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Development of Potato Starch Based Biodegradable Packaging Film

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ABSTRACT: This research work deals with incorporating the potato starch to make biodegradable packaging film using glycerol as a plasticizer. Starch is a very well known carbohydrate polymer, and plants from which it is obtained grow in nearly all the temperate zones. The experiment was conducted at Department of Processing and Food Engineering, College of Agricultural Engineering and Technology, Junagadh Agricultural University during 2019-20. The experiments were aimed to prepare a biodegradable plastic from renewable sources such as starch that would be environment-friendly. Optimization of preparation condition would help us to study the feasibility and potential of this starch with other additives to obtain a biodegradable as well as high tensile strength plastic. Development of starch film was carried out at different levels of starch concentration (5, 6.5, 8, 9.5 & 11) W/V % and glycerol concentration (0.5, 0.875, 1.250, 1.625 & 2) V/V % whereas distilled water 100 ml and acetic acid 1 ml were kept constant throughout the experiment. The films were prepared by casting technique using a film-forming solution. The results on biodegradable film were analyzed using Central Composite Rotatable Design (CCRD), Response Surface Methodology with two factors. The physical properties of potato starch powder *viz.*, water absorption index and water solubility index was found as 139 ± 1.53% and 82 ± 1.52%. Physico-chemical properties of potato starch biodegradable plastic *viz.*, moisture content, transparency, water absorption capacity and water vapour permeability was found as 23.1 %, 69.54%, 190% and 0.0058 g mm/m^2 kPa respectively. The response surface quadratic model for potato starch film optimized the treatment condition as 7.1 g starch concentration and 0.5 ml glycerol concentration.

Keywords: Biodegradable packaging, Potato starch, Glycerol, Biodegradable polymer, Synthetic polymer.

INTRODUCTION

Synthetic polymers are significant in numerous parts of industry, especially in the packaging industry. In any case, it affects the climate and causes issues with statement of waste. Materials utilized for food packaging today comprises of a variety of petroleum-derived plastic polymer, metals, glass, paper and board. Among these packaging materials, plastic is discovered to be the best due to its long life properties. Thus, its utilization is increasing day by day. Plastics can't be degraded by regular cycles in a short time; therefore, they are left as plastic waste causing environmental problems. Methods typically used to annihilate different kinds of waste, for example, burning and burying are not suitable for plastic destruction. In light of these issues related with plastic waste, there has been significant interest in the development and production of biodegradable plastics (Pablo *et al.*, 2007). At last, the average petroleum based plastics takes a long time to degrade because of the atomic bonds that make the plastics so solid and similarly impervious to regular cycles of biodegradation.

Plastics from natural polymers are biodegradable plastics. Biodegradable plastics will be decomposed due to bacteria, fungi or other micro-organisms that use them as food. New biodegradable biopolymers are developed using biotechnological processes. These biopolymers are termed as "green plastic", which are derived from plants. This green plastic is the topic of interest for contemporary scientists as it is ancillary of traditional chemical based plastics. The green plastic should be derived from renewable sources; it should be biodegradable in nature and eco-friendly (Tharanathan 2002).

Starch is a biodegradable polysaccharide, created in a lot at low consumption and shows thermoplasticity in nature. Starch an inexhaustible source, gives off an impression of being the best crude material of biodegradable polymer with low cost. Starch from different sources has been studied as a potential film-forming agent, including that from potato, barley, wheat, tapioca, and rice. Thus, it has become most promising alternative material to replace conventional plastics in individual market segments. In the present study, a biodegradable plastic film was produced by blending potato starch and glycerol. Films developed from starch are depicted as isotropic, odorless, colorless, non-toxic naturally degradable (Flores *et al.*, 2007).

Potato (*Solanum tuberosum* L.) is one of the world's major horticultural yields and is individual from the Solanaceae family. It has been in development since its presentation in the early piece of the seventeenth century and is considered critical as food crop. India is the second biggest maker of potato with a production of about 34.4 million MT after China which produces 69.06 million MT. India contributes about 11.26% of the all out potato production in the world. Potato starch based biodegradable packaging film is set up by the technique for casting.

Synthetic polymer materials have been broadly utilized in each field of human movement during the most recent decades. These artificial macromolecular substances are typically starting from petroleum and a large portion of the ordinary ones are viewed as non-degradable. Nonetheless, the petroleum resources are constrained and the sprouting utilization of non-biodegradable polymers has caused genuine ecological issues. In this manner, the polymer materials which are degradable and additionally biodegradable have been given increasingly more consideration.

Food is the necessity of our day to day life. Now a day's most of the food items are packed. In regular day to day existence, packaging is a significant zone where biodegradable polymers can be utilized. For increasingly characteristic items, bio-based films or biopolymers, improving the nature of numerous items is essential to fulfill the customers' requests of all the more environmentally friendly packaging. Other than their biodegradability, biopolymers have different attributes as air permeability, low temperature sealability, accessibility and low cost (Cutter, 2006; Satyanarayana *et al.*, 2009).

Biodegradable polymers are the one which fulfill all these functions without causing any threat to the environment. The belief is that biodegradable polymer materials was reduce the need for synthetic polymer production (thus reducing pollution) at a low cost, thereby producing a positive effect both environmentally and economically.

The performance evaluation of film was done by pigeon pea packed in potato starch film. The main objectives of this research were to produce biodegradable plastic films which are obtained from potato starch and to study its suitability for the food packaging. These plastic films are to be tested to ensure that these are appropriate for the food packaging.

MATERIAL AND METHODS

A. Raw materials

Potato starch, glycerol and acetic acid in analytical grade were procured. Potato starch was used for preparation of biodegradable plastic film from starch. Glycerol was used as a plasticizer in the filmogenic solution to increase the flexibility and plasticity of the film. Acetic Acid was also added to the solution. In packaging films, acetic acid is added to increase the antimicrobial, plasticizing and dispersing effect in biodegradable/edible films and to improve the mechanical properties and water vapor permeability. Distilled water was added to solution t as it goes about as the plasticizer and it diminishes the weakness of plastic films. So water is used to make the solution of starch.

