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Preparation and Characterization of Biodegradable Plastics Out of Food Wastes as Prospective and Eco-Friendly Medical Devices

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Abstract: Plastics are one of the most important petrochemical based polymers widely used for a variety of purposes. Most of these polymers are oil-based and are made by either addition or condensation techniques. Their adverse effect on the environment because of their resistance to microbial degradation is a matter of serious concern at present throughout the world. Biodegradable plastics is considered as a better alternative for fossil fuel plastics or petro plastics as they degrade naturally by microbial action and at a much faster rate. These biodegradable plastics can be derived from natural and renewable feedstock like cornstarch, pea starch, vegetable and fruit peels, vegetable fats and oils or micro biota. Such bio plastics are not a single class of polymers but rather a family of products. This work was aimed at utilizing food wastes like banana peels, corn starch etc., for making biodegradable plastics using simple laboratory methods and also finding new areas of the use of the plastic material that was manufactured. Using a food waste, it was possible to synthesize a bi oplastic, which, unlike the petroleum-based plastic, was not found to cause any environmental pollution. Further, on studying the mechanical, physical and chemical properties of the bioplastic material produced, it will be possible to assess the potential of its prospective applications in various fields including packaging, 3D-printing and preparation of medical devices. The hard material produced from corn-starch can be used to prepare containers. Analysis of its characteristics like water solubility and thermo tolerance also promises new avenues for medical application of the bioplastics as drug delivery systems, biodegradable bandages and suture threads.
Keywords: Bioplastics, Biodegradable, natural and renewable feedstock, eco-friendly, medical devices, drug delivery systems, bandages.

I. INTRODUCTION

Plastics have become an indispensible part of our everyday-life and so versatile in their usage, they cannot be completely avoided. However, they have become a major threat to the environment contributing to a major part of the land pollution [1].Plastics account for 25% of the total volume of landfills. The plastics produced in excess and discarded here and there are deposited as a landfill and are degraded very slowly. Hence they remain in the landfills for hundreds or even thousands of years [2].

Plastics that are made from renewable resources (plants like corn, tapioca, potatoes, sugar and algae) and which are fully or partially bio-based, and/or biodegradable or compostable are called bioplastics. The diminishing supply of petroleum along with the non-biodegradability of these petrochemical-based materials has been a source of environmental concerns and hence, the driving force in the search for ‘green’ alternatives like starch-based biodegradable plastics. Starch is a natural biopolymer consisting predominantly of two polymer types of glucose namely amylose and amylopectin. Amylose is essentially a linear polymer of glucose linked together by α-1,4 bonds while amylopectin is a branched polymer consisting of both α-1,4 and α-1,6, glycosidic linkages, with the latter found at branch points [3]. The amylose/amylopectin ratio, which is a function of the starch source, is significant as it affects some physicochemical properties of starch and in turn, influences its functionality and eventual applications. The advantages of starch for plastic production include its biodegradability, renewability, good oxygen barrier in the dry state, abundance and low cost [4,5]. Starch is a good biodegradable filler candidate because it possesses satisfactory thermal stability and causes minimum interference with melt-flow properties of most materials used in the plastic industry unlike common cellulosic fillers such as woodflour and paper pulp that were found to interfere with flow properties[6,7]. During starch extrusion, the combination of shearing, temperature and plasticization allows the production of a melted thermoplastic material [8]. Afterwards, this material can be transformed by means of thermoforming or injection moulding.

In this regard, starch has been used as fillers, thermoplastic starch (TPS), in the production of biodegradable synthetic polymer like polylactic acid (PLA), foamed starch and starch-synthetic polymer blends.Starch-based bioplastics are not a single class of polymers but rather a family of products. They have the ability to absorb humidity and hence they are being widely used for the production of capsules in pharmaceutical sector. Different types of bioplastics are combined to form materials with improved properties like increased thickness, plasticity and enhanced waterproof capacity. Banana peels are food wastes rich in starch and hence used for production of bioplastic. The ripe banana peel contains 6-9% dry matter of protein, 20-30% fibre and about 40% starch in addition to 88% moisture. Banana produces huge amount of cellulosic waste which is disposed in landfills. As the peels rot, they produce methane gas which accounts for 20% methane emissions, thus forming a major contributor to global warming. Hence, redirecting these solid wastes of banana peels into something useful and beneficial is essential. Other starch-rich natural and renewable feedstock successfully used for bioplastic production include cornstarch, potato, peastarch, vegetable and fruit peels, vegetable fats and oils or microobiota [9].

