forming solution

↓

Heating &stirring of film forming solution at 50 °C for 10 to 15 minutes

↓

Filtration using muslin cloth if any coloring agents are added

↓

Casting of filtered solution on a petri dish / polypropylene sheet

↓

Drying under electrically operated blower for 24 h

↓

Peeling of biodegradable film

Fig. 1. Flow chart for development of biodegradable film with chitosan and lemongrass essential oil

compositions 150, 100, 100, and 300 ml as 20.153, 19.564, 16.818, and 16.230 N respectively. Puncture force indicating the ability of the film to resist against punctures.

The above graphs representing the tensile strength and stress-strain curves during puncture test for biodegradable films for samples S5 and S6. The deformation of film and rupture at peak load, Sample S5 shows highest peak load and deformation indicating, best tensile strength and deformation properties (Fig. 3, 4). The peak load at which film ruptured during puncture force, Sample S5 shows highest peak load, hence it can withstand more puncture force compared to other samples (Fig. 5, 6).

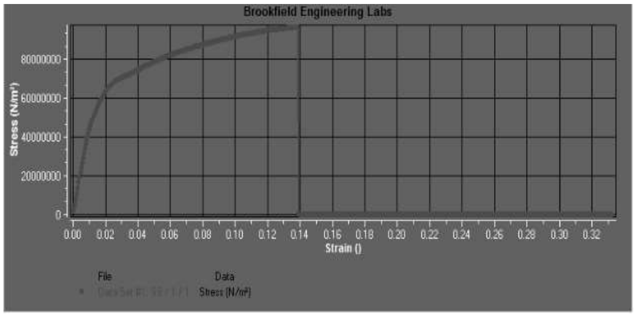

Fig. 2. Variation of tensile stress with respect to tensile strain for sample 5

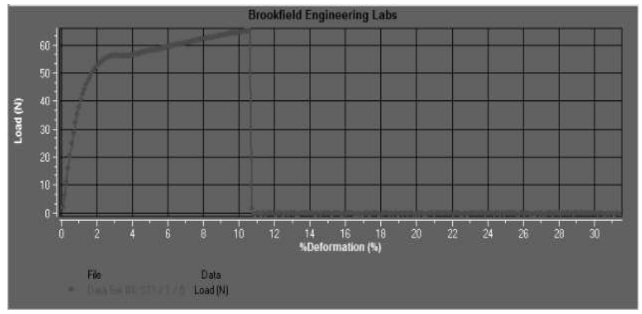

Fig. 3. Variation of tensile stress with respect to tensile strain for sample 6

Table 3. Thickness and colour of the films based on chitosan and lemongrass essential oil

Sample	Sample composition (ml)	Thickness (mm)	L	a	b
S1	100	0.05	22.86	-0.39	0.27
S2	70	0.03	21.42	-0.08	0.11
S3	50	0.02	21.37	-0.19	-0.38
S4	130	0.02	14.34	-0.51	-0.71
S5	150	0.03	20.40	-0.34	0.19
S6	300	0.05	20.53	-0.36	0.34
S7	75	0.025	18.39	-0.25	-0.50
S8	100	0.02	17.41	-0.46	-0.40
S9	100	0.04	19.38	-1.15	9.26
S10	75	0.04	14.19	-0.31	-0.53
S11	200	0.05	18.28	-0.77	1.43

Table 4. Mechanical properties of tensile strength, elasticity and puncture force of the films based on chitosan and lemongrass essential oil

Sample	Sample composition (ml)	Peak load at tensile force (N)	Deformation at peak load (mm)	Puncture force (N)
S1	100	51.583	2.93	16.818
S2	70	24.173	1.68	8.875
S3	50	16.671	1.34	4.609
S4	130	32.950	2.79	13.337
S5	150	73.256	12.45	20.153
S6	300	65.21	10.14	16.230
S7	75	12.896	1.39	13.239
S8	100	25.841	3.20	10.885
S9	100	50.72	2.82	19.564
S10	75	49.426	2.48	12.847
S11	200	46.41	4.9	11.604

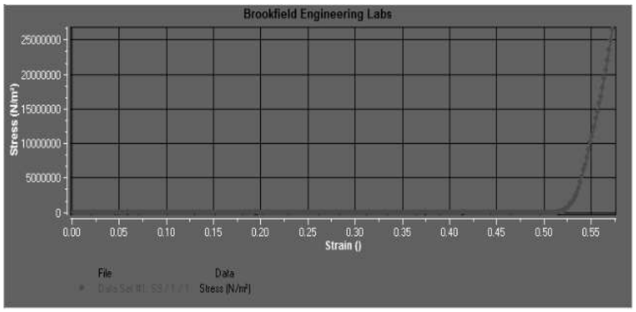

Fig. 4. Stress-strain curve for the puncture process for sample 5

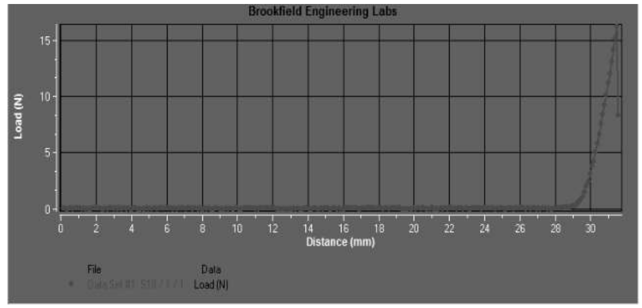

Fig. 5. Stress-strain curve for the puncture process for sample 6

Fig. 6. Biodegradability of film

Biodegradability: Biodegradable film prepared from chitosan and lemongrass essential was degraded about 0.03 g for every 10 days and was completely degraded after 4 months. Biodegradable film was kept in soil in the month of September 2022 and it was completely degraded in soil by the end of December 2022. Hence, film fabricated is biodegradable in nature and eco-friendly.

CONCLUSION

Biodegradable film from chitosan and lemongrass essential oil was developed and its physical and mechanical properties were tested. Major findings of this study were, sample with glycerol took more time for drying than usual, and the sample with coconut oil is not recommended, as the oil is separated and formed on the surface of the film. Sample with lime yellow food colour has pleasing appearance with good mechanical properties and the sample with beetroot juice has good mechanical strength but the colour of it is not durable. Best films were resulted on polypropylene petri dish and polypropylene sheets. Based on the observations and results, sample S5 is the best composition. Hence, for 150 ml of film forming solution, 500 cm^2 polypropylene sheets can be sufficient to provide required thickness and strength. It is not recommended to dry more quantity solution on a same sized mould, as it will be wasted.

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