B. Preparation of film

The films were prepared by casting technique using a film-forming solution containing potato, corn and rice starch individually. Starch Concentration (5, 6.5, 8, 9.5 & 11 % (W/V)) and Glycerol Concentration (0.5, 0.875, 1.25. 1.625 & 2 % (V/V)) was taken as variable parameter. 100 ml distilled water was added to it. The mixture of dry starch, water and glycerol was taken in a beaker. Then 1 ml of acetic acid was added to the solution. The mixture was mixed with the help of glass rod on heating with stirring on magnetic stirrer at 40˚C for 5 minutes. Now the mixture was kept in water bath at 85˚C temperature for 15 minutes and continuously agitated by glass road. Now a cast was prepared and the entire solution was poured on the cast and was left for drying at room temp for 24 hrs. After drying the films were peeled off and were kept in polythene bags away from moisture.

C. Thickness of the Film

Thickness of the Film was measured with the help of digital Vernier Calipers (Mitutoyo Corporation, Japan made, model- CD-12"), having a least count of 0.01mm.

D. Physico-chemical properties of developed biodegradable packaging film

The physico-chemical properties viz., moisture content, transparency, water absorption capacity and water vapour permeability OF developed biodegradable packaging film were determined by AOAC (2005), Han and Floros (1997), ASTM D570 and ASTM E96-95 (1995) method respectively and surface morphology investigations were performed on thermoplastic starch films of potato starch by using SEM machine model (HITACHI S-3400N).

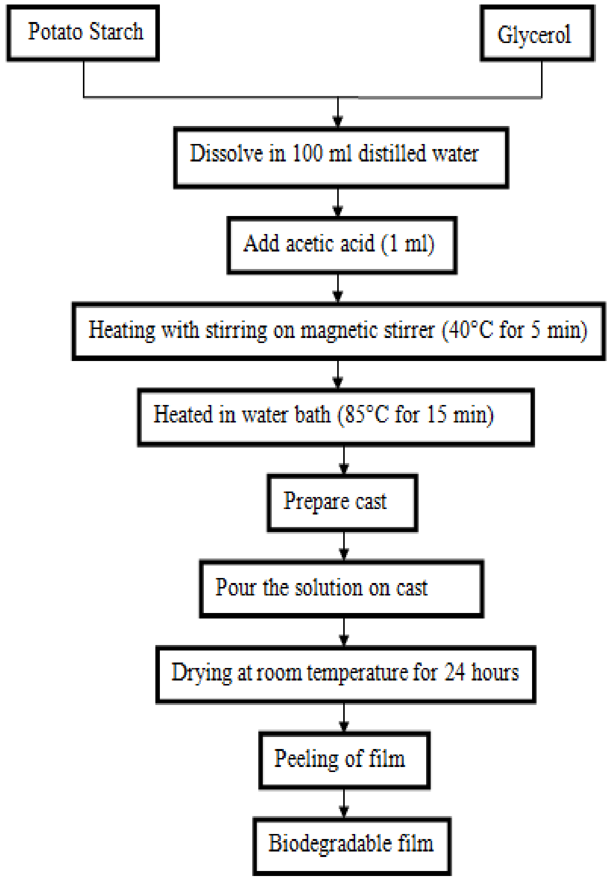


Fig. 1. Process flow chart for formation of starch based biodegradable film.

Table 1: Treatment combinations.

Run	Starch Concentration, % (W/V)	Glycerol Concentration, % (V/V)
1	8.0	1.250
2	8.0	1.250
3	8.0	1.250
4	9.5	1.625
5	5.0	1.250
6	6.5	0.875
7	9.5	0.875
8	8.0	1.250
9	11.0	1.250
10	8.0	1.250
11	8.0	1.250
12	8.0	2.000
13	6.5	1.625
14	8.0	0.500

E. Experimental design

The experiment was conducted by Response Surface Methodology (RSM), it is an empirical statistical modelling technique employed for multiple regression analysis using quantitative data obtained from properly designed experiments. The Central Composite Rotatable Design (CCRD) was used for designing the experiments using Design Expert 10 Software (Khuri and Cornell, 1987).

RESULTS AND DISCUSSION

A. Physico-chemical properties of potato starch biodegradable packaging film

Thickness of the film. Thickness of all the samples was measured and the mean thickness was calculated with the use of Vernier Calipers and showed that the mean thickness was in the range of 0.11 mm to 0.16mm. The result in agreement with Thakur *et al.*, (2017) reported that increasing the starch in the solution, the films were thicker.

Table 2: Thickness of potato starch film.

Run	Thickness (mm)
1	0.14
2	0.15
3	0.12
4	0.14
5	0.11
6	0.16
7	0.16
8	0.12
9	0.14
10	0.15
11	0.14
12	0.12
13	0.15
14	0.15
Mean	**0.14**

B. Physico-chemical properties of potato starch biodegradable packaging film

The different physico-chemical properties of potato starch biodegradable plastic were analysed and studied viz., moisture content, transparency, water absorption capacity, water vapour permeability and surface morphology were carried out as per methods and results were tabulated in Table.3.

Effect of starch and glycerol concentration on moisture content of potato starch based biodegradable film. Moisture content of potato starch film ranged between 18.19 % to 23.1%. The maximum moisture content was observed for the combination 11 g of starch concentration and 1.25 ml glycerol concentration and minimum moisture content was found for the combination of 5 g starch concentration and 1.25 ml glycerol concentration. The effect of starch and glycerol concentration on moisture content of potato starch biodegradable film is presented in Table 3.

The response surface curve of variation in the moisture content of potato starch biodegradable plastic as a function of starch concentration (X_1) and glycerol concentration (X_2) is shown in Fig 2(a). The contour plot for moisture content of potato starch biodegradable plastic film as a function of starch concentration (X_1) and glycerol concentration (X_2) is presented in the Fig. 2(b) which indicated the increase in moisture content as the starch concentration and glycerol concentration was increased up to maximum level. At this combination, moisture content of potato starch biodegradable plastic film was predicted 24.55 %.

The result in agreement with the results reported by Talja *et al*. (2008). Similar results were reported by Buso- Rios *et al.*, (2020) in purple sweet potato starch film and Kibar and Us (2013) in corn starch film.

