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Production of Agar-Agar and Sago based Bioplastic

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Abstract: Plastic garbage is the world's third-largest waste source, posing a threat to human health and the environment. Because of their outstanding barrier properties, stiffness, tensile strength, and rip strength, petrochemical plastics have long been used as packaging materials. Plastics have a lot of disadvantages, including a low water vapour transmission rate, non-biodegradability, and other difficulties that lead to environmental degradation. Newer solutions for the use of bio-plastics have emerged, all while keeping pollution and environmental harm in mind. Bio-plastics are primarily made from biological resources such as potatoes, potato peels, corn, sugarcane, wheat, and rice, as well as seaweed-based bioplastics including varied proportions of agar-agar, seaweed, starch, or cellulose, as well as plasticizers and their relative impact on physical-properties, and the bioplastic characteristics.

Replacing petroleum plastic with bioplastic is an alternate strategy to reduce plastic waste in human life while also being more environmentally friendly. As a result, the goal of this research is to gelatinize agar-agar and sago powder to make bioplastic and analysis of its properties. Unlike earlier investigations, this one looked at and evaluated new bioplastic formulations with varying ratios of agar-agar and sago to glycerol samples of 1:0.5, 1:1, and 2:1, referred to as Sets A, B, and C, respectively. The bioplastic samples (Sets A to C) with varying ratios of Agar-Agar and Sago powder to glycerol were found to have higher biodegradable qualities when the ratio of Agar-Agar and Sago powder to glycerol was 1: 1 (Set B) and It had the smallest capacity for holding water. According to the findings, Set B could only hold only 5.48 percent of the water, preventing water from interacting with the wrapped contents. In addition, as compared to other samples, Set B appears to deteriorate better in soils and dissolve more in ethanol, acetone, and oils. Set B has the potential to be employed as a fertilizer coating or other related packaging to reduce the usage rate of the petrochemical plastic since the bioplastic can decay naturally by the ethanol produced by bacteria in soils under anaerobic reactions.

Keywords: Bioplastic, Glycerol, Agar-Agar, Sago Powder, Gelatinization, Biodegradable.

I. INTRODUCTION

Humans have produced 8 billion tons of petroleum-based plastic in the previous 60 years, and the rate of production continues to rise [1]. Polyethylene, polyvinyl chloride, and polystyrene are some of the most regularly utilized ingredients in plastic production [2]. Medical devices, food packaging, toys, containers, mobile devices, sports equipment, and other objects are all examples of plastic applications [3]. Incorrect plastic waste management and unsuitable human behavior, on the other hand, result in an excess of plastic trash [14]. Most petroleum-based plastics have been around for a long time and are less sustainable than bio-based plastics [5], as well as being harder to recycle [6]. Furthermore, petroleum-based plastics are frequently broken down into micro plastics, which can harm marine ecosystems and human health [7]. Bioplastics are divided into two types: biodegradable and bio-based. Biodegradable plastics focus on the ability of the end products to degrade, whereas bio-based plastics focus on the raw material employed [8]. Biodegradable polymers are compostable, whereas bio-based plastics are not [9]. Aside from that, greenhouse gases such as carbon dioxide are released into the environment during the biodegradation of plastics [10]. Biodegradable plastics can be manufactured from fossil, renewable, or renewable-blend materials. The ultimate chemical structure of bioplastics, on the other hand, can alter their biodegradability. As a result, raw ingredients for bioplastics that influence their ultimate chemical structure, including their functional group, must be considered [11].

Biodegradable bioplastics, on the other hand, can be both photodegradable and biodegradable [12]. Photodegradable bioplastics are light-sensitive [10], with light-sensitive groups that link the polymer bioplastics' backbones acting as additive's to capture light for the degradation process [12-13]. Furthermore, when bioplastics are subjected to high levels of ultraviolet radiation for long periods of time, their polymeric structure degrades, which is referred to as photo degradation. This scenario may later allow for further bacterial breakdown of the disintegrating bioplastics [2]. When natural sunshine is inadequate to complete the process, however, photo degradation of bioplastics might be challenging. As a result, biodegradation of bioplastics appears to be more promising than photo degradation.

