Title: FLASH-GEL METHOD FOR FORMING BIODEGRADABLE OR COMPOSTABLE CONTAINERS

Abstract: The present invention provides an improved method and materials for forming biodegradable containers with increased flexibility and strength that can hold food products in dry, damp or wet conditions and provide biodegradable containers prepared according the disclosed process. The containers are produced through the use of a pre-gelled starch suspension that is unique in its ability to form hydrated gels and to maintain this gel structure in the presence of many other types of materials at low temperatures.

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FLASH-GEL METHOD FOR FORMING BIODEGRADABLE OR COMPOSTABLE CONTAINERS

CROSS-REFERENCE TO RELATED APPLICATIONS

This application claims the benefit of U.S. Provisional Application No. 61/029,152 filed February 15, 2008, and U.S. Provisional Application No. 61/084,545 filed July 29, 2008.

FIELD OF THE INVENTION

The invention relates to improved methods and materials for forming biodegradable containers that can hold food products in dry, damp, or wet conditions.

BACKGROUND OF THE INVENTION

Materials such as paper, paperboard, plastic, polystyrene, and even metals are presently used in enormous quantity in the manufacture of articles such as containers, separators, dividers, lids, tops, cans, and other packaging materials. Modern processing and packaging technology allows a wide range of liquid and solid goods to be stored, packaged, and shipped in packaging materials while being protected from harmful elements, such as gases, moisture, light, microorganisms, vermin, physical shock, crushing forces, vibration, leaking, or spilling. Many of these materials are characterized as being disposable, but actually have little, if any, functional biodegradability. For many of these products, the time for degradation in the environment can span decades or even centuries.

Each year, over 100 billion aluminum cans, billions of glass bottles, and thousands of tons of paper and plastic are used in storing and dispensing soft drinks, juices, processed foods, grains, beer and other products. In the United States alone, approximately 5.5 million tons of paper are consumed each year in packaging materials, which represents only about 15% of the total annual domestic paper production.
Packaging materials (e.g., paper, paperboard, plastic, polystyrene, glass, or metal) are all, to varying extents, damaging to the environment. For example, the manufacture of polystyrene products involves the use of a variety of hazardous chemicals and starting materials, such as benzene (a known mutagen and a probable carcinogen). Chlorofluorocarbons (or "CFCs") have also been used in the manufacture of "blown" or "expanded" polystyrene products. CFCs have been linked to the destruction of the ozone layer.

Due to widespread environmental concerns, there has been significant pressure on companies to discontinue the use of polystyrene products in favor of more environmentally safe materials. Some groups have favored the use of products such as paper or other products made from wood pulp. However, there remain drawbacks to the sole use of paper due to the tremendous amount of energy that is required to produce it. A strong need to find new, easily degradable materials that meet necessary performance standards remains.

Degradability is a relative term. Some products which appear to be degraded merely break apart into very small pieces. These pieces are hard to see, but can still take decades or centuries to actually break down. Other products are made from materials which undergo a more rapid breakdown than non-biodegradable products. If the speed of this degradation is such that the product will degrade within a period of less than approximately 180 days under normal environmental conditions, the product is said to be compostable. Achievement of products made of compostable materials which also meet a variety of needs, such as containers for products in a damp or wet condition, has posed a significant challenge.

One solution has been to make packaging materials out of baked, edible sheets, e.g., waffles or pancakes made from a mixture of water, flour and a rising agent. Although edible sheets can be made into trays, cones, and cups which are easily decomposed, they pose a number of limitations. For example, since fats or oils are added to the mixture to permit removal of the sheet from the baking mold, oxidation of these fats cause the edible sheets to go rancid. In general, edible sheets are very brittle and far too fragile to replace most articles made from conventional materials. They are also overly sensitive to moisture and can easily mold or decompose prior to or during their intended use.
Starch is a plentiful, inexpensive and renewable material that is found in a large variety of plant sources, such as grains, tubers, and fruits. In many cases, starch is discarded as an unwanted byproduct of food processing. Starch is readily biodegradable and does not persist in the environment for a significant period after disposal. Starch is also a nutrient, which facilitates its breakdown and elimination from the environment.

