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The Feasibility of using Chitosan and Glycerin with Cocoa (*Theobroma cacao*) Pod Husk Flour as Starch to Create Bioplastic Films for Food Packaging.

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CHAPTER I

INTRODUCTION

Background of the study

Plastic pollution has become one of the most pressing environmental issues, as the rapidly increasing production of disposable plastic products overwhelms the world's ability to deal with them. Plastic pollution is most visible in developing Asian and African nations, where garbage collection systems are often inefficient or nonexistent. Plastic production increased exponentially, from 2.3 million tons in 1950 to 448 million tons by 2015. Production is expected to double by 2050. Furthermore, plastics often contain additives making them stronger, more flexible, and durable. However, many of these additives can extend the life of products if they become litter, with some estimates ranging to at least 400 years to break down (Parker, 2019). It has come to the point that they create problems for wildlife and their habitats as well as for human populations (Moore, 2023).

Internationally, a study was conducted entitled "Preparation and characterization of chitosan-based bioactive films incorporating Moringa oleifera leaves extract" (Chan Matú et al., 2021). The researchers utilized the combination of moringa leaves, chitosan, and glycerin to make bioactive films. The study conducted further stated that the addition of bioactive components, especially antioxidants, and plasticizers are good strategies to improve the properties of edible films. As stated by the study, the moringa leaves ethanolic extract acted as a barrier to light transmission; glycerin and moringa oleifera leaves extract exhibited an ant plasticizing effect, and antioxidant activity; and chitosan increased the antimicrobial activity and improved mechanical properties, while reducing the hydrophilic characteristic. Each variable in the study was used to support this claim because each one possesses a distinctive property, eventually leading to the success of their study. Moreover, the moringa oleifera leaves extract has similar properties to the Cocoa (*Theobroma cacao*) Pod Husk specifically it's high content of phenolic acids. Natural phenolic compounds are ideal candidates for food packaging because they exhibit antioxidant and antibacterial activities along with a variety of relevant properties such as UV-light resistance, color, flavor, and sensory attributes (Singh et al., 2022).

A national study was carried out to investigate indigenous plants, including the potato, cassava, and gabi tubers, with the addition of glycerin as a biodegradable plastic. Cataquis, Angeles, and Dancel's 2019 study, "Utilization of Indigenous Plants as an Additive for the Manufacture of Biodegradable Plastics," at the National University of the Philippines, examined the use of starch-based biodegradable polymers from indigenous plants to create biodegradable plastic while accounting for the material's performance during use. According to the study, starch can be turned directly into a biodegradable plastic, but it must be combined with glycerin and other indigenous plants ideal for it to prevent the material from swelling and deformation when exposed to moisture. The application of glycerol to cellulose derived from bioplastics will impact the tensile strength, as stated in the study. This was further demonstrated with the cassava and glycerol setup, which showed the highest percent elongation but the lowest tensile strength among all the setups. Glycerol is intended to decrease cellulose's molecular bonding power and increase the flexibility of bioplastics. The researcher from the study encouraged other researchers to make use of other available indigenous plants with the incorporation of distilled water, corn starch, and glycerol as biodegradable plastics to help reduce plastic pollution and find other alternatives (Cataquis et al., 2019).

A study on the production of bioplastic films was also carried out locally. A research study carried out at Ateneo de Davao University concluded that "bioplastic films from modified pectin and silicified crystalline cellulose-reinforced corn starch are a promising replacement for commercial packaging plastic products" (Ferolin et al., 2019).

Globally, cocoa farmers produce around five million tons of cocoa beans per year. Among the Asian countries, the Philippines has a competitive advantage for cocoa production due to its strategic location, good climate conditions, and favorable soil (DOST, 2019). Local annual production of cocoa is estimated between 10,000 and 15,000 tons, most of which is produced in the Davao region of Mindanao, the most southern island of the Philippines (Silva, 2021). The cocoa fruit's pod husk, or outer shell, is frequently overlooked, discarded as waste, and left to pile up after harvest. Nonetheless, the cocoa pod husk is recyclable, and due to its mechanical and rheological qualities, it can be used to produce biodegradable films (Almeida, 2021). On the other hand, the sugar chitosan, which is derived from the outer skeleton of seafood such as shellfish is qualified for food protection due to its antibacterial and antifungal qualities. Given that chitosan is compatible with both natural and synthetic polymers in films, such as the cellulose present in the husks of cocoa (*Theobroma cacao*), it can also aid in strengthening the mechanical qualities of bioplastic films (Utami et al., 2021). Moreover, glycerin or glycerol weakens (low tensile strength), promotes breakdown of bioplastic in wet and dry soil, and enhances flexibility (greater elongation at break). These three factors combined qualify for the development of biodegradable films.