Despite this, microorganism-derived biodegradable plastics can create biopolymers such as polyhydroxyalkanoate, aliphatic polyesters, polylactides, and polysaccharides [14]. There is also semi-biodegradable plastic, which is a type of starch connecting plastic that links short polyethylene or polyethylene fragments [15]. However, due to a fragment of polyethylene or polyethylene that prevents bacteria from attacking the starch and several environmental variables, this type of bioplastic cannot totally breakdown [2]. As a result, chemical, physical, mechanical, and biological properties of distinct types of bioplastics can be distinguished. Furthermore, these qualities may alter over time, resulting in varying levels of tissue compatibility from their source components and making them difficult to breakdown.

Therefore, while having an acceptable usage period, biodegradable plastics should not generate a prolonged erythrogenic and harmful effect to human health. Furthermore, the time it takes for bioplastics to regenerate should be the same as the time it takes for them to degrade. Also, during the decomposition of bioplastics, no poisonous or harmful compounds that cannot be digested or removed by a human's body should be released. Its permeability and processability should also be acceptable for its intended use [16]. However, bioplastics have a high production cost and cannot be produced in sufficient quantities to replace petroleum-based plastics [17]. Due to its low cost and availability, agar-agar and sago blends can be one of the most convincing raw materials [18]. According to market research agar-agar and sago are cultivated and harvested in many areas of India, Japan, China and many more countries. Agar-Agar is a jelly-like substance made up of polysaccharide chains extracted from the cell walls of red algae and sea weeds, particularly ogonori (*Gracilaria*) and "tengusa" (*Gelidiaceae*). Sago, on the other hand, is nearly pure starch, consisting of 88 percent carbohydrate, 0.5 percent protein, and trace amounts of fat, with barely a trace of B vitamins. Furthermore, a bioplastic based on sago starch can act like a thermoplastic and have superior thermal and mechanical properties [18].

This study intends to gelatinize biodegradable agar-agar and sago-based bioplastics in a simple, more eco-friendly, and environmentally beneficial manner. Agar-Agar and Sago can be utilized in a variety of commercial packaging materials at a low cost, and they have similar content qualities to other starch sources, resulting in a more stable decomposition temperature. Unlike earlier investigations, this one looked at and evaluated new bioplastic formulations with varying ratios of agar-agar and sago to glycerol samples of 1:0.5, 1:1, and 2:1, referred to as Sets A, B, and C, respectively. Water absorption analysis, soil degradation analysis, photodegradation analysis, dissolving in acetone, ethanol, and oil analysis were among the characterization tests performed.

II. MATERIALS AND METHODS

A. Agar-Agar and Sago Characteristics

- 1) *Agar-Agar*: Chemically, the agar-agar is defined by a continuous repeating unit of Galactose and 3,6-anhydro-L-galactose with minimal changes and a low ester sulphate content. The structure of agar is made up of two polysaccharide groups: agarose, a neutral polymer, and agar, pectin, an oversimplified term for the charged polysaccharide [39-41]. Agarose is responsible for agar's capacity to gel, which makes it particularly useful in skin care, herbal medicine, and pharmaceutical applications; it also has good film characteristics. Agar-agar is widely utilized in the commercial food processing industry because of its capacity to act as a stabilizer, emulsifier, and thickening agent. Both are currently used in gel-based foods such sweets, jams, and jellies, as well as baked items. Agar gels are usually tight and lucid, but adding carbs increases their strength [43]. Agar films are physiologically inert and can easily interact with various bioactive substances and/or plasticizers to aid in the creation of elastic and soft gels [44-46]. They also have a low hydroscopic property, which is advantageous in the packaging industry.
- 2) *Sago Powder*: Sago is almost entirely made up of starch, with only a trace of B vitamins and an 88 percent carbohydrate, 0.5 percent protein, and trace levels of lipid composition. It's a common ingredient in soups, cakes, and puddings in the southwest Pacific region. Its main application in the kitchen is as a pudding and sauce thickener. In industry, it's used as a textile stiffener. Sago starch-based biodegradable plastic is an environmentally friendly product. This is done to lower the chemical composition of the mixture by substituting the starch with a starch that does not contain hazardous chemical reactions when coupled with other biodegradable components. Because sago starch is a natural product, it is completely safe. Amylose is found in sago starch, and it rapidly bonds to other substances. When heated to 70 - 120 degrees Celsius, the sago starch gelatinizes.