Bioplastics are used in an increasing number of markets – from packaging, catering products, consumer electronics, automotive, agriculture/horticulture and toys to textiles and a number of other segments. While production of most bioplastics results in reduced carbon dioxide emissions compared to traditional alternatives, there are some real concerns that the creation of a global bioeconomy could contribute to an accelerated rate of deforestation if not managed effectively. There are associated concerns over the impact on water supply and soil erosion. Hence, production of bioplastics from waste materials was found to be more prospective.Biodegradable plastics, like conventional plastics, offer a large range of packaging applications including bags for compost, agricultural foils, horticultures, nursery products, contact articles like disposable containers, toys, and textiles [10-16].

Research and development of bioplastic substances has shown their prospects to be used for medical, dental and pharmaceutical use. Biodegradable stitches, which do not require manual removal after healing, are regularly used to suture wounds and surgical incisions. Biodegradable bandages designed to promote clotting and proactive skin regeneration are also actively in use for traumatic wound care [17]. In the arena of single use health care products and packaging, reducing the environmental impact of medical waste while eliminating passive transfer of toxins into tissue can be significantly addressed via the replacement of artificial plastics with biodegradable plastics. Endoscopic (internal) drug delivery to inflamed areas and cancerous tumors via bioplastic carriers that degrade organically without a trace also offers huge promise (Figure 1a). Bioplastics with high tensile strength, including materials formulated with PHA, show great promise for orthopedic use. Though not a suitable substance for joint replacement, bioplastics may one day replace metals and artificial plastics for other orthopedic mechanisms, including the rods, screws and pins often used to assist repair of broken bones (Figure 1b). Cardiovascular uses of bioplastics where biodegradability is acceptable have demonstrated lower inflammation and healthier cell regeneration (Figure 1c). Though use of bioplastics to make endoscopic chemotherapeutic lines needs further testing, external infusion delivery systems made of bioplastics will boost healthy treatment and reduce waste (Figure 1d).

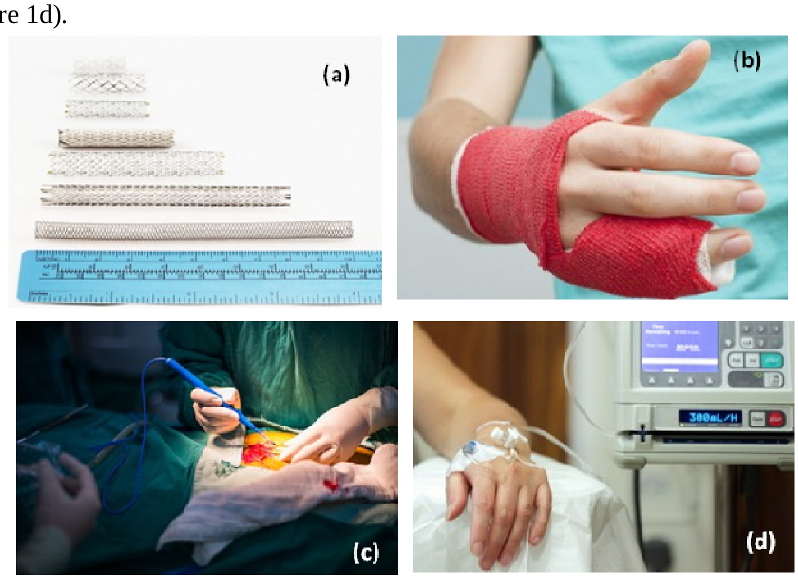

Figure 1. (a) Drug-eluting stents made of bioplastic as replacements for metal stents in cardiovascular and gastrointestial applications, (b) Rods, pins or screws and implements made of degradable bioplastics, (c) Tests of post-surgical patches, scaffolds and sutures made of bioplastics, (d) External infusion delivery systems made of bioplastics

Driven by the progress of 3D printing technology, printable thermoplastics have been developed to meet the wide range of possible applications [18]. Plant-based plastics are already a popular choice for 3D printing because they are much easier to work with during processing, eco-friendly and are food safe and odour free. Made from plant starches, Biome3D is a biodegradable plastic that combines easy processing and a superior print finish, while offering much higher print speeds [19].Further, two novel classes of biodegradable polymers, PolyActive and OctoDEX are drug delivery systems that are licensed out by OctoPlus. These biodegradable systems are used for the site-specific delivery of drugs. Because of their biodegradability and linear release properties, these two systems are an excellent technology for the controlled release of proteins and lipophilic small molecules for both local and systemic administration, and have applications in pharmaceuticals and medical technology [20]. Hence, plant based biodegradable plastic materials offer a wide scope of applications in the medical field as well.