The regression analysis and ANOVA results for the moisture content of potato starch plastic are shown in Table 4. It can be seen from the table, that starch and glycerol concentration showed positive linear effect which significant at $p<0.001$.Whilst, the interaction effect of starch concentration and glycerol concentration was positively non-significant and the quadratic effect of starch concentration was positively non- significant and quadratic effect of glycerol concentration was negatively non-significant on moisture content.

Table 3. Different physico-chemical properties of potato starch biodegradable packaging film.

Std. run	Starch concentration (%)	Glycerol concentration (%)	Moisture content (%)	Transparency (%)	Water absorption capacity (%)	Water vapour permeability (g mm/m^2 day KPa)	Tensile strength (MPa)	Puncture strength (MPa)
1	6.5	0.88	19.22	67.31	156	0.004	7.01	6.35
2	9.5	0.88	21.4	59.63	178	0.0052	11.76	8.97
3	6.5	1.63	19.72	66.25	161	0.00411	6.23	5.31
4	9.5	1.63	22.32	58.23	183	0.00524	11.07	8.36
5	55	1.25	18.19	69.54	151	0.00371	5.09	4.28
6	11	1.25	23.1	52.86	190	0.0058	13.62	10.45
7	8	0.50	18.73	63.21	165	0.00435	10.58	7.83
8	8	2.00	21.22	60.9	176	0.00473	8.54	6.95
9	8	1.25	20.23	64.23	173	0.00462	10.36	7.47
10	8	1.25	20.43	62.31	167	0.00428	10.47	7.36
11	8	1.25	20.46	62.45	172	0.00458	9.85	7.28
12	8	1.25	19.93	62.85	171	0.00459	10.2	7.08
13	8	1.25	19.95	63.10	168	0.00447	9.50	7.39
14	8	1.25	19.96	64.76	169	0.00465	9.78	7.53

Table 4: Analysis of variance (ANOVA) table and regression coefficients for response surface quadratic model of different physico-chemical properties of potato starch biodegradable packaging film.

Source	Moisture content	Transparency	Water absorption capacity	Water vapor permeability	Tensile strength	Puncture strength
Intercept	+20.16	+63.38	+169.09	+4.543E-003	+9.86	+7.32
Linear terms						
$A(X_1)$	+1.22***	-4.09***	+10.17***	5.425E-004***	+2.22***	+1.50***
$B(X_2)$	+0.53***	-0.59*	+2.67**	+7.583E-005	-0.46*	-0.28***
Interaction terms						
$AB(X_1 X_2)$	+0.10	-0.085	+2.82878E-014	-1.750E-005	+0.023	+0.11
Quadratic terms						
$A^2(X_1^2)$	+0.13	-0.51**	+0.10	+5.724E-005*	-0.19	-1.103E-004
$B^2(X_2^2)$	-0.034	-0.30	+0.10	+3.493E-006	-0.14	+6.140E-003
Indicators for model fitting						
R^2	0.9552	0.9744	0.9751	0.968	0.9628	0.9892
Adj-R^2	0.9272	0.9585	0.9595	0.9489	0.9395	0.9825
Pred-R^2	0.7391	0.9470	0.9316	0.9054	0.7923	0.9346
Adeq Precision	20.81	29.97	30.16	27.11	24.61	46.92
F-value	34.13	61.01	62.55	49.29	41.36	146.90
Lack of fit	NS	NS	NS	NS	NS	NS
C.V. %	1.76	1.33	1.21	2.66	5.76	2.67

A or X_1 = Starch Concentration, B or X_2 = Glycerol Concentration, ***Significant at $p<0.001$, **Significant at $p<0.01$,*Significant at $p<0.05$,NS=Non-significant

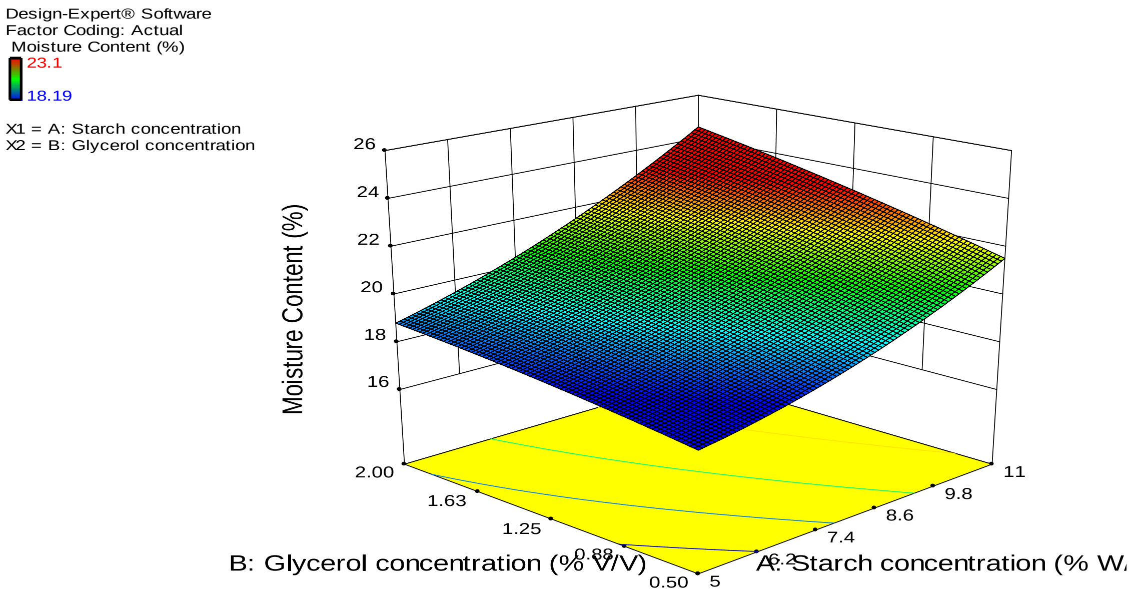


Fig. 2 (a) Response surface plot for moisture content of potato starch biodegradable film.

