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Chitin Based Bioplastics Production and Insilco Studies using Optimization software

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Abstract
The aim of this project is to study the different properties of produced bioplastics and the relationship between several factors and responses. Corn starch, potato starch and a combination of both were used as the source for production, this resulted in good bioplastics which were biodegradable in nature and had better properties. Design Expert Software and Minitab software were utilized to employ Insilco studies involving interpretation, comparison, characterization, optimize, etc. with the help of several tables and graphs. Optimized values for the factors using Design Expert Software are: Chitosan = 0.527, Starch=3.89 and Glycerol=1.72. The optimum values using Minitab Software were found to be 0.4 for Chitosan, 3.0 for Starch and 1.6 for Glycerol.
Keywords: *Bioplastics, Corn Starch, Potato Starch, Optimization, Design Expert Software, Minitab*

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I. INTRODUCTION

Plastics are regularly used extensively in every field. It is made from non-renewable sources. Each year, over 300 million tons of plastics are produced. More than 50% of this amount is said to be single purpose use. The production process gives toxic by-products like carbon dioxide into the environment [7]. It is robust, lightweight, cheap, has longevity, etc. Though it's given advantages, it is not a healthy practice due to its non-biodegradable properties leaving the environment to be contaminated, it can take up to thousands of years to decompose. Plastics are a key cause of pollution, resulting in islands of garbage dumps on land. Plastics cause health hazards in humans and is also one of the leading reasons for the cause of death of various marine species. Ever-growing industries such as Pharmaceutical, Food, Transportation, Aerospace, Electronics and more have new and exponential demands every year [9]. For this reason, an immediate cost-effective, efficient and easily available alternative has to be produced. The proposed alternative is called Bioplastics. Bioplastics are made from renewable natural sources like recycled food waste, polysaccharides, etc. By definition, bioplastics are capable of being broken down by the environment [17]. The tensile strength of bioplastics is suitable for production of disposable packing material. In the production, glycerol is added as a plasticizer to enhance the flexibility and reduce brittleness [11]. Bioplastics are proposed to be used for applications such as packaging, pots, straws, paper coatings.

Approximately 50% of bioplastics are produced from Starch [16]. Starch is the most common carbohydrate in human diet, present in foods such as potato, rice, maize. It contains two molecules- amylose present in the linear chain and amylopectin which are branched. Mechanical factors like tensile strength, water absorption and biodegradability were tested for these bioplastics. It was found by the results that using this filler along with potato starch increased the bioplastic performance. Corn starch, sometimes also referred to as maize starch is derived from the endosperm of a kernel of maize. It has several characteristic features such as versatility, easy modifications, thickening the solution and medicinal properties. It is also reported that presence of corn in a bioplastic production has increased biodegradability [7]. Potato starch contains minimal fat or protein. It is a clear white powder, easily available, has neutral taste, high binding strength, low gelatinization temperature and minute tendency to make the solution yellow in colour or produce foam. These properties make it useful in the food industry. One disadvantage which pulls back the advancement is high hygroscopicity and high sensitivity to water or moisture [8,18].

Corn starch and chitosan microparticles are observed to be crosslinked with glutaraldehyde [19]. The polysaccharide composition affects the thermal stability of the bioplastic. Starch granules are both amorphous and crystalline in nature, mostly composed of amylose, which binds to another amylose molecule by (1→4) bonds, and amylopectin which binds to another amylopectin molecule by (1→4) bonds as well as branching at (1→6) bonds at intervals of approximately 20 units. Chitin, a derivative of glucose is a long polymer chain of N-acetyl glucosamine. These are the second most abundant polysaccharide found in nature after cellulose, found primarily in cell walls of fungi and the exoskeleton of arthropods for example, crustaceans and insects, making it easily available. Hence, chitin is used as an additive in bioplastic production.

Response Surface Methodology is a methodology which uses statistical models to give details about the relationship between several variables and one or multiple responses. RSM uses artificial neural networks which apply mathematical concepts to the model. This technology is widely used for its accuracy and obtaining the most efficient results. The main purpose of RSM is to study optimization, to obtain topmost quality production [11,12,14]. Optimization is a technical process which involves the search for the most optimum conditions for an effective and desirable outcome. Optimum characteristics are fine-tuned by one's requirement adjusting the additives. It can use explanatory variables as well as response variables and features such as Rotatability, Uniformity and Orthogonality. Central Composite Design is used to build a second order quadratic model which is used for the response variable.

