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Utilization of food wastes to prepare film forming biomolecules

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Abstract
Food wastes are produced by a variety of sources, ranging from agricultural operations to household consumption. About 38% (approx.) of waste occurs during food processing and disposal of these food wastes represents a huge challenge as an environmental issue. Apart from being used as animal feeds or fertilizers, in the recent year's research clearly showed that the by- products from food processing industries contain valuable nutrients. These valuable by-products can be used in the development and production of new biomolecules. There has been a constant interest in finding new sources of plant- derived polysaccharides from the bio agro- waste streams as they are a promising source. In this study, cellulose was extracted from food wastes (Potato peels and Rice bran) and the extracted biomolecule was converted into Hydroxypropyl methylcellulose (HPMC). HPMC is a semi-synthetic polymer which possesses good film-forming characteristics and it is widely used in the capsule manufacturing process by pharmaceutical industries. In this study, the food waste was utilized efficiently for the development of valuable raw material HPMC from a natural plant source for usage in the food and pharmaceutical industries world-wide.

Keywords: cellulose, HPMC, film, food wastes, potato peels and rice bran

1. Introduction

Tremendous amounts of municipal solid waste (MSW) are being generated and dumped off at landfills every day. Among these one of the largest single category of wastes disposed are represented by biodegradable food waste which resulted in several environmental and economic issues over the past few years. Food waste, constituting the largest MSW category (38%) are being sent to the landfills as much as 3,648 tons per day (TPD) [1, 2]. Generally, the wastes generated from the food industry can be separated into two main categories: plant- derived wastes and animal- derived wastes. The animal- derived wastes can be subdivided into three subcategories: (i) meat products, (ii) fish and seafood and (iii) dairy products, whereas the plant- derived wastes can be classified into four subcategories: (i) cereals (e.g. rice bran, wheat bran and brewers' spent grain), (ii) root and tubers (e.g. potato peel, sugar beet and molasses), (iii) oil crops and pulses (e.g. sunflower seeds, soybean seed and olive pomace) and (iv) fruit and vegetables (e.g. orange peel, grape pomace, apple pomace, tomato skin and pomace) [3, 4].

Disposal of huge quantities of waste have always been a special concern to the food industries. Apart from being used as animal feeds or fertilizers, the research conducted in the past few years indicated that there are valuable molecules which can be obtained from the by-products and wastes generated from food industries. These biomolecules can be converted into value added biomolecules and can be used for various purposes [5]. Regarding the potentially marketable components present in foods wastes and co-products, the aim of the study was to exploit high-value components such as proteins, polysaccharides, fibers, flavor compounds, and phytochemicals as nutritionally and pharmacologically- functional ingredients [6]. Nearly 90% of dry matter in vegetables' and fruits' is composed of carbohydrate out of which 75% is sugars and hemicellulose, 9% is cellulose and 5% is lignin; whilst, the major plant biopolymers with largest application possibilities are cellulose and starch [7, 8].Thus, the main focus of the feasibility study was primarily focused on starch and cellulose.

Cellulose being the most abundant natural biopolymer on earth, is considered as one of the most promising polymeric resources. Advantages of using cellulose include low cost, biocompatibility and biodegradability. It has a wide range of application such as clothing, packaging, paper, and medical products in our everyday life, but also has a potential usage in numerous applications such as bio-based materials, fibers, films, food casings and membranes. Therefore, effective utilization of cellulose from food waste will help in the reduction of exploiting of our limited fossil resources so as to protect the environment and sustainable resources for the future generations. In recent years, considerable attention has been directed towards biodegradable cellulose-based materials, due to the serious "white pollution" made from the non-biodegradable plastic film [9].

Chemically modified polymers have been extensively investigated in order to develop new biomaterials with different physio-chemical properties. Important class of modified polymers includes cellulose ethers, such as methylcellulose (MC), hydroxypropyl methylcellulose (HPMC), hydroxyethyl cellulose (HEC) and carboxymethylcellulose (CMC). Cellulose, the most abundant polysaccharide found in nature; it is a regular and linear polymer composed of (1→4) linked β-D-glucopyranosyl units. This particular β-(1→4) configuration together with intramolecular hydrogen bonds gives a rigid structure [10]. Among the class of modified polymers, Hydroxypropyl methylcellulose (HPMC) [11, 12] is one of the most general hydrophilic polymers, which are being extensively used in many fields owing to its ease of use, excellent film-forming ability, good biocompatibility and biodegradability [13].Starch and HPMC are good candidates for making hard and soft gelatin capsules [14]. One of the limitations of using them is the initial high capital investment [15]. However, potato peels [16] and rice brans are the most common food wastes and agricultural waste all around the world. These two sources would contribute to the efficient utilization of food wastes with the additional benefit of developing the raw materials in an inexpensive way. Major utilization of HPMC in the production of capsule shells would be able to replace the animal-derived gelatin in conventional two-piece capsules. The main aim of this study is to recover cellulose from food wastes and to check its suitability for vegetable capsules by preparing HPMC films.