II. MATERIALS AND METHODS

Various food wastes like banana peels, potatoes, agar agar, cornstarch, etc. were used to produce bioplastics using different procedures as follows:

A. *Bioplastic from banana peels*[21]

Banana peels were dipped in 0.5% sodium met bisulphite solution for 30 minutes after cleaning with distilled water and drying on filter paper. The treated peels were dried on filter paper and boiled for 30 minutes. The softened peels were pureed into fluid paste using a laboratory blender. To 25ml of banana paste 0.5M HCl,2ml of propan-1,2,3-triol and 3ml NaOH were sequentially added with intermittent mixing using a glass rod. The mixture was poured evenlyonto a large petridish and baked in hot air oven at 130°C for 30 minutes (Figure 2). Similar procedure was repeated using sorbitol instead of glycerol, skipping the process of boiling the treated peels.

Figure 2. Processing of banana peels to produce bioplastic, (a) sodium metabisulfite treatment, (b) boiling of treated peels, (c) softened peels drie after boiling, (d) grinding into fine paste, (e) even layering onto petri dish, (f) plastic after baking in oven.

B. *Bioplastic from potato starch* [22]

Potatoes were peeled and pureed using distilled water, and filtered with the help of muslin cloth. The starch was allowed to settle down for 5 minutes and then the supernatant was decanted. 100ml distilled water was added to rinse the starch. The water was decanted, this leaves clean wet starch. The starch was dried in an oven to obtain white powder (Figure 3). To 2.5g starch powder 25ml distilled water was added along with 3ml of HCl (0.1M) and 2ml of glycerol and this mixture was heated for 15 minutes. The pH was found to be acidic. To neutralize the pH, 3ml NaOH was added. The mixture was poured evenly onto a large petridish and and was baked in hot air oven for 30 minutes at 130^{o}C.

C. *Bioplastic from corn starch [23]*

Cornstarch solution was prepared by dissolving 10 grams of corn starch in 60 ml of distilled water. To this mixture 5 ml of acetic acid and 5 ml of glycerol was added sequentially and heated on low flame with constant stirring (Figure 4). The mixture was spread

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onto a largepetriplate and then baked in hot air oven set to 130° C for 90 minutes. Similar procedure was repeated with sorbitol in place of glycerol.

D. Bioplastic from agar [24]

Agar solution was prepared by adding 3.0 g agar in 100ml distilled water and 2ml glycerol was added to it. The mixture was heated on a low flame, poured evenly onto a petriplate and baked in hot air oven for 30 minutes at 130^{o}C.

E. Bioplastic from banana peels and agar

The banana peels were processed and made into paste as per earlier protocol. To about 25g of banana paste, 1 gram of agar was mixed followed by addition of HCl, Glycerol and NaOH. The steps of layering and baking were followed as per earlier protocol.

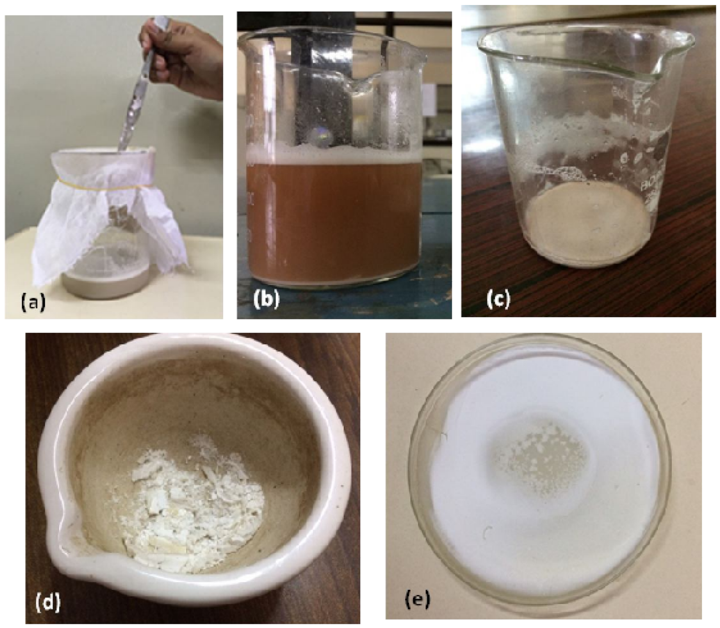

Figure 3. Processing of potato starch to produce bioplastic, (a) filtration of potato puree using muslin cloth, b) settling of potato starch, (c) settled starch after decantation, (d) mashing of heat dried starch, (e) even layering onto petridish.