Due to the biodegradable nature of starch, there have been many attempts to incorporate it into a variety of materials. Starch has been incorporated into multi-component compositions in various forms, including as filler and binder, as has been used as a constituent within thermoplastic polymer blends.

Starch can be used as a binder or glue to adhere solid constituents together to form a heterogenous mixture of different components. At some point before or during the molding phase, the starch is typically dissolved or gelatinized in an appropriate solvent, such as water, so that the starch becomes a flowable material into which the other components can be dispersed. Since native starch has a melting point that approaches its decomposition temperature, it is necessary to add polar liquids or solvents to allow the starch to become molten, solvated or otherwise liquefied into a plastic state at a temperature that is safely below its decomposition temperature. Upon resolidification of the gelatinized starch, typically by removing enough of the water by evaporation so that the starch recrystallizes or otherwise dries out, the starch forms a solid or semi-solid binding matrix that can bind the remaining components together. Although many have attempted for years to perfect a starch blend that would yield an environmentally sound material while, at the same time, being economical to make, such a combination has not yet been achieved.

There remains a need in the art to provide a fully compostable product that is strong, not prone to mold or pests, and can be readily and inexpensively made. Furthermore, there is a need to develop a robust method to develop compostable products that can be used to hold dry, wet or damp material at a range of temperatures.

U.S. Patent Nos. 5,736,209 and 5,810,961, and PCT Publication No. WO 97/37842, also assigned to Khashoggi Industries, disclose methods to develop biodegradable paper and products which include a binding matrix of starch and cellulosic ether, and fibers substantially
homogeneously dispersed throughout the matrix. The ‘209 patent discloses a concentration range for the starch of about 5% to about 90% by weight of solids in the sheet, for the cellullosic ether a range from about 0.5% to about 10% by weight of solids, and for fibers a concentration range from about 3% to about 40%. Optionally, an inorganic mineral filler can be added. Sheets produced using this biodegradable material having a thickness less than about 1 cm and a density greater than about 0.5 g/cm³ are described.

PCT Publication No. WO 01/51557, also filed by Khashoggi, is directed to compositions and methods for manufacturing thermoplastic starch compositions having a particulate filler (present in an amount greater than about 15% by weight of the thermoplastic starch) and, optionally, fiber reinforcement. Native starch granules are made thermoplastic by mixing and heating in the presence of an appropriate plasticizer (including somewhat polar solvents such as water or glycerin) to form a starch melt. The starch melt is then blended with one or more non-starch materials in order to improve the properties and reduce the cost of the resulting thermoplastic starch composition. A particulate filler component is thereafter blended with the starch melt, preferably an inexpensive, naturally occurring mineral particulate filler ("inorganic filler"), included in an amount greater than about 15% by weight of the thermoplastic starch composition. In addition, this reference discloses a composition comprising a thermoplastic starch melt having a water content of less than about 5% by weight while in a melted state, wherein at least one plasticizer has a vapor pressure of less than about 1 bar when in a melted state and in which a solid particulate filler phase is dispersed and included in an amount from about 5% to about 95% by weight. An additional embodiment discloses dispersion of a solid particulate filler phase in an amount from about 5% to about 95% by weight of the thermoplastic starch composition and a fibrous phase in a concentration of from 3% to about 70% by weight.