Design Expert is a software used to perform design of experiments, comparison studies, study the characteristics, optimization, interpretation and more [12]. It can screen up to 50 factors. It provides tools to lay out an experiment on your process, mixture or combination of factors and components. Here, the design expert version used is design expert 13. The interpretations are done using ANOVA (analysis of variance). This feature of interpreting optimized data can be used to analyze and obtain the optimum parameter(s) and assess the significance and validity of the models [13]. It is based on the law of variance. ANOVA result is independent of constant bias, errors made during scaling and units. There are no prerequisites to learn Design Expert and can be easily used by beginners. Minitab is a software which was developed at Pennsylvania State University. It allows users to analyze data and interpret results with the help of several calculations and graphs. Minitab allows us to input statistical 12 data, manipulate the data and identify its trends and finally extrapolate the answers to the problems at hand in a simple and effective way. It provides a quick and effective solution to problems. There are no prerequisites to learn Minitab and can be easily used by beginners, but for specific tasks, we need prior training. It is used for experiments and projects which use several factors to analyze and interpret results. It also gives details about characteristics of the factors, and mostly used for optimization. Minitab 20.3.0.0 version is used.

1.1 Production of Bioplastics

1.1.1 Production using Corn Starch: 0.4g of chitosan and 1.6ml of glycerol were added to a beaker and mixed to obtain a paste. Further, 3g of corn starch powder and 40ml of 1% acetic acid were added to obtain a mixture. The mixture is stirred and 10ml of distilled water is added. The mixture is heated in a microwave for 5 seconds, stirred and heated again. This mixture is immediately poured onto silicone sheets with boundaries. Air bubbles are removed using a safety pin. The film is dried for 24-48 hours, peeled and observed.

1.1.2 Production using Potato Starch: The same methodology as stated for Corn Starch is adapted, using Potato Starch instead of Corn Starch.

1.1.3 Production using Corn and Potato Starch: 3g of potato starch and 1g of corn starch (co-biopolymer was used) to get a improve the previously obtained potato starch film. • Here again, the same procedure was followed, by only changing the compositions of all the chemicals according the 4g of starch which was used here.

1.1.4 Coating of bioplastic on thick sheet: 1.5% chitosan and 0.4% acetic acid were mixed and heated till it dissolved completely. To this solution, 4% corn starch and 2% glycerol were added and mixed and heated again till it became a homogenous mixture. This mixture was stored for 1 day to cool it down and a jelly like bioplastic paste was obtained. This mixture was used to coat the craft papers by using a special type of spring roller rod which spreads the mixture evenly onto the paper uniformly. The paper was secured and held tight and after applying some of the bioplastic mixture on the paper, the rod was pulled over it with uniform pressure being applied. This coated paper was then dried in the oven for 5 mins and its properties were observed. Higher the GSM, better is the coating and overall properties.

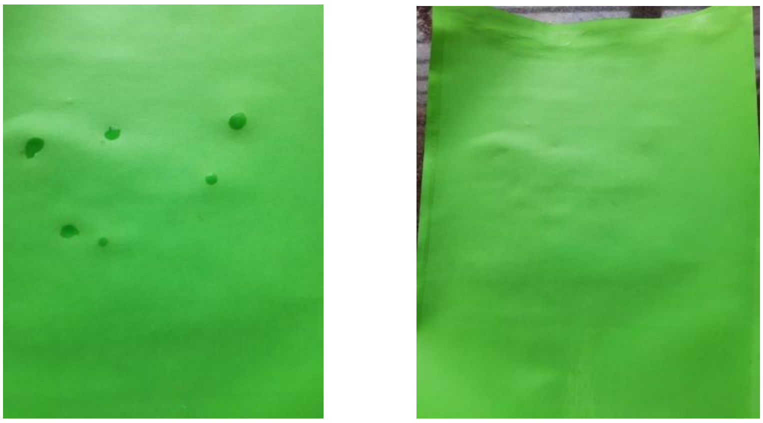


Figure 1: (A) Water droplets are dropped on the sheet coated with the bioplastic coating. (B) The Water droplet is evaporated from the surface at room temperature without seeping through the paper.