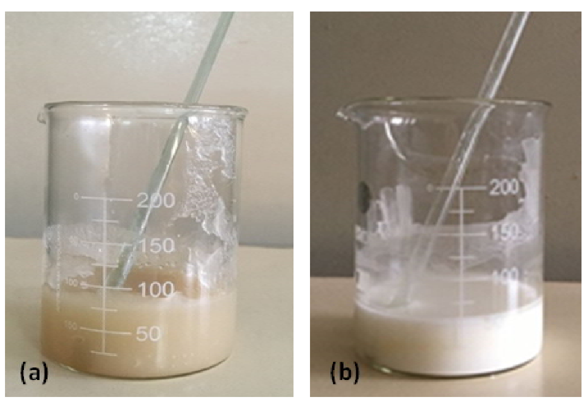

Figure 4. (a) Corn starch solution with glycerol, (b) Corn starch solution with sorbitol

Various physical & chemical properties of the bioplastic material produced were studied. The colour, texture, odour and transparency of the material were visually observed while the thickness of the plastic material was determined by using a micrometer (0-25μm, Mitutoy Company). The material was heated and microwaved to study its thermal characteristics. The shelf-life (decay) was assessed by visual inspection on a daily basis. Three best composites obtained(treated banana peel, untreated banana peel and agar and treated banana peel with sorbitol) were tested for tensile strength in Universal Testing Machine of DAK

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system. Five of the banana peel derived composites were selected and characteristics like water permeability & water-resistance were studied by sprinkling water on the sample and placing it in water for about 1 hour. The material was boiled in water to study the changes in structure and was also treated with cold water, acids(0.1N $H_2SO_4$and 0.1N HCl) & alkali (0.1NNaOH). Biodegradability tests were conducted on selected samples (banana peels treated with $Na_2S_2O_5$, untreated banana peel + agar and banana peel treated with $Na_2S_2O_5$ + sorbitol) by halophilicdegradability and soil biodegradability. Soil degradability was tested by layering 10 grams of moist soil over the composite and observation for degradation over a few weeks. For studying the degradation by halophilic microbes, 0.1 ml of the sample of sea water containing a mixture of halophilic microbes was plated and grown on halophilic agar medium. The composites were cut into pieces, dimensions were noted and were placed on the agar plate with halophilic microbes. The plates were checked regularly for a period of 15 days. All qualitative and quantitative data was recorded.

III. RESULTS AND DISCUSSION

The success of bioplastic from banana peels, agar agar, corn starch and potato starch is based on the fact that they are rich in starch. The films were prepared successfully by the mixing and casting method (Figure 5). A brown coloured elastic material with tensile strength similar to a paper (when manually pulled) was produced using banana peels. The bioplastic formed was comparatively fragile. The composite produced from banana peels was thin, papery and showed good tensile strength. The composite with potato starch was thin and transparent but brittle. Composite from corn starch was thick, flexible, opaque and hard. Agar gave the toughest material, which was like a transparent film and tensile strength was greater than other composites. The visual physical characteristics studied are presented in Table 1.

Thickness of the composites ranged approximately between 0.17-0.3μm (±0.4 std. error). The composites were found to be thermo labile (heat-sensitive) as most of them turned black n burnt when heated. They were found to be non-microwavable. Acid- base and water solubility tests conducted on five different banana peel derived composites gave variable results as shown in Figure 6. Almost all the composites were found to be insoluble in water, but a slight discolouration was observed in sample 4 (banana peel treated with $Na_2S_2O_5$ + potato starch) and sample 5 (untreated banana peel + sorbitol) with hot and cold water treatments. When the samples were treated with acids (0.1N H_2SO_4, and 0.1N HCl), the composites turned to a lighter shade of brown as compared to the controls and became moist. However they were insoluble in acids. Treatment with 0.1N NaOH showed uniform results for almost all the samples, as they turned into dark brown, softer materials with partial or no solubility in alkali.