U.S. Patent No. 5,618,341, 5,683,772, 5,709,827, and 5,679,145 and PCT publication No. WO 97/2333, assigned to Khashoggi Industries, disclose starch-based compositions that can be used in making containers. U.S. ’341 and ’145 teach methods for dispersing fibers within a fibrous composition comprising the steps of: (a) combining together water, fibers, and a thickening agent such that the thickening agent (such as a pregelatinized starch) and water interact together to form a fluid fraction that is characterized by a yield stress and
viscosity that enables the fibers to be substantially uniformly dispersed throughout the fibrous composition as the fibers and fluid fraction are mixed together, the fibers having an average length greater than about 2 mm and an average aspect ratio greater than about 25:1; and (b) mixing together the combined thickening agent, water, and fibers in order to substantially uniformly disperse the fibers throughout the fibrous composition. The thickening agent is included in an amount in a range from about 5% to about 40% by weight of the fluid fraction. The inventive method involves a fluid system that is able to impart shear from a mechanical mixing apparatus down to the fiber level in order to obtain a starch-based composition having substantially uniformly dispersed fibers. U.S. Patent '772 additionally discloses an inorganic filler to enhance the strength and flexibility of the articles. '827 additionally discloses methods to make the article of manufacture that is developed from mixtures including fibers having an average aspect ratio greater than about 25:1. The '341, '772, '827, and '145 patents and WO 97/2333 application disclose high aspect ratios (i.e., about 25:1 or greater) and long-length (i.e., at least about 2 mm) fibers to reinforce the structure. PCT publication No. WO 97/2333 discloses articles that contain high starch contents (from about 50% to about 88% by weight ungelatinized and about 12% to about 50% by weight of gelatinized starch).

U.S. Publication No. US 2002/0108532 and PCT Publication No. WO 00/39213 filed by Apack AG disclose methods to produce a shaped body made of biodegradable material that shows good expansion behavior during thermoforming from 7.6 to 8.5% by weight of cellulosic fibers, from 16.1 to 17.6% by weight of native starch, from 5.4 to 6% by weight of pregelatinized starch and from 68.0 to 70.6% by weight of water. First, the pregelatinized starch is produced by mixing between 5.4-6% starch and 94-94.6% water, heating the mixture to 68-70°C, holding the mixture constant at 68-70°C for 10 minutes, and cooling the pregelatinized starch to 50°C. Then, adding the 16.1 to 17.6% by weight of native starch, 7.6 to 8.5% by weight of cellulosic fibers, and 68.0 to 70.6% by weight of water to the pregelatinized solution at a temperature of 50°C; mixing for 5 minutes to achieve a homogeneous mixture at 40°C, not allowing the mixture to substantially cool, and placing the mixture in a baking mold, and baking the mixture at 100-200°C for 10-100 seconds to form the shaped body.
German patent DE 19706642 to Apack Verpackungen Gmbh discloses the production of a biodegradable article from 25-75% fibers, 13-38% starch and 13-38% water. First, the 25-75% fibers, 13-38% starch are mixed in a dry state in a continuous process; then water is admixed continuously. The mixture is then subjected to a baking process to obtain the finished molded article, and then the molded article is coated with a biologically degradable film that is impermeable to humidity.

EP 0773721 B1 filed by Cooperatieve Verkoop-en Productievereniging van Aardappelmeel en Derivaten B.A. discloses a method for manufacturing a biodegradable molding suitable for containing liquid or more or less solid food or non-food products without loss of firmness or the molding. The molding contains 500-1500 parts by weight of a starch product containing 5-75% by weight of a starch derivative which in comparison to native starch has a reduced swelling capacity at an increased temperature, 0.5-50 parts by weight of a thickener, 25-300 parts by weight of an inert filler, and an adhesion promoting substance are prepared with water and mixed to obtain a homogenous suspension. The suspension is introduced into a baking mould and is baked to obtain a self-supporting base molding. A biodegradable wax composition containing at least 50% by weight of wax components and having a melting range beginning at temperature of at least 40 degrees C.

U.S. Patent No. 5,849,152 teaches a process for the production of shaped bodies, in particular packaging shaped bodies, from biologically decomposable material using a viscous mass containing biologically decomposable fiber material, water and starch, which is baked with the formation of a composite of fiber material and starch in a baking mould, and a shaped body produced according to this process. The use of native and pre-gelatinated or modified starch in connection with a fiber material feed stock from fibers or fiber bundles of differing lengths is said to be especially advantageous.