1.2 Tests Performed

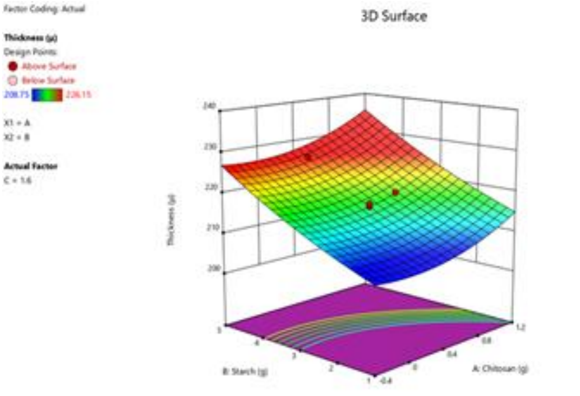
i. Hydrophobicity test: Water droplets were added onto the paper to observe whether or not the biofilm helps it and makes the water stay on the film, without seeping through the paper.
ii. Biodegradability test: Bioplastics are buried in the soil. They are checked time to time and noted if biodegraded and when.
iii. Solubility test: The bioplastic was dipped in water for 1 hour and results are observed and noted.
iv. Thermostability test: The bioplastic is kept in fridge for a couple of days and changes are observed.

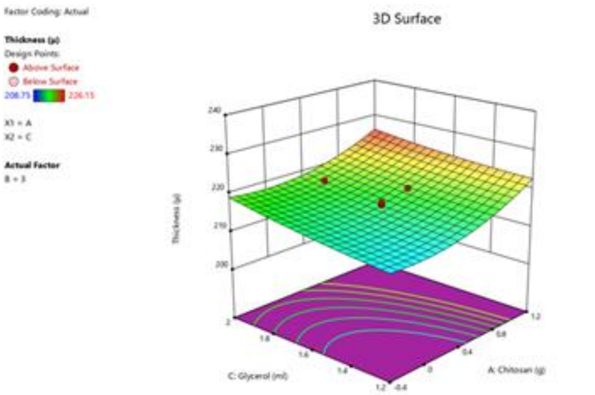
1.3 RSM using Design Expert Software

A new Central Composite Design (CCD) is selected under Response Surface. The number of factors along with their units and ranges which are from high to low are entered and so are the corresponding responses and their units. The total number of Runs is 20 runs. Here, the factors taken were Chitosan, Starch and Glycerol and the responses were Thickness, Elongation, Young's modulus and Water contact angle. The resulting table is sorted in ascending order with respect to the standard order. In this experiment, the values of the responses were entered using the previous values from CIPET (Central Institute of Petrochemicals Engineering & Technology, Bangalore). Check the information on the navigation panel to interpret the various results.

The first set of results obtained was the summary regarding the information about the factors and the responses. Graph columns like scatter plots, box plots and histograms could be observed and evaluations can be observed. Analysis for the 4 responses was then calculated (no transformations were required) and the results were interpreted. ANOVA for the different models were analyzed and their fit summaries and statistics were observed where the best suggested models were chosen. Final equations for the responses in the terms of actual and coded factors were obtained. Several model graphs were obtained such as normal plots, perturbation graphs, residual vs predicted and runs, Predicted vs actual, Box-Cox, Cook's distance plots, etc. Contour plots and 3D surface plots for different combinations of the factors can be observed for the 4 responses and the predictions were acquired.

Numerical Optimization is performed and the results of the constraints were obtained in the form of a report and graphs. 100 solutions were found here.