This study aims to assess the feasibility of the bioactive components, namely cocoa (*Theobroma cacao*) pod husk flour as starch combined with glycerin and chitosan for general use. Now, in an effort to address plastic pollution and provide an alternative to plastic manufacture, researchers are concentrating on the combination of the three components that have been shown to be suitable for the development of biodegradable films. Finally, this work has the potential to further research and understanding of the characteristics and applications of glycerin, chitosan, and cocoa (Theobroma cacao) pod husk. It may also encourage the usage of environmentally friendly bioactive components.

Statement of the Problem

This study seeks to find out the viability of using Chitosan and Glycerin with Cocoa (*Theobroma cacao*) pod husk flour as starch to create a bioplastic film and an alternative to plastic films used in food packaging. Specifically, it is sought to answer the following questions:

1. Can Cocoa (*Theobroma cacao*) pod husk flour as starch combined with Glycerin and Chitosan be made to a bioplastic film?

2. Can Cocoa (*Theobroma cacao*) pod husk flour as starch combined with Glycerin and Chitosan be an alternative to plastic films used in food packaging utilized by the food industry?

3. Will it be equally effective and beneficial as other bioplastics, or plastic food packaging in general?

4. Will the production of this bioplastic help to reduce the current plastic pollution crisis?

Statement of the Hypothesis

The following are the research hypotheses employed in this study:

1. **Null Hypothesis**: If the Cocoa (*Theobroma cacao*) pod husk flour as starch with the combination of Glycerin and Chitosan are not feasible as bioplastic films, then usable food packaging cannot be created with these variables.

2. **Alternative Hypothesis:** If the Cocoa (*Theobroma cacao*) pod husk flour as starch with the combination of Glycerin and Chitosan are feasible as bioplastic films, then usable food packaging can be created with these variables.

Significance of the Study

The goal of this research is to use environmentally benign materials and bioactive components that can be converted into bioplastic films with antibacterial qualities and acquire a shortened degradation time, such as glycerin, chitosan, and cocoa pod husk. Therefore, it would be valuable for the following if the results of this study were positive.

This study is deemed useful for:

Community: The results of this study could offer them guidance in addressing their plastic waste-related challenges, ultimately helping in the prevention of the harmful effects of plastic pollution on the environment. Further, the product might provide a means of livelihood and income for community members.

Government: The study has the potential to inform them about strategies for reducing the environmental impact of plastic waste. They might influence this research to promote the adoption of biodegradable plastics over non-degradable ones, which, in turn, reduces the need for extensive clean-up efforts. Furthermore, the provinces and municipalities of the nation might be able to introduce this product as another source of income.

Garbage Collectors: This study has the potential to assist them in reducing the accumulation of plastic waste, which can lead to a decrease in plastic pollution and its harmful consequences.

Food Industry: With this product, companies in the food industry will be able to adopt a more environmentally friendly option for their product's food packaging, which will help them lessen their own plastic pollution footprint.

Future Researchers: This study can be utilized as a reference and instructional tool to undertake research into expanding the incorporation of the cocoa plant in various investigations. By broadening the current study's scope, they have the opportunity to draw more universally applicable conclusions.

Department of Science and Technology: This study will aid DOST in gathering recent and new data on the cocoa plant, particularly its pod husk and its advantages for environmental preservation.

Scope and Delimitation

The aim of this research is to investigate the potential of cocoa (*Theobroma Cacao*), pod husk starch, glycerin, and chitosan as bioactive components for the production of biodegradable films intended for use in food packaging, other things will also be utilized like the distilled water and Vinegar. The objective of this experiment is to determine the thickness of the biodegradable film. When assessing whether or not the aforementioned bioactive variables are feasible for the creation of biodegradable films, these will be the factors that the researchers consider; other indicators will not be included and will be restricted to the previously specified.