U.S. Patent No. 5,916,503 discloses a process for the manufacture of new shaped parts, preferably made from chipboards, wherein a mass formed from at least a binder and a small-particled material brought into contact with the binder is subjected to extrusion at elevated temperature and pressure. According to this process, a mass possessing a total moisture content of 6 to 25 wt. % or adjusted to the stated moisture content and made up of at
least one biopolymeric preferably starch-containing binder, which converts into a melt and/or gel at extrusion temperatures and pressures, and further made up of the small-particled material, is subjected to extrusion and immediately thereafter undergoes decompression and spontaneous expansion.

DE 19751234 discloses biologically degradable foamed molded articles of a fiber/starch material. Starch is provided in an amount of 8 to 26%, further hydro- colloids in an amount from 5 to 15%, and a foaming means in an amount of up to 3%, whereby a foam means acts at approximately 80 to 140°C, so that the volume of the mass becomes larger in such a manner by expansion of the water vapor and the sets free gas from the foam means thereby filling in the figuration cavity. The invention is applied to producing shaped elements, in particular for packaging uses.

EP 0524920 describes new shaped elements for construction, insulation and packaging purposes on the basis of biogenic, biologically degradable materials including starch or a starch-containing material, characterized in that it is formed by a plurality of performs connected to each other at least in places by a plurality of contact points with a structure-providing base matrix comprising a multiplicity small dimensioned voids, bubbles or pores on the basis of at least one melt solidified or gelled after exposure to an elevated temperature, elevated humidity, elevated pressure and/or mechanically stress or a gel of at least one starch or one starch-containing material thus obtained.

U.S. Patent Nos. 6,878,199 and U.S. 7,083,673 disclose method and materials for forming biodegradable containers that can hold food products in dry, damp or wet conditions and provides the biodegradable containers prepared according to the disclosed process. The containers are produced through the use of a pre-gelled starch suspension that is said to be unique in its ability to form hydrated gels and to maintain this gel structure in the presence of many other types of materials and at low temperatures.

Although numerous attempts have been made to provide suitable biodegradable and compostable materials for packaging, the resulting substances are not ideal. The currently available materials either cannot successfully be used to package materials, particularly those that are wet, or do not effectively degrade under normal environmental conditions. A need
exists to develop materials that will reduce the build up of disposed, slowly degrading materials, and to limit the environmental damage caused by toxic chemicals used in the production of packaging materials. Biodegradable or compostable materials of this sort, which are lightweight and flexible, are also highly desired.

SUMMARY OF THE INVENTION

The present invention provides improved methods for batch processing or continuous processing of materials for use in the formation of biodegradable containers that can hold food products in dry, damp, or wet conditions. In addition, the present invention provides novel formulations that result in improved properties of the biodegradable containers, such as but not limited to, increased flexibility and strength.

In one aspect, the present invention is a process for forming a biodegradable material involving

(i) forming a pre-gelled waxy starch suspension, the "pre-gel," comprising at least one or more starches, and optionally, one or more of cellulose pulps or powders, and one or more plant derived oils, such that the pre-gel is maintained at temperatures between about 0-60°C, preferably about 0-40°C;

(ii) adding to the pre-gel a dry or damp, homogenous mixture containing at least one or more waxy or native starches, and optionally, one or more of, wood fibers or wood flour, diatomaceous earth, mold release agents, foaming agents, and citric acid to form a homogenous moldable composition; and

(iii) molding the homogenous moldable composition with heat to form a biodegradable material.

In another aspect, the present invention is a "flash-gel" process for forming a biodegradable material involving:
(i) forming a pre-gelled starch suspension, the "pre-gel," by first pre-heating and maintaining water at temperatures between 65-80\(^\circ\)C, preferably between 65-75\(^\circ\)C, adding a cellulose pulp or powder, followed by addition of at least one or more starches, and optionally, one or more plant derived oils;

(ii) adding to the pre-gel a dry or damp, homogenous mixture containing at least one or more waxy or native starches, and optionally, one or more of, wood fibers or wood flour, diatomaceous earth, mold release agents, foaming agents, and citric acid to form a homogenous moldable composition; and

(iii) molding the homogenous moldable composition with heat to form a biodegradable material.