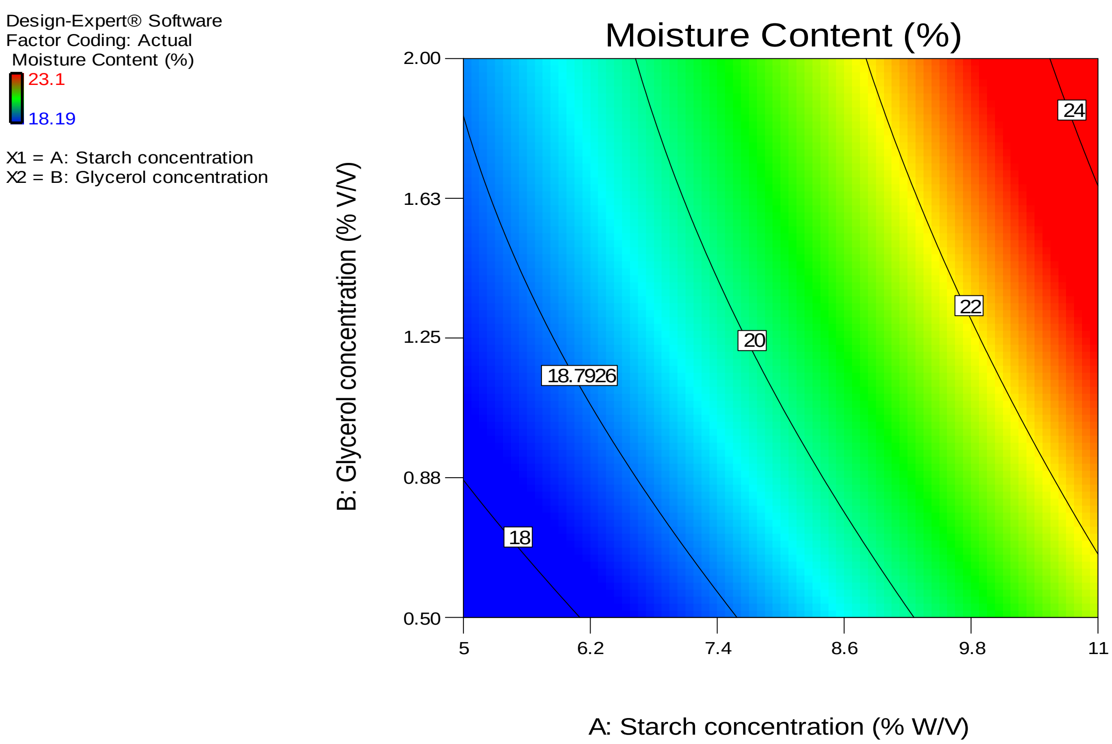


Fig. 2 (b) Contour plot for moisture content of potato starch biodegradable film.

Effect of starch and glycerol concentration on transparency of potato starch based biodegradable film. Transparency of potato starch film was ranged between 52.86 % to 69.54 %. The maximum transparency was observed for the combination of 5g starch concentration and 1.10 ml glycerol concentration and minimum transparency was found for the combination of 11g starch concentration and 1.25 ml glycerol concentration. The effect of starch and glycerol concentration on transparency of potato starch biodegradable film is presented in Table 3.

The response surface curve of variation in the transparency of potato starch biodegradable plastic film as a function of starch concentration (X_1) and glycerol concentration (X_2) is shown in Fig 3(a). It represents the interactive effect of starch concentration and glycerol concentration on the, transparency of potato starch film. The contour plot for transparency of potato starch biodegradable plastic film as a function starch concentration (X_1) and glycerol concentration (X_2) is presented in Fig. 3 (b) which indicated the increase in transparency as the starch concentration was decreased up to minimum level and transparency was increased with an increase in glycerol concentration up to 1.10 ml; with further increase in glycerol concentration transparency was decreased. Transparency at the combination of 5g starch concentration and 1.02 ml glycerol concentration may be observed 69.65 %. The result in agreement with the result reported by Dai *et al*., (2010). Similar result was found by Khairunnisa *et al*., (2018) in the film made from alginate.

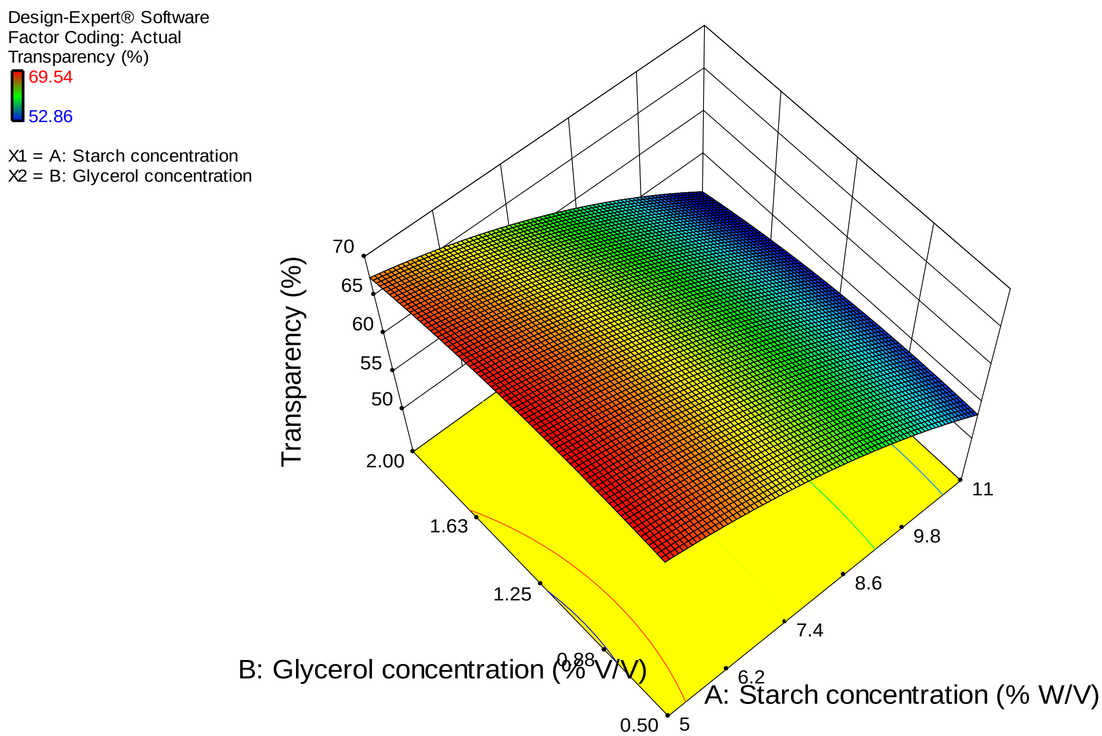


Fig. 3 (a) Response surface plot for transparency of potato starch biodegradable film.