2. Materials and methods

Materials

The Rice Bran and Potato were purchased from Nilgiris, Thiruvanmiyur, Chennai. Schultz regent, NaOH (Sodium hydroxide), Benedict's reagent, Copper sulphate, Propylene oxide, Ninhydrin reagent, Sulphuric acid and Concentrated Hydrochloric acid were purchased from Fisher Scientific. Sodium hypochlorite was purchased from Jeyam Scientific for the experimental study.

Extraction of cellulose from rice bran

The rice bran was thoroughly ground using a mixer. The bran was then passed through 50mm mesh and weighed. The samples were stored at room temperature (25°C) for further analysis. The stored samples were washed thoroughly with distilled water and dried at 100°C. Further, roughly 100 grams of rice was treated with 100 ml of 10% aqueous Sodium hydroxide (NaOH) solution. 900 ml distilled water was added to the samples and was subjected to stirring for 30 minutes on a magnetic stirrer at constant temperature of 60°C. The obtained slurry was filtered and filtrate (which consists of cellulose) was washed with distilled water. The solution was neutralized by adding 1N Hydrochloric acid (HCl) drop by drop. The obtained solution was spin cast at 500 rpm and was allowed to dry in a vacuum oven at 60°C [17]. The extracted and dried cellulose was stored in a desiccator for further analysis.

Extraction of cellulose from potato peels

The potatoes were hand peeled and the peels were washed thoroughly with water to clear away soil and other bulk contaminants from the peels. The cleaned peels were added to water at a water-to-pulp ratio of 20:1 and was made into a slurry using a blender for 10 mins to remove the majority of potato flesh. The resulting slurry was screened using a 250 mm sieve and repeatedly washed with distilled water. Potato peels comprised of starch, cellulose, hemicellulose, lignin, and other impurities, and therefore several pre-treatment steps including alkali treatments and chlorine bleaching were required to isolate its cellulosic component prior to acid hydrolysis. Initially, the peels were treated with a 0.5 N aqueous sodium hydroxide solution at 80°C for 2.5 h under mechanical agitation. The treatment was repeated thrice in order to completely eliminate the lignin, hemicelluloses, and other impurities. After each treatment, the pulp was filtered using a 75 mm sieve to remove the alkali solution and washed with distilled water to remove dissolved impurities. The alkali washing was followed by bleaching with 2.3% sodium chlorite solution in an acetate buffer (pH = 4.9) to remove any organic residue still present in the sample. The bleaching treatment was carried out twice at 70°C for 2 h. The extracted cellulose fibers from the potato were washed, dried and weighed [18, 19]. The cellulose extracted was stored in a desiccator for future analysis.

Conversion of cellulose to HPMC

The cellulose obtained from the potato peels and rice bran were first converted to methyl cellulose by an alkylation reaction using Methyl chloride. 74 g of cellulose from the potato peels and rice bran was reacted with methyl chloride to obtain 73 g of methylcellulose on heating. The methylcellulose is then reacted with propylene oxide to form (49g) Hydroxypropyl methyl cellulose [20]. Several detection and identification tests were done to assess the characteristics and film forming ability of HPMC.

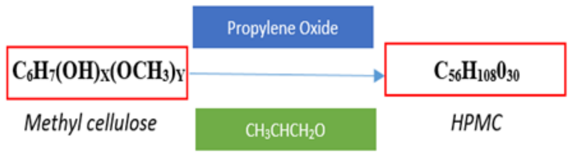

Fig 1: Conversion of methyl cellulose to HPMC

Preparation of HPMC Film

Film formation is the most important characteristic of HPMC for it to be used as capsule shell in oral medications. 2% (2 g in 100ml), 6% (6g in 100ml) and 10% (10g in 100 ml) HPMC solution were prepared by boiling water and slowly adding HPMC powder with constant stirring. The solution was then allowed to cool at room temperature. It was then poured into glass petri-dishes and dried at room temperature.