The flash gel method, while useful for batch preparation, allows for faster and more efficient production of the moldable homogenous compositions using continuous batch preparation. For example, a typical batch preparation that requires adding the components of the pre-gel to water and then heating until a pre-gel forms can take 30 minutes to 1 hour, or more to prepare the final moldable homogenous composition. In contrast, the flash gel method allows for formation of a pre-gel in 30 to 120 seconds and production of a moldable homogenous composition within 10 to 15 minutes.

In one embodiment, the pre-gel comprises 3-10\%, preferably 3, 5, 7.5, or 10\% starch by weight of the pre-gel, and 90-97\% water by weight of the pre-gel.

In another embodiment, the pre-gel comprises 2-15\% starch by weight of the pre-gel, 5-15\% cellulose pulp or cellulose powder by weight of the pre-gel, and an amount of water sufficient to equal 100\% of the total weight of the pre-gel.

In another embodiment, the pre-gel comprises 5-15\%, preferably 10\% starch by weight of the pre-gel, 5-10\% paper pulp, preferably 5.9-8.0\%, by weight of the pre-gel, and an amount of water sufficient to equal 100\% of the total weight of the pre-gel.
In yet another embodiment, the pre-gel comprises 5-15% starch, preferably 8.2%, by weight of the pre-gel, 5-10% wood fibers or wood flour, preferably 9.2%, by weight of the pre-gel, and an amount of water sufficient to equal 100% of the total weight of the pre-gel.

In one embodiment, the starch of the pre-gel is a waxy potato starch. Waxy starches include starches containing at least about 85% by weight amylopectin, preferably about 90% by weight amylopectin, more preferably about 95% by weight amylopectin, even more preferably about 100% amylopectin (i.e., amylose-free). In another embodiment, the starch is a native pea starch.

In one embodiment, the pre-gel further comprises 0.01-2.0% of a plant derived oil by weight of the pre-gel. In one exemplary embodiment, the plant-derived oil is soy oil.

In one embodiment, the homogenous mixture comprises a native or waxy corn or potato starch. In another embodiment the homogenous mixture comprises a waxy potato starch and a native corn starch. In yet another embodiment the homogenous mixture comprises a native potato starch, a native corn starch and a tapioca starch. In another embodiment the homogenous mixture comprises a native potato starch, a native tapioca starch, and a modified tapioca starch. In yet another embodiment, the homogenous mixture comprises native potato and tapioca starch.

In one embodiment, the homogenous mixture comprises one or more starches selected from native corn, native potato, waxy potato, native tapioca, and modified tapioca, and diatomaceous earth. In another embodiment the homogenous mixture comprises a native tapioca starch and/or a modified tapioca starch and diatomaceous earth. In one embodiment the diatomaceous earth is in an amount of 30-60% by weight of the homogenous moldable composition. In yet another embodiment, the diatomaceous earth is in an amount of 50% by weight of the homogenous moldable composition.

Optionally, wood fibers or wood flour can be included in the dry or damp homogenous mixture. In a particular embodiment, the wood fibers and wood flour have an aspect ratio of about 1:2 and 1:8.
In addition, the homogenous mixture may optionally include fillers such as, but not limited to, bentonite clay, gypsum, calcium sulfate, limestone and fly ash.

The biodegradable or compostable materials formed according to the process of the present invention can be coated or filmed, either before, during or after any of the molding processes.

In one embodiment, the materials are coated or filmed by applying a heated biodegradable film, wherein the temperature of the biodegradable or compostable material is approximately the melt point of the biodegradable film, as described in co-pending U.S. Patent Application No. 11/605,169.