Definition of Terms

Cocoa Pod Husk -

- is a waste by-product of the cocoa industry, obtained after the removal of the cocoa beans from the fruit. The fibrous shells surrounding cacao beans. It acquires mechanical and rheological qualities.

Chitosan -

- is a sugar that comes from the outer skeleton of shellfish, including crab, lobster, and shrimp. Chitosan aids in the antibacterial and antifungal properties.

Glycerin -

- makes bioplastic more flexible (higher elongation at break), weaker (low tensile strength), and easier to degrade under wet and dry soil. It aids in the degradation process.

Distilled Water -

- is the solvent to be used in the washing process when producing starch or when combining glycerin, chitosan, and cocoa pod husk starch.

Starch -

- is a natural polymer; Amylose content in starch is a key factor in the manufacture of bioplastics since it is responsible for gelatinization and retrogradation, which are required during film formation.

Vinegar -

- is usually often added to starch-based biopolymers to modify the molecular structure of the starch, making it stronger and more workable.

CHAPTER 2

METHODOLOGY

The experiment involves glycerin and chitosan as its two independent variables. The control set-up will consist of one 100% pure glycerin set-up and one 100% pure chitosan set-up.

The investigation is divided into five phases: Phase I - Preparation of Materials, Phase II - Making the Cocoa (*Theobroma Cacao*) Pod Husk flour, Phase III - Mixing and Heating all the Ingredients, Phase IV - Pouring the mixture into a Mould/Tray and Drying, Phase V - Evaluating the Set-ups, and Phase VI - Proper Disposal of Materials Used.

Materials/Ingredients

The researchers will use a variety of materials and ingredients in each set-up throughout the whole experimental study. The materials and ingredients will be as follows: Cocoa Pod Husk, Chitosan, Glycerin, Distilled Water and Vinegar are the dependent and independent variables seen in each set-up measured using measuring spoons/kitchen scale together with a bowl for mixing; Knife, Heat Source, sieve, bowl, pan, and meat tenderizer/hammer will be used in the making of the Cocoa (Theobroma Cacao) Pod Husk flour; heat source, pan, bowl, spatula and together with the variables will be used to create the mixture; and a Mould/Tray where the mixture will be placed upon drying.

PHASE I - Preparation of Materials

The cocoa (*Theobroma Cacao*) Pod Husk will be obtained from the farm of one of the researchers, namely from Barangay Subasta, with the authorization of the Cocoa landowners; a sieve, knife, and meat tenderizer/hammer will also be prepared for the flour production. Chitosan and Glycerin, on the other hand, will be acquired in stores online; water, vinegar, a bowl, a heat source, a pan, measuring apparatus, a Mould/Tray, and a spatula will also be prepared. Moreover, the different ratios will be prepared by the researchers: 100 mL of water : 1 tbsp of cocoa pod husk flour : 10 mL of vinegar : 10 mL of glycerin, 100 mL of water : 1 tbsp of cocoa pod husk flour : 10 mL of vinegar : 2 tsp of chitosan, 100 mL of water : 1 tbsp of cocoa pod husk flour : 10 mL of vinegar : 5 mL of glycerin : 1 tsp of chitosan.

PHASE II - Making the Cocoa (*Theobroma Cacao*) Pod Husk flour

Cocoa pod husks will be cut into strips. Following that, the strips will be kept on a dehydrator set at 70°C for six hours. To make the flour, the dehydrated cocoa pod husk strips will be blended and then sieved.

PHASE III - Mixing and Heating all the Ingredients

In a bowl, 100 mL of water, 1 tbsp of cocoa pod husk flour, 10 mL of vinegar, and 10 mL of glycerin will be mixed for set-up A; 100 mL of water, 1 tbsp of cocoa pod husk flour, 10 mL of vinegar, and 2 tsp of chitosan will be mixed for set-up B; 100 mL of water, 1 tbsp of cocoa pod husk flour, 10 mL of vinegar, 5 mL glycerin and 1 tsp of chitosan will be mixed for set-up C. Once the combinations are ready, put a pan containing ¼ cup of water on top of the heat source and place it to medium heat. The bowl containing the mixture will be positioned above the pan and mixed until it becomes slightly solid when the water reaches a boil.