B. Materials and Equipment

The materials needed to synthesize or to make agar-agar and sago based bioplastics were fine agar-agar powder, sago powder, distilled water, and glycerol, some amount of vinegar. In addition, this project used 98% pure glycerin as a plasticizer. In the meantime, it required other common laboratory equipment, such as a small beaker, a hot plate with a magnetic stirrer, a spatula, a balance, a pipette, and a Petri dish. Secondly I prepared the bioplastics with different ratios of (agar-agar, sago powder) to glycerol samples were 1: 0.5, 1: 1 and 2: 1, respectively and named as sets A, B and C, respectively.

C. Preparation of Agar-Agar and Sago Based Bioplastic

The agar-agar and sago-based bioplastics were made using a modified approach adapted from [20], and [21]. In a tiny beaker, required amount of agar-agar and sago powder were mixed with distilled water, vinegar, and glycerol. The respective mixer was kept well in the beaker. Here the vinegar was diluted with acetic acid, which was utilized to break down the amylopectin found in sago polysaccharides, and then added to the mixture. After that, the mixture was placed on a hot plate and heated to between 80 to 120 degrees Celsius. The stirrer was used to constantly stir the mixture until it boiled. The hot plate was switched off when the mixture became translucent. The mixture was stirred for another 2 to 5 minutes. The mixture was poured on the petri dish and moulded as thinly as possible after 2 to 5 minutes. At room temperature, the mixture was deposited and dried. When the bioplastic was totally dry, it was carefully removed from the petri dish. As previously mentioned, novel formulations with three different ratios of agar-agar and sago to glycerol samples (1:0.5, 1:1, and 2:1) were tested, and they were labelled Sets A, B, and C, respectively. To evaluate their qualities and characteristics, all of these samples were subjected to characterization procedures and biodegradation assessments.

D. Bioplastic Methodologies

1) *Biodegradation Analysis:* Biodegradation analyses were carried out in this part on six different aspects: water absorption, soil degradation, photodegradation, and dissolving in acetone, ethanol, and oil to properly analyze the properties of the bioplastic. All of these analyses might reflect the bioplastic's biodegradability qualities, and then to identify the bioplastic's prospective application and function. All of the samples were cut into 5 cm x 5 cm of squares for each biodegradation analysis test. Meanwhile, the relative change percentage (RC) for the weight of bioplastics was calculated for each of the biodegradation analyses using the equation (1) taken from [22]:

$$RC = \left[\frac{x_2 - x_1}{x_1} \right] 100 \quad \dots(1)$$

Where, x1 and x2 indicate the initial and final weights of the bioplastic, respectively after the designated biodegradation analysis.

2) *Water Absorption Analysis:* Water absorption is used to determine the amount of water absorbed under specified conditions. Factors affecting water absorption include: type of plastic, additives used, temperature and length of exposure. The data sheds light on the performance of the materials in water or humid environments. In this study, the water absorption test was done on the bioplastic samples according to the American Society for Testing and Materials (ASTM) D570 [23]. For the water absorption test, the specimens are dried in an oven for a specified time and temperature and then placed in a desiccator to cool. Immediately upon cooling the specimens are weighed. The material is then emerged in water at agreed upon conditions, often 23°C for 24 hours or until equilibrium. Specimens of 5 cm in diameter are removed, and dried with a clean piece of cloth, and weighed. The RC of the samples indicating the amount of water absorbed by the bioplastics were calculated using (1).

$$RC = \left[\frac{x_2 - x_1}{x_1} \right] 100$$

Where, x1 and x2 indicate the initial dry weight and final wet weight of the bioplastic sample, respectively after the designated biodegradation analysis.