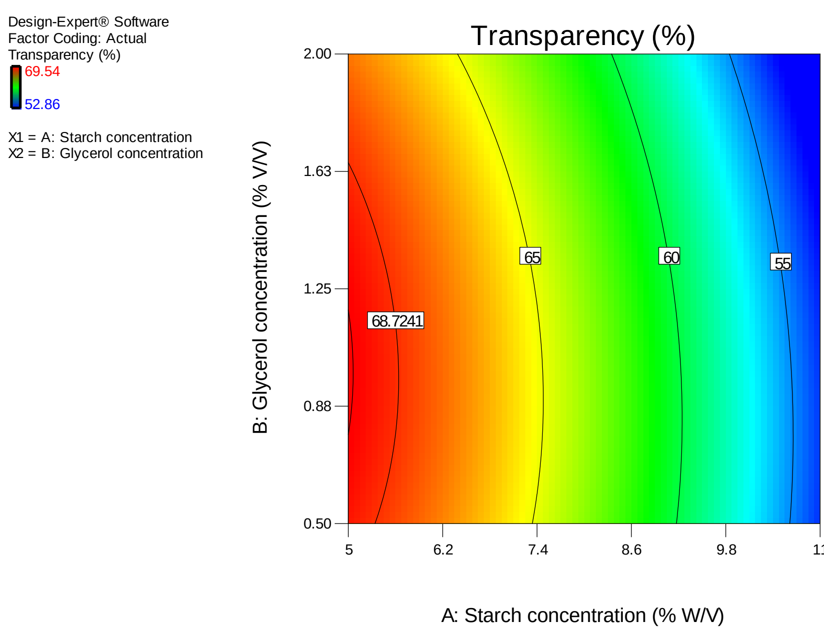


Fig. 3 (b) Contour plot for transparency of potato starch biodegradable film.

The regression analysis and ANOVA results for the transparency of potato starch film are shown in the Table 4. It can be seen from the table, that starch concentration showed negative linear effect on transparency which there were significant value at $p<0.001$ also glycerol concentration showed negative linear effect on transparency which was significant at $p<0.05$. Whilst, the interaction effect of starch concentration and glycerol concentration was negatively non-significant and the quadratic effect of starch concentration showed negatively significant at $p<0.01$ and glycerol concentration was negatively non-significant on transparency.

Effect of starch and glycerol concentration on water absorption capacity of potato starch based biodegradable film. Water absorption capacity of potato starch film was found between 151 to 190%. The maximum water absorption capacity was observed for the combination of 11g starch concentration and 1.25 ml glycerol concentration and minimum water absorption capacity content was found for the combination of 5g starch concentration and 1.25 ml glycerol concentration. The effect of starch and glycerol concentration on water absorption capacity of potato starch biodegradable film is presented in Table 3.

The response surface curve of variation in the water absorption capacity of potato starch biodegradable plastic film as a function of starch concentration (X_1) and glycerol concentration (X_2) is shown in Fig 4 (a). It represents the interactive effect of starch concentration and glycerol concentration on the, water absorption capacity of potato starch film. The contour plot for water absorption capacity of potato starch biodegradable plastic film as a function of starch concentration (X_1) and glycerol concentration (X_2) is presented in the Fig. 4(b) which indicated the increase in water absorption capacity as the starch and glycerol concentration was increased up to maximum level. At this combination, the water absorption capacity of potato starch film was predicted up to 145 %. The result in agreement with result reported by Farahnaky *et al.* (2013) in wheat starch film and similar result was found by Bourtoom and Chinnan (2008a) in rice starch-chitosan film.

The regression analysis and ANOVA results for the water absorption capacity of potato starch film are shown in the Table 4. It can be seen from the table, that starch concentration showed positive linear effect on water absorption capacity which significant at $p<0.001$ also glycerol concentration showed positive linear effect on water absorption capacity which was significant at $p<0.01$. Whilst, the interaction effect of starch concentration and glycerol concentration was positively non-significant and the quadratic effect of starch concentration and glycerol concentration was positively non-significant on water absorption capacity.