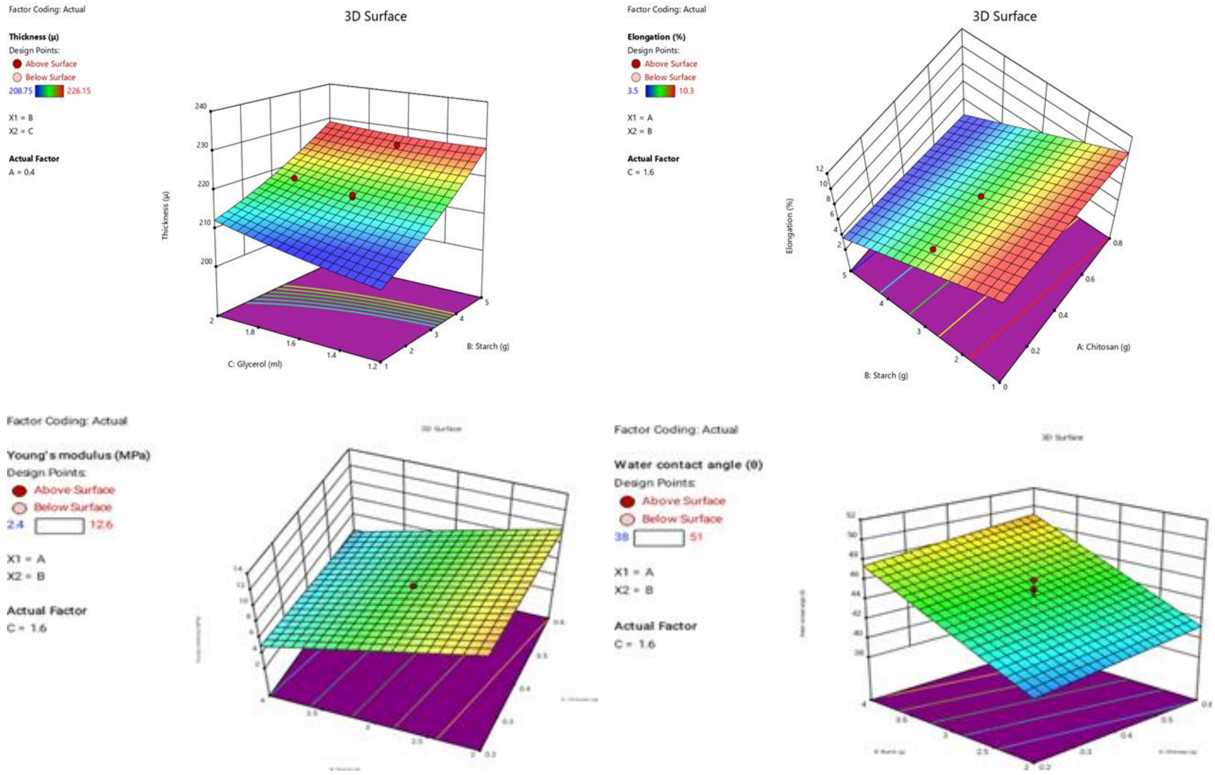
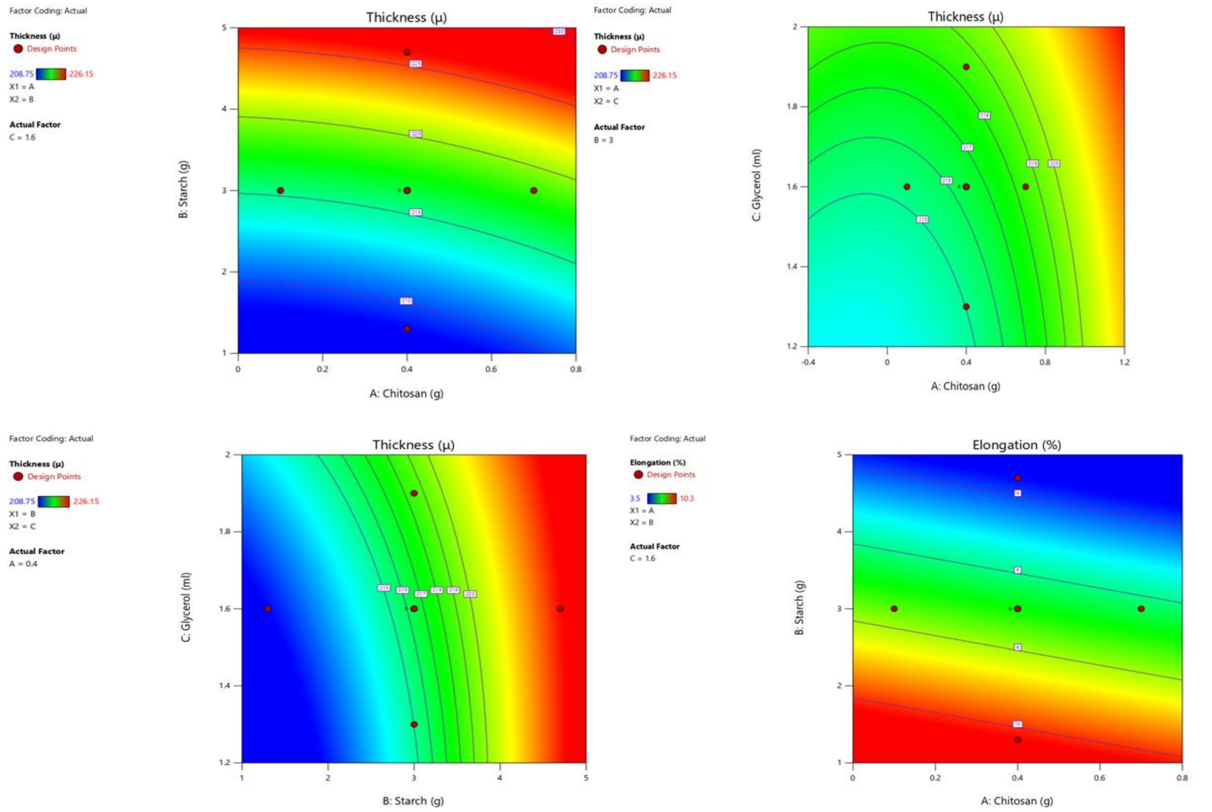