3) *Soil Degradation Analysis by Soil Burial Test:* As per ASTM G21-70 the soil burial test is conducted of both indoor and outdoor microorganisms from the soil that react to the water within the biofilm and start decomposing due to breakdown of the polymer chain in the biofilm. The samples were weighed, and the initial weights were recorded. The samples were buried in the mineral soil taken from farm field site (near Jalgaon district region, Maharashtra, India) in the beakers. In the Soil Burial Test. Bioplastic samples (1 cm x 1 cm) were buried in compost soil at 7.5 cm depth, then incubated at room temperature for 10 days with sampling time every two days. The buried samples were then cleaned from the soil and weighed. The RC of the samples that showed the soil degradation rates of the bioplastics were determined by using equation (1).

$$RC = \left[\frac{x_2 - x_1}{x_1} \right] 100$$

Where, x1 and x2 indicate the initial and final weights of the bioplastic, respectively after the designated biodegradation analysis.

Later, the weight loss of the bioplastics was calculated using equation (1) and relatively defined the class score of bioplastic as per shown in Table – 1.

Score	The growth of fungi on the surface of bioplastic samples (%)
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0	0
1	10
2	10 – 30
3	30 – 60
4	60 - 100

Table 1- Biodegradability rank of bioplastics

4) *Photodegradation Analysis*: This test was referred from ASTM G21 – 70. According to that the respective samples were weighed, and the initial weights were recorded. Next, the samples were placed under sunlight in Raver, District Jalgaon, Maharashtra, India with the temperature of 23 °C to 42 °C [24] for 7 days. During the photodegradation analysis, there was only sunny day. The samples were collected and weighed every day. Any type of dirt present on the surface of the sample specimens was removed, and then the final weights were recorded accordingly. The same steps were repeated for all the sets specimens for 7 days, and the weight data were recorded. Then, the RC of the samples which denoted the photodegradation rates of the bioplastics were obtained using the equation (1).

$$RC = \left[\frac{x_2 - x_1}{x_1} \right] 100$$

Where, x1 and x2 indicate the initial and final weights of the bioplastic, respectively after the designated biodegradation analysis.

5) *Dissolution in Acetone Analysis*: The samples were weighed, and the initial weights were recorded. Then, the samples were placed in the beakers filled with acetone, and the samples were completely immersed in the acetone. All the beakers carrying specimen were covered with the preservative parafilm. The samples were taken out of the acetone after 24 hours. Then the samples were dried, and then the final weights were recorded accordingly. The same procedures were repeated to all other sets for 24 hours respectively, and the weights data were collected. After that, the RC of the bioplastics which displayed the dissolution rate of bioplastic in acetone were calculated using the equation (1).

$$RC = \left[\frac{x_2 - x_1}{x_1} \right] 100$$

Where, x1 and x2 indicate the initial and final weights of the bioplastic, respectively after the designated biodegradation analysis.

6) *Dissolution in Ethanol Analysis*: The samples were weighed, and the initial weights were recorded. Then, the samples were placed in the beakers filled with ethanol, and the samples were completely immersed in the ethanol. The beakers were covered with parafilm to avoid the evaporation of ethanol. The samples were taken out from the ethanol after 24 hours. Then the samples were cleaned, and then the final weights were recorded accordingly. The same steps were repeated by all sets for 24 hours and the weights data were collected. Next, the RC of the samples which illustrated the degradation rates of the bioplastics in ethanol was calculated using of the equation (1).

$$RC = \left[\frac{x_2 - x_1}{x_1} \right] 100$$

Where, x1 and x2 indicate the initial and final weights of the bioplastic, respectively after the designated biodegradation analysis.

7) *Dissolution in Oil Analysis*: The samples were weighed, and the initial weights were recorded. Then, the samples were placed in the beakers filled with cooking oil from reputed Company Group. The oil is a refined, bleached, and deodorized with high-quality sunflower oil which gives better taste, flavor and less odor. Then, the samples were completely immersed in the oil. The respective beakers carrying all sets specimen were covered with the help of preservative parafilms, and then the samples were taken out from the oil after 24 hours. After that, the remaining oil on the surface area of the samples was removed and the final weights were recorded. The same steps were repeated for other sets and all weight data were collected. Later, the RC of the bioplastics that indicated the degradation rates of the bioplastics in oil were determined using the equation (1).

$$RC = \left[\frac{x_2 - x_1}{x_1} \right] 100$$

Where, x1 and x2 indicate the initial and final weights of the bioplastic, respectively after the designated biodegradation analysis.