The plastic material produced from banana peels was found to have a better shelf life as compared to other products. There was no decay observed even after 7 days. However after 15 days, the material started darkening which suggested initiation of decay.The composites were tested for soil biodegradability and degradation by halophilic microbes. Slight variations were seen in the dimensions after 15 days of soil degradability. The composites turned thinner and darker (Figure 7a-d). In the studies of biodegradability using halophilic microbes, there was no visible change in the dimensions, but a growth of halophilic microbes could be seen on the composite with gradual darkening (Figure 7e-h).

The composites which were tested for tensile strength in Universal Testing Machine of DAK system showed reasonably good tensile strength. Tensile strength of approximately 0.443 Mpa (0.22mm thick), 1.098 Mpa (0.15mm thick) and 1.596 Mpa (0.37mm thick)were observed for banana peels treated with $Na_2S_2O_5$, untreated banana peel + agar and banana peel treated with $Na_2S_2O_5$ + sorbitol composites. The replacement of glycerol with sorbitol showed promising prospects in increasing the tensile strength of the plastic. Attempt was made to increase the tensile strength of the product by addition of different plasticizers similar to glycerol and manipulation of the concentrations of HCl and NaOH. The maximum load endured by the samples (banana peels treated with $Na_2S_2O_5$, untreated banana peel + agar and banana peel treated with $Na_2S_2O_5$ + sorbitol) were found to be 1.500 N, 3.000 N and 8.600 N respectively. The replacement of glycerol with sorbitol showed promising prospects in increasing the tensile strength of the plastic. The maximum load endured by the sample of banana peel + sorbitol composite was found to be substantially higher than the samples in literature [21] which would yield a stable bioplastic. The bioplastics with lesser tensile strength and load could be useful in suture preparation in medical field and those with higher strengths could be used to replace the polythene bags.

The manufacture of bioplastic from food waste is most desirable as it is a low-cost and high-value usable product. Adding other materials / chemicals to food wastes during processing enhanced the mechanical properties of the bioplastic. All the materials produced were water-soluble, hence, coating with oil or adding non toxic chemicals is suggested to favour water-proofing ability. Different types of raw materials were combined to form materials with improved properties like increased thickness, plasticity and enhanced waterproofing. As per similar reports, in order to improve the tensile properties of the starch based materials, chemically modified hydroxy-propylated or oxidized potato starch as well as high amylose starch were developed by genetic engineering and

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have been used as raw material for plastics [4, 25-28]. Tensile properties of the modified starches as compared to the native starch based materials were found to be improved. Another recommendation for the future is to use sodium bicarbonate ($NaHCO_3$), instead of sodium metabisulfite ($Na_2S_2O_5$), as a preservative for the plastic after manufacturing because it has higher efficiency and the plastic would bear higher temperatures and pressures without decaying.