Figure 2: 3D Surface Plots for the responses. (A) Thickness- i. A vs B ii. A vs C iii. B vs C (B) Elongation (C) Young's Modulus (D) Water Contact Angle



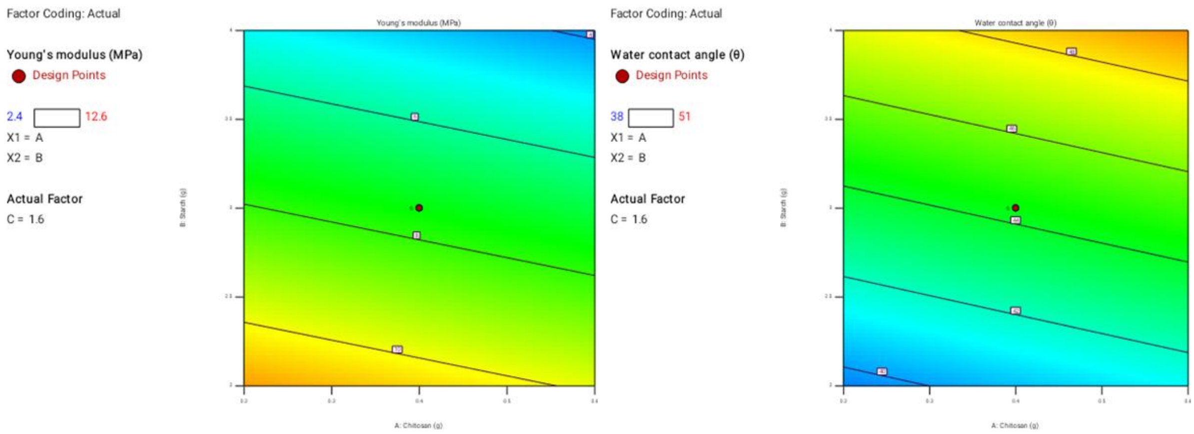


Figure 3: 2D Contour Plots for the responses. (A) Thickness- i. A vs B ii. A vs C iii. B vs C (B) Elongation (C) Young's Modulus (D) Water Contact Angle