DETAILED DESCRIPTION OF THE INVENTION

Definitions

The term "sheet" as used herein refers to any substantially flat, corrugated, curved, bent, or textured sheet made using the methods described herein. The sheets can also include organic coatings, printing, other sheets laminated thereto. The sheets within the scope of the present invention can have greatly varying thicknesses depending on the particular applications for which the sheets are intended. The sheets can be as thin as about 0.001 mm or as thick as 1 cm or greater where strength, durability, and or bulk are important considerations.

The term "film" is not inherently different from the term "sheet" except that "film" normally denotes a very thin sheet. Films are often formed by processes that are different from how sheets are normally formed, such as by film blowing rather than sheet calendering. In general, films will be defined as sheet-like articles having thicknesses as low as about 1 micron and up to about 1 mm.

The term "molded article" shall refer to articles that are shaped directly or indirectly from starch compositions using any molding method known in the art.
The term "injection molding" shall refer to articles that are made in classic injection machines such as, but not limited to, the Arburg Model 42 Allrounder. In an injection molding process the dough is injected directly into a heated mold at temperatures between 176°C and 210°C, preferably between 196°C and 218°C, more preferably between 198°C and 210°C. The dough is injected into a heated mold via a plunger type system rather than a runner type system.

The term "container" as used in this specification and the appended claims is intended to include any article, receptacle, or vessel utilized for storing, dispensing, packaging, portioning, or shipping various types of products or objects (including, but not limited to, food and beverage products). Specific examples of such containers include, among others, boxes, cups, "clam shells," jars, bottles, plates, bowls, trays, cartons, cases, crates, cereal boxes, frozen food boxes, milk cartons, bags, sacks, carriers for beverage containers, dishes, egg cartons, lids, straws, envelopes, or other types of holders. In addition to integrally formed containers, containment products used in conjunction with containers are also intended to be included within the definition "container". Such articles include, for example, lids, liners, straws, partitions, wrappers, cushioning materials, utensils, and any other product used in packaging, storing, shipping, portioning, serving, or dispensing an object within a container.

As used herein, the term "dry or damp" refers to a solid composition that can be dry, or can be moist or wetted, generally with water, although other solvents may be used. The amount of liquid in the composition is not sufficient to act as a carrier between particles in the composition.

As used herein, the term "homogeneous mixture" refers to mixtures of solid particulates or of solids in a liquid carrier which are substantially uniform in composition on a macroscopic scale. It will be appreciated that mixtures of different types of solid particles or of solids in a liquid carrier are not homogeneous when viewed on a microscopic scale, i.e., at the particle size level.

As used herein, the term "native starch" refers to a starch that has been derived from its natural source, wherein the natural source has not been genetically or chemically modified to increase its amylopectin content.
As used herein, the term "modified starch" refers to a starch that has been derivatized or modified by typical processes known in the art such as, e.g. esterification, etherification, oxidation, acid hydrolysis, cross-linking, and enzyme conversion. Typical modified starches include esters, such as the acetate and the half-esters of dicarboxylic acids/anhydrides, particularly the alkenylsuccinic acids/anhydrides; ethers, such as the hydroxyethyl and hydroxypropyl starches; oxidized starches, such as those oxidized with hypochlorite; starches reacted with cross-linking agents, such as phosphorus oxychloride, epichlorohydrin, hydrophobic cationic epoxides, and phosphate derivatives prepared by reaction with sodium or potassium orthophosphate or tripolyphosphate, and combinations thereof. Modified starches also include seagel, long-chain alkylstarches, dextrins, amine starches, and dialdehyde starches.

Pre-gelled starch suspensions

Starches used in forming the pre-gelled starch suspension used in the method of the invention desirably possess the following properties: the ability to form hydrated gels and to maintain this gel structure in the presence of many types of other materials; and the ability to melt into plastic-like materials at low temperatures, for example, between 0-60°C, preferably between 0-40°C, and in the presence of a wide range of materials and in moist environments and to exhibit high binding strengths and produce an open cell structure for both insulation and cross linking of components.