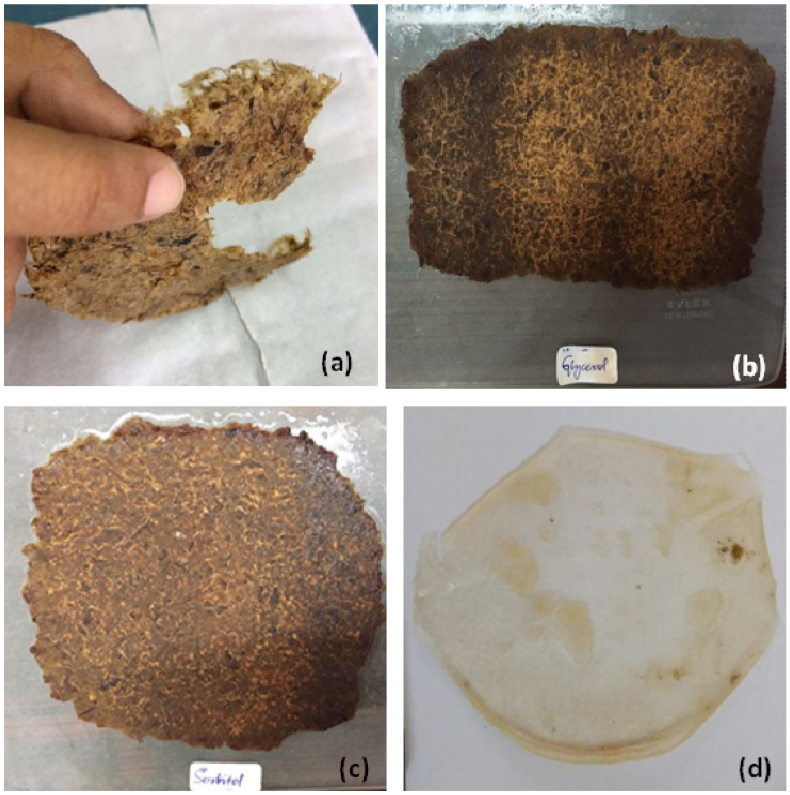

Figure 5. Bioplastic materials prepared from various food wastes,(a) banana peels, (b) banana peels + glycerol, (c) banana peels + sorbitol, (d) corn starch.

Table 1. Physical characteristics of the bioplastics produced.

Material used	Banana peel + glycerol	Banana peel + sorbitol	Corn starch	Potato starch	Agar
Colour	Brown	Brown	White	White	Transparent
Odour	Fruity	Fruity	Nil	Nil	Nil
Texture	Rough	Rough	Smooth	Smooth	Smooth
Transparency	Opaque	Opaque	Translucent	Translucent	Transparent

Finally, this research work can be scaled-up by manufacturing the plastic in a bigger scale at a factory that includes all of the stages of the manufacturing. Biodegradable plastics made from food wastes offer wide applications in various fields.Potato starch has shown suitable properties for film formation using extrusion and film blowing techniques, which have great potential for packaging applications [4, 29, 30]. Plant-based bioplastic products can be used to assist the body's natural ability to regenerate healthy cells during healing also include bandages, sutures, and gingival patches. Additionally, because some wounds and incisions take longer to heal than others, biodegradable sutures can be constructed to degrade according to a specific time-frame for a specific level of healing. Biodegradable staples, plastic pins, tacks, and screws are used to hold shattered bones together while they heal, to reattach ligaments, and for delicate reconstructive surgery on ankles, knees, and hands. Biodegradable dental implants, made of porous polymer particles, are being used to quickly fill the hole after a tooth has been extracted [31]. Hence, the bioplastic materials

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produced from various food wastes in our studies offer wide scope and potentials, depending on their physical and chemical properties and further characterization.