III. RESULTS AND DISCUSSION

A. Water Absorption Analysis

The water absorption of bioplastics was tested, and the results obtained were tabulated in Table 2. From Table 2, it can be seen that Set B had the lowest average water absorption of 5.48 % while Set A and Set C had the highest average 9.72 % and 6.93 % of water absorption. However, the only Set A had a markable increased water absorption rate at 23°C for 24 hours or until equilibrium respectively. It can be said that Sets B which had a high content of glycerol absorbed less water than Sets A and Set C. These results are due to glycerol that has a characteristic of hygroscopic which can increase the moisture content of bioplastic [33]. Therefore, the water absorption ability is correlated to glycerol concentration. Set B, which contained more glycerol as compared to Set A and Set C, showed better water absorption ability.

Sets	RC (%)
Set A	9.725
Set B	5.480
Set C	6.932

Table 2 - Calculation of relative change percentage (RC) of water absorption for Sets A, B and C.

B. Soil Degradation Analysis

The produced bioplastics are expected to degrade in soil due to their main raw materials, glycerol, and agar-agar and sago. Glycerol is a kind of plasticizer that is sustainable and biodegradable [34]. Meanwhile, a study had proved that agar-agar and sago could be biodegraded in soil with the presence of microorganisms in the natural environment [35]. In this study, soil degradation analysis for bioplastic was done for all the samples by burying them in the soil, and the results are summarized in Table 3. The Table 3 shows that the percentages of average soil degradation for three sets from 13.89 % to 10.65 %. These results indicated that all the sets were degradable in the soils since there was a reduction of weights. The bioplastics were degraded by microbes in the soil due to their natural materials. These results indicated that the cornstarch-based plastic blended with glycerol could degrade in the soil, and they had environmentally friendly characteristics.

However, Figure 4 that displays the average weights of bioplastic versus time shows that the biodegradable rates of the bioplastic samples were low. In Figure 4, Sets A and C had a similar and lesser biodegradable rate than Sets B. According to [36], the cassava starch or agar-agar bioplastic blending with the least amount of glycerol requires a longer time, which is 10 days to degrade in soil completely.

Thus, these results could be due to the lower content of glycerol in Sets A and C. The higher concentration of glycerol that is hydrophilic will attract more water which helps to increase the microbial growth and rapid the degradation process [36].

Sets	RC (%)
Set A	11.175
Set B	13.891
Set C	10.650

Table 3 - Calculation of relative change percentage (RC) of soil degradation for Sets A, B and C.

Therefore, all the limit scores of all the samples lie between 10 – 30 so, all the samples get the score of 2 as per Table 1.

C. Photodegradation Analysis

Photodegradation is a chemical reaction when the ultraviolet light provided by the sunlight is absorbed by the bioplastic and breakdown down the polymer chain. The obtained results for photodegradation is tabulated in Table 4. All the samples showed a reduction in the average weight of bioplastic. Sets A and C showed almost similar photodegradation percentages, which was around 6.2 %. Meanwhile, Sets B showed a relatively high reduction percentage in weight. The higher photodegradation rate was Set B, which was around 0.8 % per day. Set C had the lowest photodegradation rate, which was only around 0.6 % per day. In other words, bioplastic will require 100 to 200 days to degrade if it is fully under the sunlight. The weather was a huge influence factor in this analysis. Throughout these 7 days, every day was a sunny day except 1 day which was cloudy. As a result, 1 day out of 7 days showed the least reduction of weight for all the samples because of cloudy weather. According to [37], the presence of water will accelerate the degradation of the bioplastic. This is because water will wash off the degraded surface and provide a new surface of bioplastic to carry out photodegradation further [37]. Thus, rainy days will increase the photodegradation rate.

Sets	RC (%)
Set A	6.240
Set B	7.892
Set C	6.175

Table 4 - Calculation of relative change percentage (RC) of photodegradation for Sets A, B and C.