While not intending to be bound to any specific mechanistic explanation for the desirable properties observed when the method of the invention is employed, it is believed that the gel property holds other components in suspension until the product can be molded and to hold the moisture levels constant within the mixture until and during molding. The second property is evident in the transition in the mold of the gel structure into a drier and dried form that will then melt into the binding plastic-like product within the confines of the mold. This complex three dimensional cross linked structure provides a backbone for the product, exhibiting both strength and insulation properties.
Starch is produced in many plants, and many starches may be used (e.g., corn, waxy corn, wheat, sorghum, rice, and waxy rice), which can be used in the flour and cracked state. Other sources of starch include tubers (potato), roots (tapioca (i.e., cassava and maniac), sweet potato, and arrowroot), and the pith of the sago palm. Suitable starches can also be selected from the following: ahipa, apio (arracacha), arrowhead (arrowroot, Chinese potato, jicama), baddo, bitter casava, Brazilian arrowroot, casava (yucca), Chinese artichoke (crosne), Japanese artichoke (chorogi), Chinese water chestnut, coco, cocoyam, dasheen, eddo, elephant's ear, girasole, goo, Japanese potato, Jerusalem artichoke (sunroot, girasole), lilly root, ling gaw, malanga (tanier), plantain, sweet potato, mandioca, manioc, Mexican potato, Mexican yam bean, old cocoyam, saa got, sato-imo, seegoo, sunchoke, sunroot, sweet casava, tanier, tannia, tannie, tapioca root, taro, topinambour, water chestnut, water lily root, yam bean, yam, yautia, barley, corn, sorghum, rice, wheat, oats, buckwheat, rye, kamut brand wheat, triticale, spelt, amaranth, black quinoa, hie, millet, plantago seed husks, psyllium seed husks, quinoa flakes, quinoa, teff and legumes such as the field pea.

Starch is typically considered a natural carbohydrate chain comprising polymerized glucose molecules in an alpha-(1,4) linkage. Starch contains a mixture of two molecules amylose and amylopectin. Both consist of polymers of α-D-glucose units in the 4Cl conformation. In the amylose molecule, these are linked -{(I→4)}-, with the ring oxygen atoms all on the same side, whereas in amylopectin molecule about one residue in every twenty or so is also linked -{(I→6)}- forming branch-points. Starches vary from one another in several ways, including amylose:amylopectin ratio, structure of amylose and amylopectin molecules, granual size and shape and other variations.

The relative proportions of amylose to amylopectin depend on the source of the starch. See J-Y. Li and A-I. Yeh, Relationships between thermal, rheological characteristics and swelling power for various starches, J. Food Engineering 50 (2001) 141-148. (b) N. Singh, J. Singh, L. Kaur, N. Singh Sodhi and B. Singh Gill, Morphological, thermal and rheological properties of starches from different botanical sources, Food Chem. 81 (2003) 219-231. This ratio varies not only among the different types of starch, but among the many plant varieties within a type. In the native form, most starches contain 18% to 28% wt.% amylose. The commercially most important starch types (maize starch, potato starch, wheat starch and
tapioca starch) contain 15 to 30 wt. % amylose. Native, non-waxy potatoes are typically about 20-25 wt. % amylose. Native pea starches contain amylose at higher levels above 30 and as high as 70%.

"Waxy" starch is a term used to describe starches with high amylopectin ratios. In particular, the term "waxy" is often used to describe starch or flour containing at least about 85% by weight amylopectin, and more preferably, about 90 % by weight amylopectin, and even more preferably, about 95% by weigh amylopectin, and most preferably about 99% by weight amylopectin. Certain waxy starches are 100% amylopectin or, put another way, amylose-free.


Of some cereal types, such as barley, maize, millet, wheat, milo, rice and sorghum, there are varieties of which the starch granules nearly completely consist of amylopectin. Calculated as weight percent on dry substance, these starch granules contain more than 95%, and usually more than 98% amylopectin. The amylose content of these cereal starch granules is thus less than 5%, and usually less than 2%.