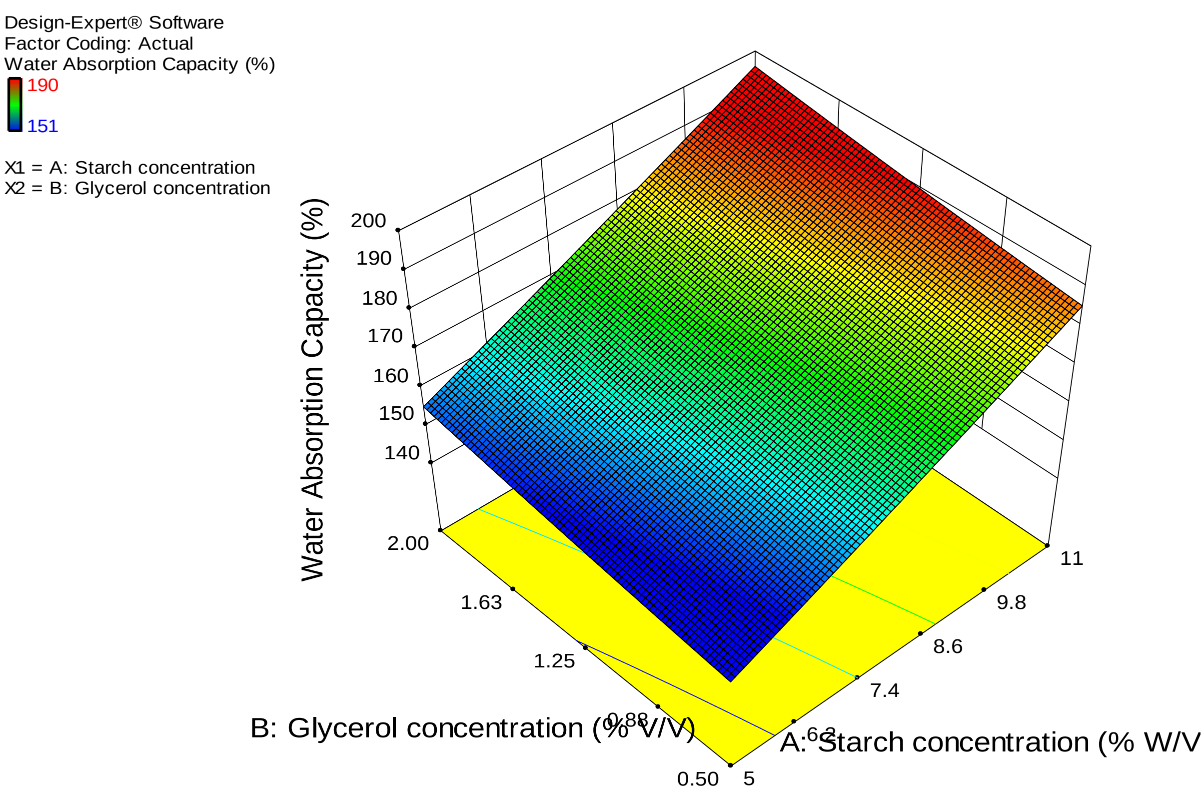


Fig. 4 (a) Response surface plot for water absorption capacity of potato starch biodegradable film.

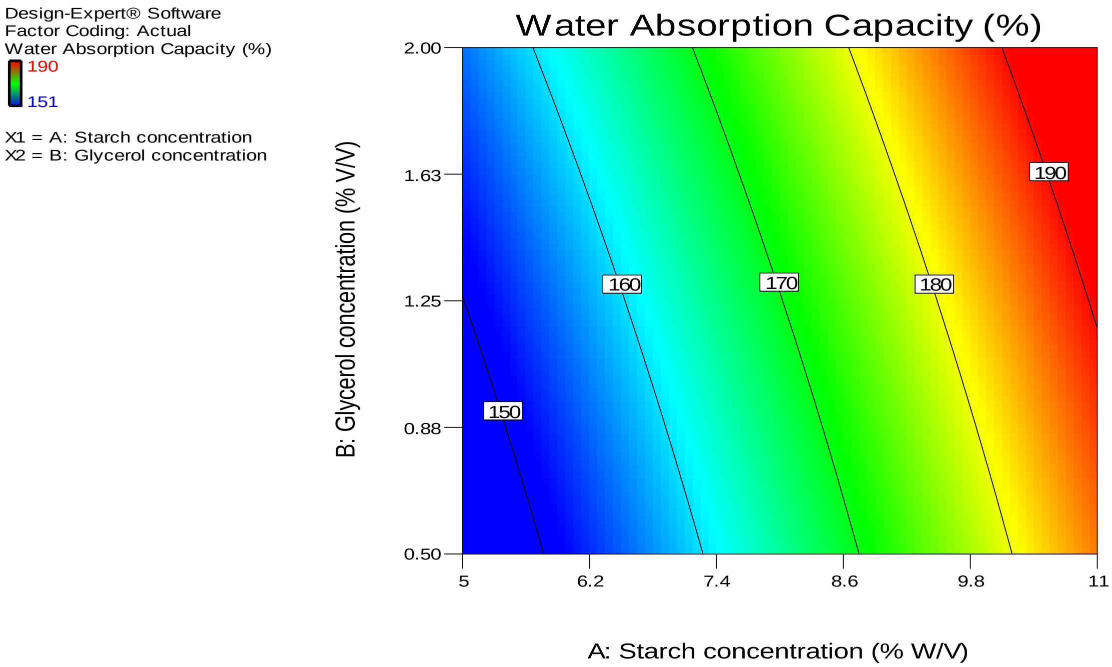


Fig. 4 (b) Contour plot for water absorption capacity of potato starch biodegradable film.

Effect of starch and glycerol concentration on water vapor permeability of potato starch based biodegradable film. Water vapor permeability of potato starch film ranged from 0.00371 to 0.00580 g.mm/m^2kPa. The maximum water vapor permeability was observed for the combination of 11g starch concentration and 1.25 ml glycerol concentration and minimum water vapor permeability was found for the combination of 5g starch concentration and 1.25 ml glycerol concentration. The effect of starch and glycerol concentration on water vapour permeability of potato starch biodegradable film is presented in Table 3.

The response surface curve of variation in the water vapor permeability of potato starch biodegradable plastic film as a function of starch concentration (X_1) and glycerol concentration (X_2) is shown in Fig 5 (a).

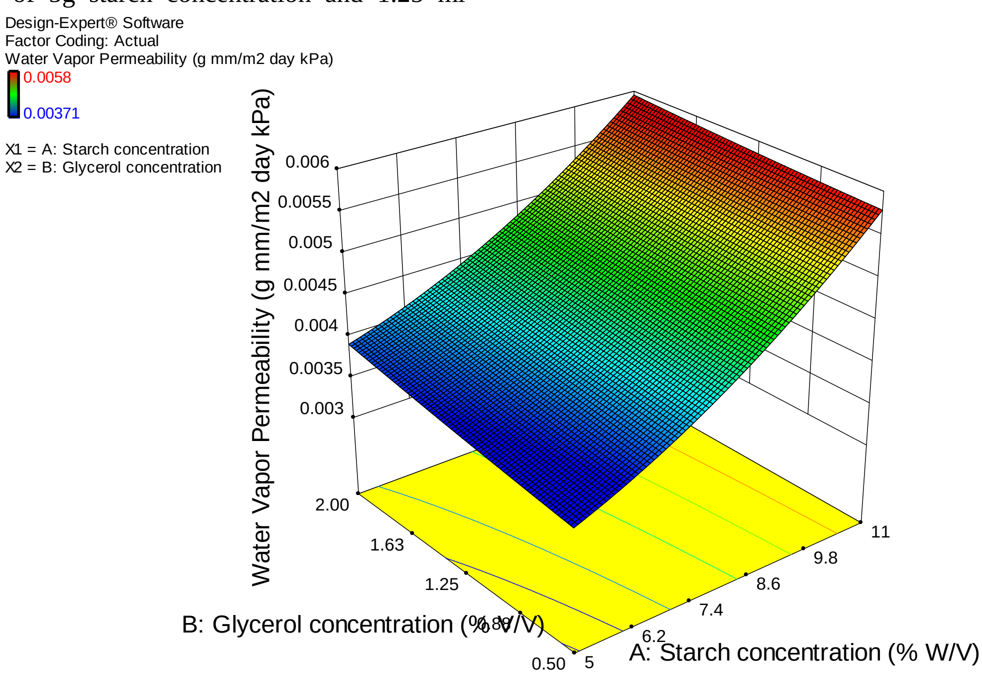


Fig. 5 (a) Response surface plot for water vapor permeability of potato starch biodegradable film.