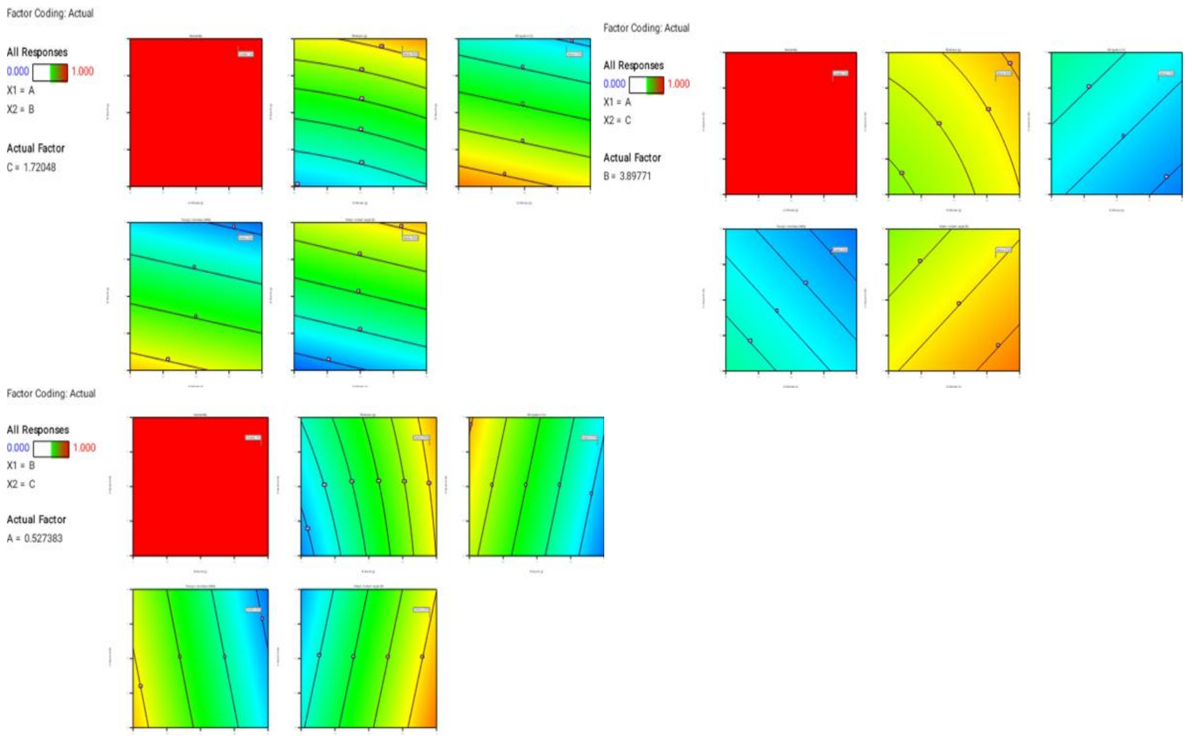


Figure 4: Optimization plots for all responses showing the optimized values for the factors Starch, Glycerol and Chitosan. (A) Glycerol=1.72 (B) Starch=3.89 (C) Chitosan = 0.527

1.4 RSM using Minitab

A RSM design was created by selecting it from the DOE (design of experiment) found in Stat in the menu. Here, select CCD and select the number of continuous factors (3), the number of runs (20) and input the names and low and high values as the factors. The factors taken were Chitosan, Starch and Glycerol.

The results are obtained in the form of summary tables and design tables. The resulting spreadsheet is then sorted in ascending order with respect to the standard order and the values of the responses were entered. The responses were Thickness, Elongation, Young's modulus and Water contact angle. After entering all the values, analyze the Response surface design and select all the 4 responses and the terms. For the graphs, all the model terms and the Normal and Pareto effect plots were selected. For the Residual plots, the Four in one option was chosen. Finally for storage, Fits and Residuals were selected.

The results for all the 4 responses against the 3 factors were obtained and were analyzed. The results included the ANOVA tables. Coded coefficients, the model summary, the regression equation and the Fits and

Diagnostics for Unusual Observations. The main effects, the interaction plots, the contour plots and the surface plots were attained. All the selected plots were obtained and interpreted.