PHASE IV - Pouring the mixture into a Mould/Tray; Oven-drying and sun-drying

After that, the slightly solid mixture will be thinly placed on a tray or mold with the appropriate labels for each setup. All the set-ups will be placed on the oven to dry in a temperature of 105 degrees Celsius for an hour. Then, each setup will be further dried for 10 days in the same location where it can get sufficient heat.

PHASE V - Evaluating the Set-ups

After drying them, the researchers will check and measure the biodegradable film's appearance and texture. Due to the researchers' limited resources, the only biodegradable film factor used to test the product's utility is its appearance and texture.

PHASE VI - Proper Disposal of Materials Used

The researchers will use disinfectants to clean the utensils and equipment before sterilizing them following the experiment. The experiment's wastes will all be disposed of in the appropriate garbage cans.

Set-ups

This study's experimental design for biodegradable film manufacture as food packaging using cocoa (*Theobroma Cacao*) Pod Husk flour as starch with glycerin and chitosan is made up of setups and combinations of dependent and independent variables.

Set-ups	Trial 1	Trial 2	Trial 3
Set-up A (Cocoa Pod husk flour combined with Glycerin)	100 mL of Water	100 mL of Water	100 mL of Water
	1 tbsp of cocoa pod husk flour	15 mL of Cocoa Pod Husk Flour	15 mL of Cocoa Pod Husk Flour
	10 mL of Vinegar	10 mL of Vinegar	10 mL of Vinegar
	10 mL of Glycerin	10 mL of Glycerin	10 mL of Glycerin
Set-up B (Cocoa Pod Husk Flour combined with Chitosan)	100 mL of Water	100 mL of Water	100 mL of Water
	1 tbsp of cocoa pod husk flour	15 mL of Cocoa Pod Husk Flour	15 mL of Cocoa Pod Husk Flour
	10 mL of Vinegar	10 mL of Vinegar	10 mL of Vinegar
	2 tsp of Chitosan	2 tsp of Chitosan	2 tsp of Chitosan
Set-up C (Cocoa Pod Husk Flour combined with Glycerin and Chitosan)	100 mL of Water	100 mL of Water	100 mL of Water
	1 tbsp of cocoa pod husk flour	15 mL of Cocoa Pod Husk Flour	15 mL of Cocoa Pod Husk Flour
	10 mL of Vinegar	10 mL of Vinegar	10 mL of Vinegar
	5 mL of Glycerin	5 mL of Glycerin	5 mL of Glycerin
	1 tsp of Chitosan	1 tsp of Chitosan	1 tsp of Chitosan

Research Method

The method of this experiment is an experimental feasibility study because the researchers will use experimentation to find out whether the variables are deemed possible or achievable, in other words feasible. A feasibility study is a detailed analysis that considers all of the critical aspects of proposed project in order to determine the likelihood of its success (The Investopedia Team, 2023).

Chapter III

RESULTS AND DISCUSSIONS

This chapter presents an overview of the data gathered and the findings. The three trials were divided into distinct figures by a table including images of every setup from the researchers' prior investigations. The researchers' observations regarding the appearance and texture of the three different bioplastic film set-ups—cocoa pod husk flour combined with glycerin, cocoa pod husk flour combined with chitosan, and cocoa pod husk flour combined with both glycerin and chitosan—are stated after each figure.