4. Methodology analysis and results

Detection Tests

The following detection tests were done to check the presence of HPMC and ensure the absence of major sugars, proteins and fats from the food waste samples. The results and observations are summarized in table 3.

Benedict's test

1 ml of filtrate solution was prepared by dissolving 0.5 grams of sample in 1ml of distilled water. To the sample solution Benedict's reagent was added and heated gently to test the presence of carbohydrates particularly reducing sugars. The solution showed no appearance of reddish precipitate indicating the absence of reducing sugars [12] in the sample.

Biuret test

0.5 mg of the extract residue was mixed with 1ml of water and additionally 1mL of 40% sodium hydroxide solution was added [22] in the mixture. To the mixture, a drop of 1% copper sulphate was added to test the presence of protein in the sample. The samples were clear and there was no formation of violet or pink color indicating the absence of protein in the sample.

Spot test
0.5g of sample was pressed or rubbed on a clean white sheet of paper to test for the presence of fat. There was no appearance of a greasy spot (translucent spot) indicating the absence of fat in the sample [23]. Negative results indicate the absence of proteins, fats and carbohydrates and in turn indicate that the sample comprises of HPMC.

Iodine test
Few drops of iodine solution were added to 1ml of sample solution (0.5g) to detect the presence of starch in the sample. There was no formation of blue-black color indicating the absence of starch in the sample.

Schultz test
A Few drops of Schultz Reagent was added to the 0.5g of sample in 1ml of water to test for the presence of cellulose. There was a noticeable appearance of purple color solution which indicated the presence of Cellulose in the sample.

Alcohol-precipitate test for Methyl cellulose
0.5g of the cellulose sample after treatment with methanol to methyl cellulose is mixed with 5 ml of distilled water and mixed with 25 ml of 95% alcohol solution. To this mixture, 2 to 3 drops of Saturated NaCl solution was added to test the presence of Methyl cellulose. The absence of precipitate formation indicated the presence of Methyl Cellulose in the sample.

Characterization studies For HPMC [24]
The following characterization tests were done to characterize the synthesized HPMC from the food waste and the results and observations are summarized in table 4.

pH Test
1% solution of synthesized HPMC from rice bran and potato peels was prepared and checked for the pH. The pH was found to be 7.1 which is in accordance with the literature review for HPMC.

Aggregate test
The 0.5 grams of sample was gently added to the top of 100ml beaker filled with distilled water. It was allowed to disperse over the surface and container was held constant to ensure an even dispersion of the substance. It was then allowed to stand for 1-2 min to check for aggregate formation. The presence of powdered aggregate on the surface indicated that the sample synthesized was HPMC.

Viscosity test
0.5g of the sample was added to 100ml of boiling water and the same was continuously stirred using magnetic stirrer to form a slurry. It was then cooled to 10°C and then checked for the appearance of the solution with respect to viscosity thickness. The solution resulted in a clear and slightly turbid solution with thickness dependent on viscosity grade which indicated that the sample was HPMC.

Ninhydrin test
To 1ml of the sample solution, 9ml solution of sulphuric acid and water in the ratio of 9:10 was added and shaken well. The solution was further heated in a water bath for exactly 3 minutes and immediately cooled in an ice bath. Following this, 0.6 ml of ninhydrin TS was added, and the solution was vigorously shaken and allowed to cool down to room temperature. The color change of the solution from red to purple indicated that the solution contains HPMC.

Film test
2-3 ml of the sample from aggregate test was spread into a glass slide as a thin film and the water was allowed to evaporate. The formation of clear and coherent film indicated that the sample was HPMC.

Flocculation test
The sample solution prepared from the aggregate test was added to exactly 50 ml of water in a beaker. A thermometer was inserted into the solution and was continuously stirred using a magnetic stirrer and simultaneously heated on the hot plate at a rate of 2-5 °C/min. The temperature at which the turbidity increase began was noted as 80°C and this temperature was called the flocculation temperature. According to literature, the flocculation temperature for HPMC should be higher than 50°C and the experimental temperature agrees well with the range.