D. Dissolution in Acetone Analysis

Bioplastic samples were immersed in acetone to observe the reaction between acetone which consists of the organic compound, carbonyl group with the ignored. However, even though the weight loss of bioplastic was not obvious in this analysis, the properties of bioplastic changed. During the experiment, the bioplastic became a bit more brittle. Different conditions such as temperature, acetone concentration, external disturbance, etc. will affect the reaction of acetone with glycerol in the bioplastic and acetone can change the character of bioplastic [38]. Acetone is commonly used for sterilization in ophthalmic surgery or any medical equipment. Hence, these results showed that the produced bioplastics are not recommended to be used in medical industries which use acetone as bioplastics. All the results obtained were tabulated in Table 5. In Table 5, it is shown that there was little weight change in bioplastic in every set of samples. The average dissolve rate in acetone for all three sets of samples was in the range of 0.7 % to 1 %. These per cent values were relatively small, which can be a sterilization reagent too as it will dissolve when contact or immersed in an acetone solution. Besides, they are hard to be recycled using acetone as a solvent because their weight reduction in acetone is too small. It may require a longer time or a higher concentration of acetone to completely dissolve the bioplastic, which is not eco-friendly and economical.

Sets	RC (%)
Set A	1.279
Set B	0.865
Set C	0.730

Table 5 - Calculation of relative change percentage (RC) of dissolve rate in acetone for Sets A, B and C.

E. Dissolution in Ethanol Analysis

Apart from that, bioplastic was immersed in ethanol as well to investigate the relationship between bioplastic and ethanol, which is organic compound alcohol with a functional group of the hydroxyl group. Table 6 shows the initial and final weights of all the samples precipitate in ethanol.

As shown in Table 6, Set B showed the highest precipitation rate of 12.1 % in ethanol among all the sets. Besides that Sets C showed lowest precipitation rates which were 5.3 %, respectively. Compared to acetone, bioplastic precipitated much more in the presence of ethanol. Thus, it means that it is easier for bioplastics to be recycled using ethanol than acetone. For its application, these bioplastics are recommended to be used as fertilizer coating since they can be degraded by ethanol from bacteria in the soils that undergo anaerobic reactions.

Sets	RC (%)
Set A	8.251
Set B	12.115
Set C	5.370

Table 6 - Calculation of relative change percentage (RC) of precipitation rate carried in ethanol for Sets A, B and C.

F. Dissolution in Cooking Oil Analysis

The results of the dissolution test of bioplastic in cooking oil are tabulated in Table 7. Based on Table 7, all the samples showed a very low dissolution rate in cooking oil, which was around 0.3 % to 0.4 %. Here, Set B showed less dissolution as compared to other sets. In general world the oil-soluble bioplastic is not suggested to be used in the food and beverage packaging industries for oily food such as oil bottles or fried food packaging. The reduction in number of bioplastic weight in cooking oil is suspected to be due to the release of plastic particles, which are harmful to human health. Apart from that, oil is not suitable to be the medium for dissolving bioplastic for recycling purposes.

Sets	RC (%)
Set A	0.460
Set B	0.310
Set C	0.385

Table 7. Calculation of relative change percentage (RC) of dissolve rate in oil for Sets A, B and C.

IV. CONCLUSION AND FUTURE RESEARCH DIRECTIONS

In this study, bioplastics were produced from agar-agar and sago via gelatinization using different ratios of agar-agar and sago to glycerol, and its application is recommended based on its characteristics. It can be concluded that agar-agar and sago based plastic with a ratio of agar-agar and sago to glycerol (1:1), which is Set B acts more like plastic as compared to other sets since, Set B absorbs less water. It shows a high temperature tolerance, it is biodegradable in soil, photodegradable, and can dissolve in ethanol, albeit it was rarely dissolved in cooking oils. In the future, if the bioplastic needs to be recycled, it can be dissolved in ethanol and processed to make other goods.

For future studies it can be find out that how many times it can be recycled, more research is needed. Furthermore, additional research can be conducted to investigate the bioplastic's mechanical strength in order to improve the bioplastic's tensile strength and elongation break for future applications in a variety of fields. More time can also be spent studying soil degradation. Finally, different types of plasticizers can be combined with agar-agar and sago to investigate property changes. Varied plasticizers may have an effect on mechanical or thermal characteristics, resulting in different bioplastic behavior. Furthermore, creating homogeneous smooth sheets with a similar thickness to the bioplastic, investigating photodegradation using FTIR or Gel permeation chromatography, and assessing plastic mechanical strength can all be investigated further.

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