Trial 1

Figure 1

Set-ups	Trial 1
Set-up A (Cocoa Pod husk flour combined with Glycerin)	
Set-up B (Cocoa Pod Husk Flour combined with Chitosan)	

Set-up C (Cocoa Pod Husk Flour combined with Glycerin and Chitosan)	

Figure 1 shows that there is a difference between the biofilms in each set-up in terms of appearance and texture after 10 days of drying. It shows that in the produced film Set-up A, bubble formation became visible. The film is light brown and somewhat translucent, with remnants of the cocoa pod husk powder still visible. Even after drying, the film in Set-up A has a sticky texture. The biofilm created on Set-up B is somewhat darker brown than on Set-up A and closer to the color of Set-up C. After oven-drying, Set-up B became hard, dry, and brittle; the hardened bubble formation is still apparent along with bits of the Cocoa Pod Husk Flour. The cracked product is slight to not translucent. As for its texture, Set-up B has a hard surface. Lastly, in Set-up C, the color of the biofilm is slightly darker brown than in Set-up A. Bubble formation is still apparent with little particles of Cocoa Pod Husk flour and is slightly translucent. Set-up C has a texture that is slightly sticky on one side and nearly smooth on the other, a texture similar to that of a plastic film.

Trial 2

Figure 2

Set-ups	Trial 2
Set-up A (Cocoa Pod husk flour combined with Glycerin)	
Set-up B (Cocoa Pod Husk Flour combined with Chitosan)	
Set-up C (Cocoa Pod Husk Flour combined with Glycerin and Chitosan)	

Figure 2 illustrates that the biofilms in each setup for the second trial differ in look and texture after 10 days of drying. It demonstrates how bubble formation is visible in the biofilm created by Set-up A. The film is relatively translucent and pale brown, making it the lightest of all the setups. In the biofilm, brown traces of the cocoa pod husk powder are still discernible. Even after drying, the film in Set-up A has a sticky texture but does not stick to the skin. The biofilm produced by Set-up B is a little bit darker brown than that of

Set-up A. Upon oven drying, Set-up B turned crisp, firm, and dry; the crisped bubble formation and fragments of the cocoa pod husk flour in the cracked product are still visible on one side of the film and are not translucent. Set-up B features a firm surface in terms of texture. Finally, in Set-up C, the biofilm is slightly darker brown, almost black. With tiny bits of cocoa pod husk flour, bubble formation is still noticeable and is only faintly transparent. Set-up C has a texture that is practically smooth on both sides, similar to a plastic film.

Trial 3

Figure 3

Set-ups	Trial 3
Set-up A (Cocoa Pod husk flour combined with Glycerin)	
Set-up B (Cocoa Pod Husk Flour combined with Chitosan)	
Set-up C (Cocoa Pod Husk Flour combined with Glycerin and Chitosan)	

As shown in *Figure 3,* the biofilms in each setup for the last trial differ in appearance and texture based on the researchers' observations following ten days of drying. There is clear bubble formation in the bioplastic film produced by setup A. The film is a light brown color and somewhat transparent. There are still noticeable dark particles of the cocoa pod husk powder in the biofilm similar to the previous trials. The film in Set-up A has a sticky texture and slightly adheres to the skin even after drying. In contrast to Set-up A, Set-up B generated a darker brown biofilm. Set-up B dried in the oven until it became crisp, and dry. However, in trial 3, Set-up B did not crack. On one side of the film, which is partially to completely translucent, the crisped bubble formation with pieces of the cocoa pod husk flour is still visible. The surface of Set-up B has a hard texture. Lastly, the biofilm is somewhat lighter and darker brown—almost black—in Set-up C. There are still traces of cocoa pod husk flour present, and the bubble creation is just slightly translucent. Set-up C's texture is nearly smooth on both sides, resembling a standard plastic film.

CHAPTER IV

CONCLUSION AND RECOMMENDATION

Conclusion

Based on the findings, a useable bioplastic film with the ideal texture and appearance across all setups was successfully created using chitosan and glycerin in combination with cocoa (*Theobroma cacao*) pod husk flour. This film's appearance—more especially, its color and visible particles—may differ significantly from a regular plastic film, but its texture—which is almost entirely smooth on both sides—is considered as well to be comparable to a regular plastic. These factors are therefore viable and have the potential to be used in the manufacturing of bioplastic sheets for food packaging.

Recommendations

The researchers recommend to the future researchers:

1. To use other methods in drying the bioplastic films;
2. To choose a fruit that has more starch content when extracted;
3. To increase the blending time of the variable to be made as starch to produce finer starch; and
4. To assess other bioplastic film quality factors for a better evaluation.
5. To investigate the degradation of the setups made.