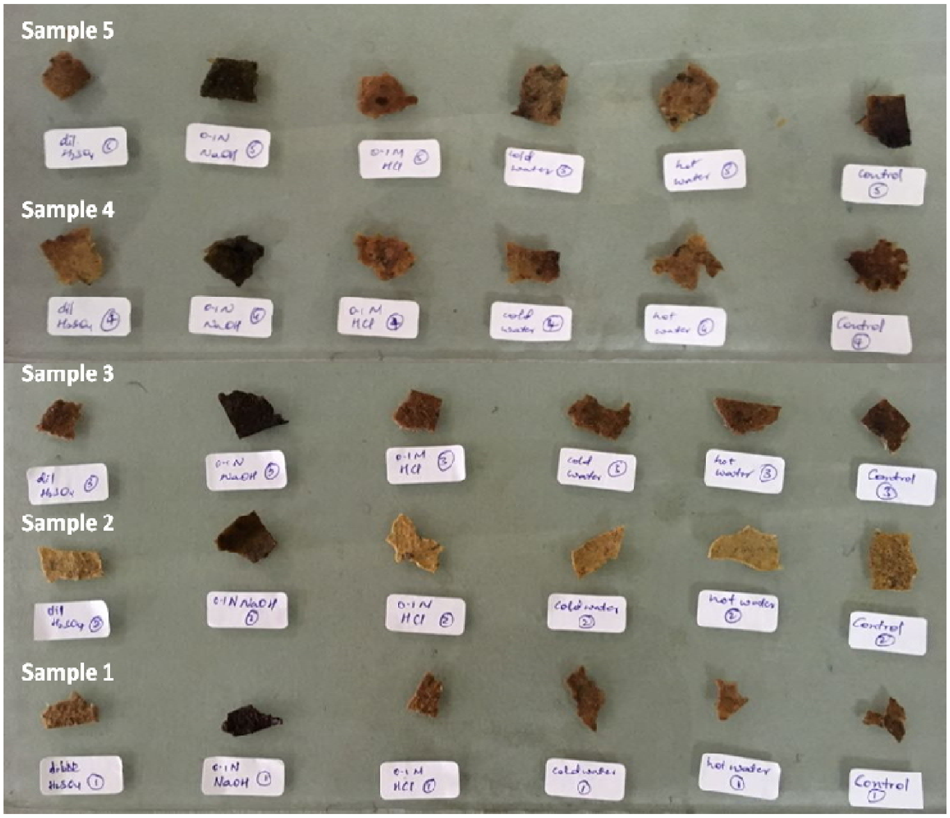

Figure 6. Acid-base and water solubility results of various banana peel derived composites; Sample 1: Banana peel treated with $Na_2S_2O_5$ + Sorbitol, Sample 2: Banana peel treated with $Na_2S_2O_5$ + Glycerol, Sample 3: Banana peel treated with $Na_2S_2O_5$ + Glycerol + Agar, Sample 4: Banana peel treated with $Na_2S_2O_5$ + Potato starch, Sample 5: Untreated Banana peel + Sorbitol

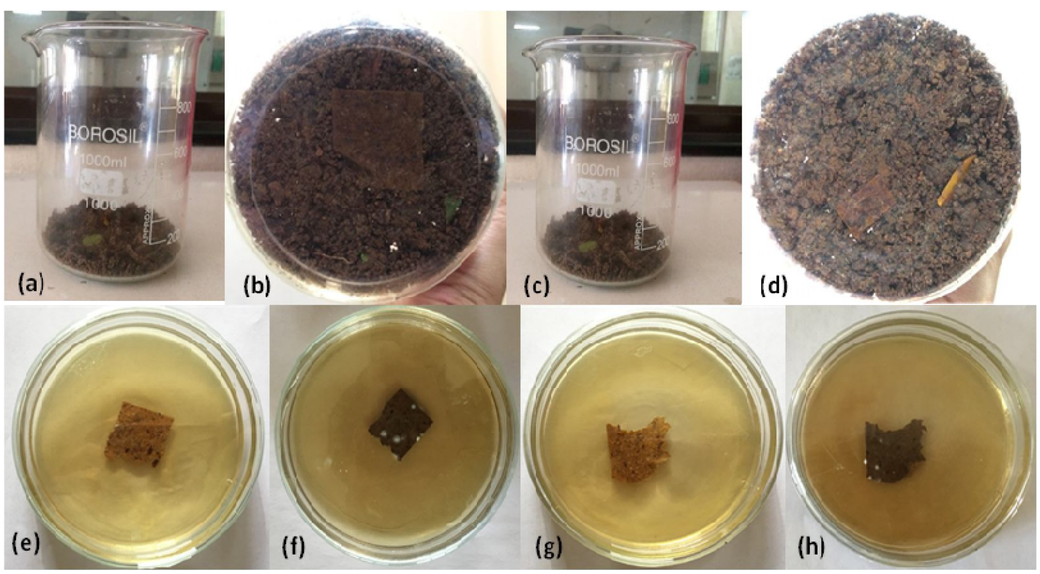

Figure 7.Biodegradability tests of bioplastics produced. (a-d) Soil biodegradability of (a) test sample 1 in beaker, (b) bottom view of test sample 1, (c) test sample 2 in beaker,(d) bottom view of test sample 2. (e-h) Biodegradability with halophilicmicrobes of (e) test sample 1 as observed on Day 1, (f) test sample 1 as observed on Day 15, (g) test sample 2 as observed on Day1, (h) test sample 2 as observed on Day 15.

IV. CONCLUSION

Thin layered biodegradable plastic materials were successfully derived from various food wastes. The physical and chemical characteristics of the composites produced indicate their prospective uses as replacement for petroleum-based non-biodegradable plastics which are causing environmental hazards. The films produced from banana peels had potential application to be used as food packaging because it can enhance the food quality and at the same time can protect the environment. The hard material produced from cornstarch can be used as container. Water solubility is a useful property as the material may be useful for drug delivery systems, provided, the material can be sterilized. More applications of bioplastics to replace polythene bags can be ensured by imparting waterproofing ability and increased tensile strength. Since these bioplastics are made out of natural wastes, their applications in medical field is very prospective as they may tend to cause less allergies as compared to chemical based plastics. These kind of bioplastics may also find use as the basis for sanitary products like diaper foils, bed underlay, disposable gloves etc., as they are breathable & allow water vapour to permeate, but at the same time waterproof. If the bioplastic is fragile (as in our case), it may be dried, powdered & coloured to be used for 3D printing technology. The 3D prints prepared can later be successfully decomposed into environment without causing pollution. Proactive skin regeneration using potato skin is prevalent in traumatic wound care. Bioplastics made out of potato skin may find use in skin grafting and artificial skin transplantation techniques. Future research on the compatibility and biodegradability of these materials opens avenues for using these plastics as drug delivery systems, biodegradable bandages and suture threads, 3D printing and medical devices.