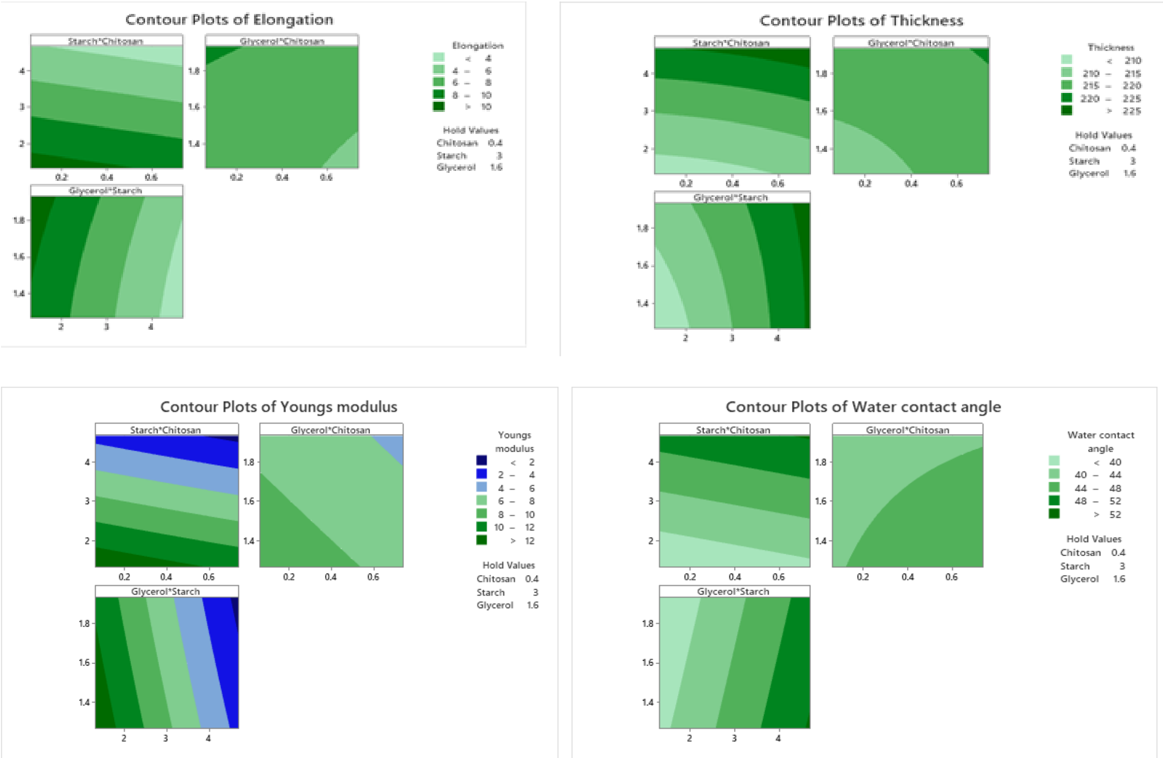


Figure 5: 2D Contour Plots (A) Elongation (B) Thickness (C) Young's Modulus (D) Water Contact Angle

II. RESULT AND DISCUSSION

The results obtained are as discussed below.

2.1 Bioplastics Produced

The bioplastic obtained from corn starch, potato starch and combination of both were successfully produced. Potato starch bioplastic films were shinier and more brittle compared to corn starch films. The bioplastic obtained by using the combination of both Potato and Corn starch resulted in better properties as well as lesser air bubbles.

2.2 Tests Performed

i). Hydrophobicity test: Water did not seep through the biofilm and dried eventually.

ii). Biodegradability test: Bioplastic was degraded sometime between 4 and 5 months.

iii). Solubility test: The thickness of the bioplastic decreased, and it had become more brittle as it lost a lot of its water content. After drying the obtained bioplastic for a day, it became very hard and curled in nature.

iv). Thermostability test: Curling was observed with no other changes observed.

2.3 RSM using Design Expert Software and Minitab Software

The model fit value of the four responses indicates that the model is significant. Final equation in terms of respective responses with the factors taken are obtained. This can be used for further studies. The response for thickness gives a quadratic equation and hence the factor coding involves 3 results of relationships between a combination of any 2 of the factors taken. The other responses give a linear equation. The optimized values for the factors using Design Expert Software are: Chitosan = 0.527, Starch = 3.89 and Glycerol = 1.72 for the mechanical values entered. The optimized values using Minitab Software are: Chitosan = 0.4, Starch = 3.0 and Glycerol = 1.6.

By analyzing the results from Minitab and Design Expert, the following relations were obtained:

i. Thickness: As the concentration of chitosan, starch and glycerol increased, the thickness also increased and it was found to be directly proportional to all the 3 factors.

ii. Elongation: Elongation was found to be directly proportional to glycerol, but indirectly proportional to Chitosan and Starch.

iii. Young's modulus: young's modulus is indirectly proportional to chitosan, starch and glycerol.

iv. Water contact angle: Water contact angle is directly proportional to starch and chitosan, whereas it is indirectly proportional to glycerol.

III. CONCLUSION

It was observed that the Bioplastics produced were of good quality. Repeated slight modifications such as amount of ingredients and temperature could make a vast difference. The Bioplastic in a semi-solid form also serves as a fair enough coating which could be applied to making candy wrappers. The bioplastic from the same source would have different mechanical properties if the composition is modified even slightly. From these results one can get the optimum values of the factors that are required to get the most efficient bioplastic with the best characteristics.

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