4. Discussion
Cellulose was successfully extracted from both rice bran and potato peels and the amount of cellulose obtained from the food waste was calculated as 54g and 25g respectively. The appearance of cellulose was found to be slightly crystalline and watery. The amount of cellulose yield from rice bran (54%) is found to be more when compared to amount obtained from the potato peels (27.7%). The yield of cellulose extracted from both the sources is summarized in Table No. 1& 2. The various confirmatory and detection tests were used to detect the presence of other components like carbohydrates, proteins, amino acids and fats and the negative results of the tests indicated the complete purification of the sample to HPMC. Other tests for hemicelluloses and lignin content were not detected as those constituents were removed during the extraction procedure which was already discussed in research papers. The characterization tests were also positive and proved that the HPMC synthesized was reliable for making capsules. The 2% solution of the prepared HPMC powder was found to be too dilute (watery) and films prepared using 6% and 10% solutions were reliable and good for making capsules. Thus, the HPMC powder synthesized from the two sources namely rice bran and potato peels exhibited sufficient film forming property and can be used for making plant derived capsules for oral medications.

Table 1: Yield of Cellulose from Rice Bran

Content	Amount
Weight of the sample before drying	200 g
Weight of the sample after drying	100g
Moisture content in the sample (Initial-Final)	100g
Percentage Moisture content(Initial – Final/Initial)*100	50%
Weight of cellulose obtained from Rice Bran	54g

Table 2: Yield of Cellulose from Potato Peels

Content	Amount
Weight of the sample before drying	200 g
Weight of the sample after drying	90g
Moisture content in the sample (Initial-Final)	110g
Percentage Moisture content (Initial –Final/Initial)*100	55%
Weight of cellulose obtained from Rice Bran	25g

Table 3: Summary of results for the detection tests

Tests	Procedure	Observation	Results
Benedict's test	1 ml of filtrate solution was prepared by dissolving 0.5 grams of sample in 1ml of distilled water. To the filtrate solution Benedict's reagent was added and heated gently to test the presence of carbohydrates particularly reducing sugars	No reddish-brown precipitate	Absence of reducing sugar
Biuret test	0.5 mg of the extract residue was mixed with 1ml of water and additionally 1mL of 40% sodium hydroxide solution was added [22] in the mixture. To the mixture, a drop of 1% copper sulphate was added to test the presence of protein in the sample	No violet or pink color	Absence of proteins
Spot test	0.5g of sample was pressed or rubbed on a clean white sheet of paper to test for the presence of fat.	No greasy spot	Absence of fat
Schultz test	A Few drops of Schultz Reagent was added to the 0.5 grams of sample in 1ml of water to test for the presence of cellulose.	Appearance of purple color solution	Presence of cellulose
Iodine test	A few drops of Iodine was added to the sample, the appearance of blue black color indicates the presence of starch.	No blue-black color	Absence of starch
Sodium hydroxide test	0.5g of the cellulose sample after treatment with methanol to methyl cellulose is mixed with 5 ml of distilled water and mixed with 25 ml of 95% alcohol solution. To this mixture, 2 to 3 drops of Saturated NaCl solution was added to test the presence of Methyl cellulose.	Absence of precipitate	Presence of methyl cellulose

Table 4: Characterization tests for HPMC

Tests	Observation	Results
pH test	The pH of the solution was found to be 7.1	Presence of HPMC
Aggregate formation	Formation of powdered aggregate on the surface	Presence of HPMC
Viscosity test	The solution was found to be more viscous and the viscosity was found to be 110cp (2% solution, 20°C)	Indicates that the solution contains HPMC
Ninhydrin	The color of the solution changes from red to purple	Indicates that the solution is HPMC
Film formation	A thin transparent and clear film was formed	Indicates the film formation ability of HPMC
Flocculation temperature	The flocculation temperature was found to be 80°C (>50°C)	Presence of cellulose was confirmed

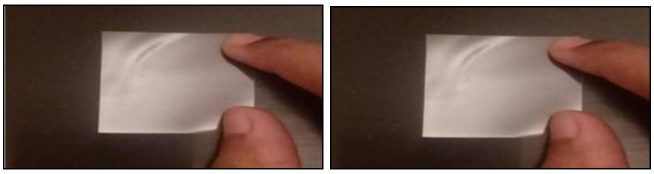

Fig 2: HPMC film obtained from 10% and 6% solutions that were prepared from the HPMC powder

5. Conclusion

The overall waste production of potato peels mainly in the fried snack products and rice bran which is also used for the extraction of oil can be utilized for an alternative usage from this study. This alternative method for the production of HPMC will reduce the waste disposal on the whole and serve as a method to benefit from the WASTE generated in a food Industry. Thus, the efficient utilization of the by-products from food industry can help in reducing the negative cost, reduce environmental pollution, demonstrating sustainability in food industry and that has direct impact on the economy of the country. Further studies can be conducted on how the drug delivery differs from a normal gelatin capsule versus the plant derived HPMC capsules from this study.

6.Acknowledgement

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