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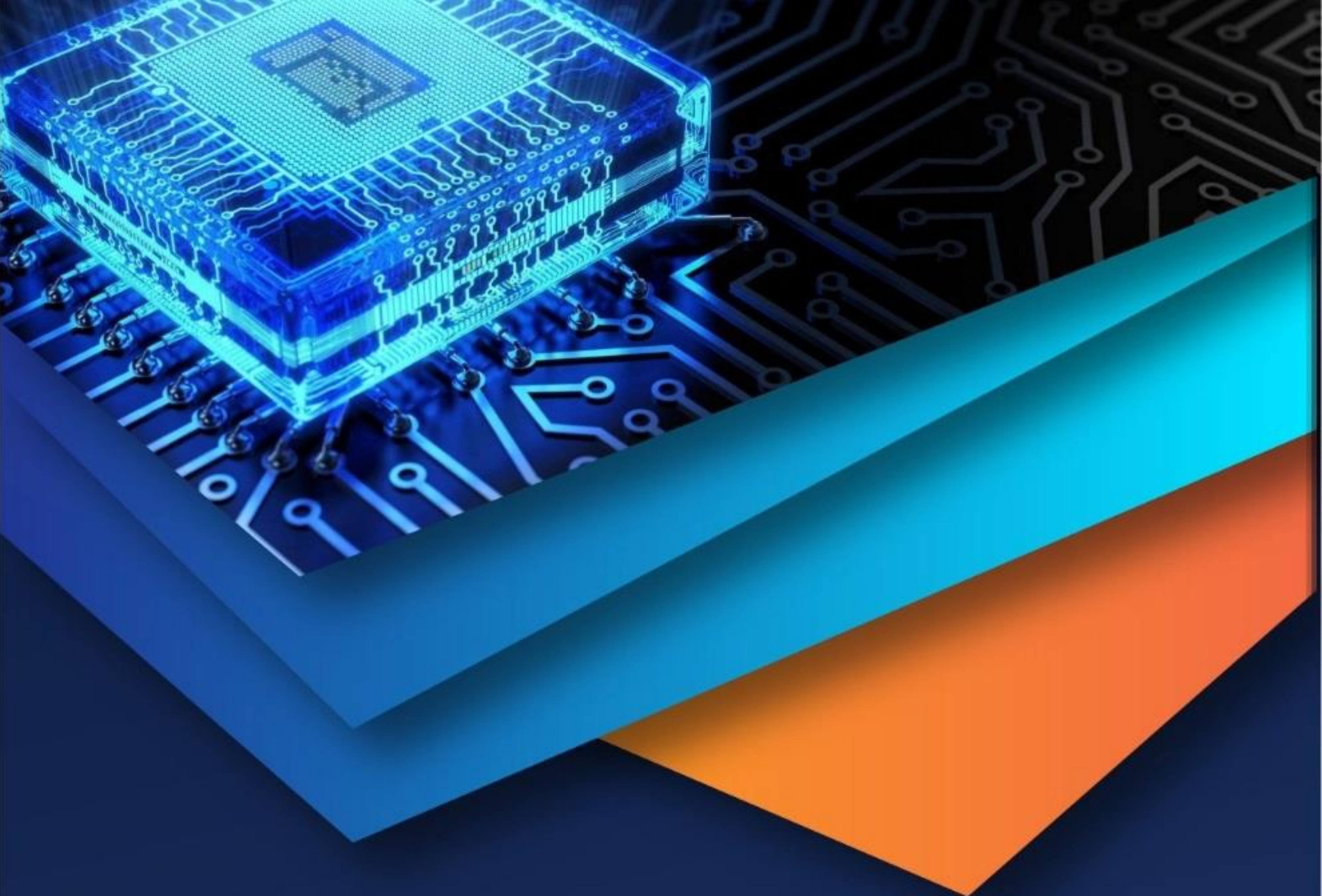

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