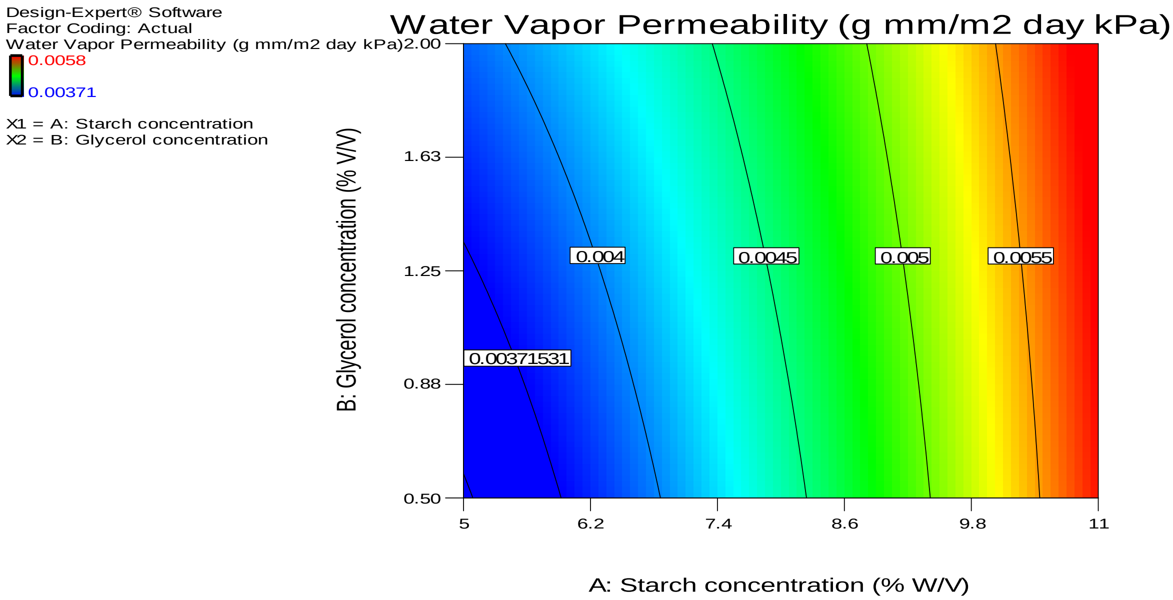


Fig.5 (b) Contour plot for water vapor permeability of potato starch biodegradable film.

It represents the interactive effect of starch concentration and glycerol concentration on the, water vapor permeability of potato starch film. The contour plot for water vapor permeability of potato starch biodegradable plastic film as a function of starch concentration (X_1) and glycerol concentration (X_2) is presented in the Fig. 5 (b) which indicated that decreases the starch and glycerol concentration, water vapour permeability was decreased up to minimum level.

At this combination, the water vapour permeability of potato starch film was predicted up to 0.00348 g.mm/m^2 kPa. The result was agreement with the result reported by Ghasemlou *et al.,* (2013) in corn starch films incorporated with plant essential oils. Similar result was found by Farahnaky *et al.*, (2013) in films made of wheat starch and glycerol and Muhammed *et al.,* (2015) in sugar palm starch film incorporated with glycerol and sorbitol.

The regression analysis and ANOVA results for the water vapor permeability of potato starch film are shown in the Table 4. It can be seen from the table, that starch concentration showed positive linear effect on water vapor permeability which significant at $p<0.001$; glycerol concentration showed positive linear effect on water vapor permeability which was non-significant. Whilst, the interaction effect of starch concentration and glycerol concentration was negatively non-significant and the quadratic effect of starch concentration was positively significant at $p<0.05$ and glycerol concentration was positively non-significant on water vapor permeability.

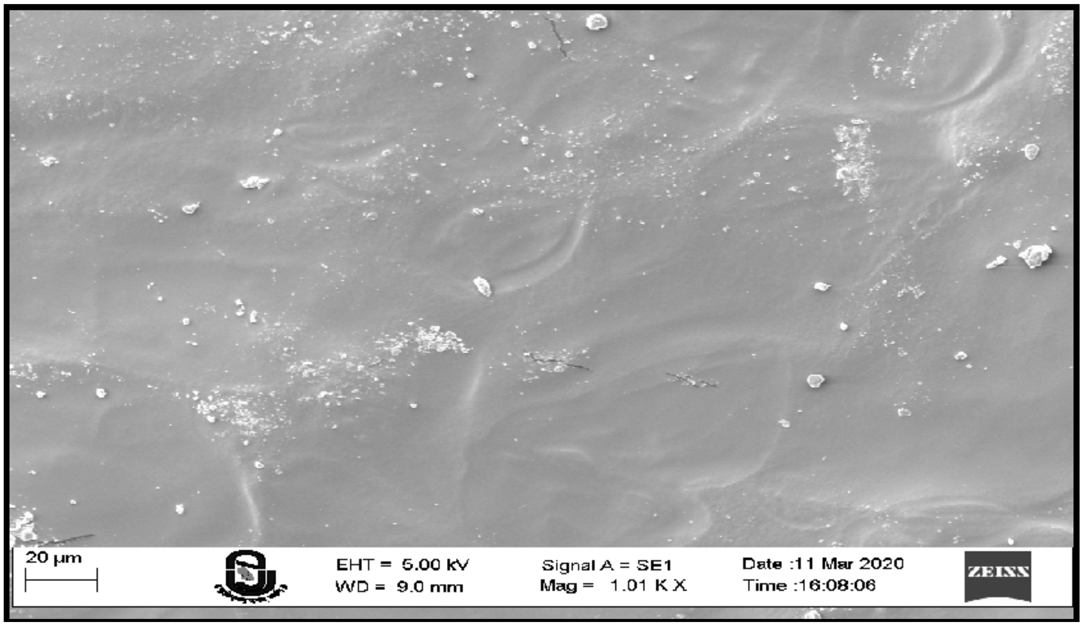
Surface morphology of developed potato starch packaging film. The surface morphology of potato starch/glycerol films was studied with SEM is represented in Fig. 6. In order to have the leading effect, the samples were gold plated and the scanning was synchronized with microscopic beam in order to keep up the little size over a huge distance comparative with the specimen. The subsequent images had a great depth of the field. A remarkable three dimensional appearance with high resolution was gotten in case of cross linked matrix.

The potato starch films had several unequalized holes, suggesting that the miscibility and compatibility in each component in potato starch films were increased. With increase in glycerol content the surface of the film gets smoother.

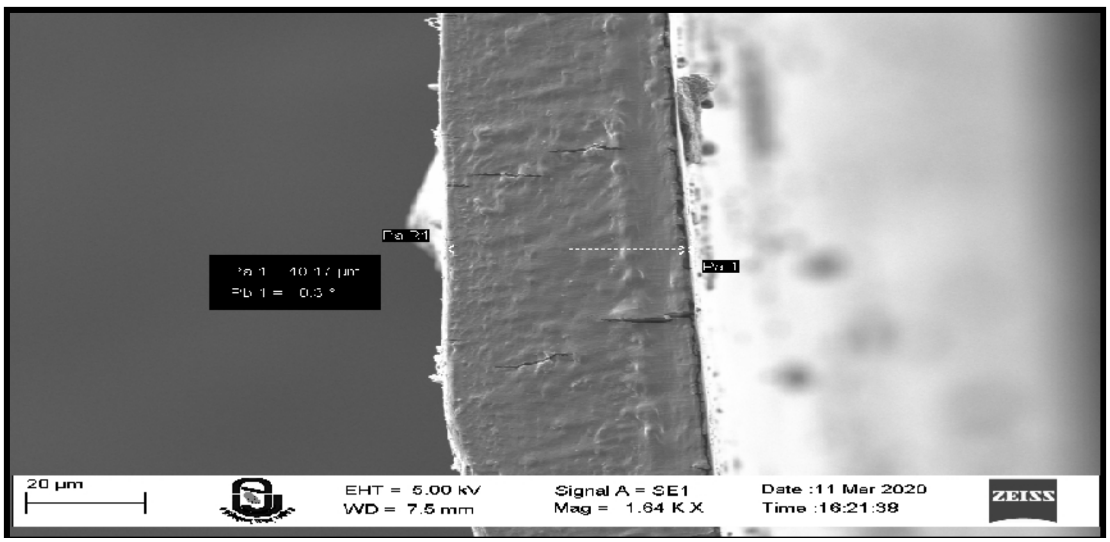
From the SEM micrograph, it can be concluded that the potato starch granules dispersed well in the glycerol. This dispersion assists with improving the mechanical properties of the film and shows a concurrence with the tensile property results.

Optimization and validation of process variables

The optimum condition for the development of starch based biodegradable film was determined by the numerical optimization technique, using Design Expert software version 10 (State-Ease Inc., Minneapolis, MN, USA). The optimum treatment conditions for potato starch film were found to be, 7.1 g starch concentration and 0.5 ml glycerol concentration. The analysis showed that at this combination of starch and glycerol concentration, it would be possible to produce a potato starch based biodegradable plastic film with a moisture content of 18.52 %, transparency 65.47%, water absorption capacity 159 %, water vapour permeability 0.004 g mm/m^2 day kPa.



(A) Surface or top view



B) Side view

Fig. 6. Scanning electron microscopic images of cross-section of potato starch film.

C. Performance evaluation of potato starch biodegradable film for packaging

Packaging is any material that is used in holding, protection, handling, delivery and presentation of goods, from raw materials to finished products providing from producers to consumers. The ability of developed biodegradable film to seal at sides and corners to form plastic carry bag was evaluated. The results indicated that the prepared bioplastic samples have good sealing properties. The heat-sealing feature was estimated through visual inspection. The sample was inspected manually. The sealed sample seems to have excellent sealing properties. Since sealing properties are important for preparing plastic bags, hence, it is concluded that the bioplastics produced in this study can be used to manufacture bioplastic carry bags. A sample bag produced from the starch is used for packaging is shown in Plate 1.

Plate 1. Pigeon pea packed in potato starch film.

CONCLUSION

Biodegradable plastics have been successfully produced from potatoes using the aforementioned approach and experimental procedures. The plastic samples produced were characterized in order to determine their physical and physico-chemical properties. Based on the results obtained, it could be concluded that the potatoes which are available in large quantity or it is a good source of starch for the production of biodegradable plastics. Their renewability and minimization of environmental pollution/hazards are of great achievement. The moisture content of the film was increase with increased the starch and glycerol content. This behavior could be explained because increasing the starch content promotes the formation of more hydrogen bridges and plastic containing glycerol absorb more moisture which is likely due to the hydrophilic nature of glycerol. Transparency of the film was decreases with increase in the starch concentration and glycerol concentration due to more molecules of starch give darker colour of the film. Water absorption capacity of the film increased with the starch and glycerol content due to starch and glycerol being hydrophilic in nature. Water vapour permeability increases with increase in the glycerol content due to hydrophilic nature of biopolymers and presence of voids in their structure have a considerable influence on the WVP of resulting films. This data can be used to design specific food packaging film system.

FUTURISTIC APPROACH

Focused research is needed in bringing more values such as making the packaging material simpler yet smarter, where consumer is able to assess the quality, safety, shelf-life, and nutritional values of the contents of packet with cost effectiveness. The benefits however should not come at the cost of the cost of curing environmental issues and should eco-friendly.

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