Legume sources often contain high levels of amylase. Native pea starches contain amylose at higher levels above 30% and as high as 70%.

Regular corn contains 72-76% amylopectin and 24-28% amylose. Waxy corn, containing higher levels of amylopectin, including up to 100% amylopectin, has been known
for nearly a century. The waxy trait is controlled by a single recessive gene, the wx gene. See See M. G. Nueffer, L. Jones, and M. Zuber, "The Mutants of Maize" (Crop Science Society of America, Madison, Wis., 1968), pp. 72 and 73. Common waxy maize is grown for many purposes and is commercially available.

Rice with a high percentage of amylopectin has grains that become sticky and tend to disintegrate upon cooking. Sticky or glutinous rice consists essentially of starch that is pure amylopectin.


Wheat starch is normally 75% amylopectin and 25% amylose. Three genes encode GBSS in hexaploid wheat. Naturally occurring mutations (null alleles) resulting in the loss of one or more GBSS isoforms have been identified. The presence of one or two GBSS null alleles results in the production of starch with reduced amylose content. Reduced amylose wheats have been termed "partial waxy". Wheats with three GBSS null alleles produce essentially amylose-free, or waxy, starch. See Waxy wheats: Origin, properties, and prospects Graybosch RA. In: Trends in Food Science & Technology, 1998, 9(4):135-142.

In contrast to the situation of cereals, root and tuber varieties of which the starch granules nearly exclusively consist of amylopectin are not known in nature. For instance, potato starch granules isolated from potato tubers usually contain about 20-25% amylose and 75-80% amylopectin (wt. % on dry substance). The elimination of these genes can be realized by recessive modification or genetic modification of potato plant material. An example thereof is the amylose-free mutant of the potato of which the starch substantially only contains amylopectin through a recessive mutation in the GBSS gene. This mutation technique is described in, inter alia, J. H. M. Hovenkamp-Hermelink et al., "Isolation of amylose-free starch mutant of the potato (Solanum tuberosum L.)", Theor. Appl. Gent., (1987), 75:217-221, and E. Jacobsen et al., "Introduction of an amylose-free (amf) mutant into breeding of cultivated potato, Solanum tuberosum L., Euphytica, (1991), 53:247-253.
Recombinant modification of potato plants aimed at altering the starch synthesized in these plants has been described (see, e.g., International Patent Cooperation Treaty Publications WO 92/1375 and WO 92/14827). Elimination or inhibition of the expression of the GBSS gene in the potato is also possible by using so-called antisense inhibition. This genetic modification of the potato is described in R. G. F. Visser et al., "Inhibition of the expression of the gene for granule-bound starch synthase in potato by antisense constructs", Mol. Gen. Genet., (1991), 225:289-296. See U.S. Patent No. 6,784,338 and US Patent No. 5,824,798.

Commercial sources of high amylopectin potato starches are known. Eliane™ (Avebe Group, the Netherlands) is a high amylopectin potato produced by physical processing and natural breeding techniques which is sold in a variety of formulations, including Eliane™100. See U.S. Patent No. 6,660,093 (Visser et al.) which discloses a potato plant producing essentially amyllose-free starch. Amflora™ (BASF, Ludwigshafen, Germany) is a genetically engineered high amylopectin product, with an amylpectin content of around 98 percent. Amflora™ is produced as renewable raw material and only used in the paper, textile or glue industry, rather than human consumption.

By using genetic modification, it has been found possible to cultivate and breed roots and tubers, for instance potato, yam, or cassave (Patent South Africa 97/4383), of which the starch granules contain little or no amyllose.

The composition and properties of root and tuber starches, such as amylopectin-potato starch and tapioca amylopectin-starch differ from those of the waxy cereal starches. Amylopectin-potato starch has a much lower content of lipids and proteins than the waxy cereal starches. Problems regarding odor and foaming, which, because of the lipids and/or proteins, may occur when using waxy cereal starch products (native and modified), do not occur or occur to a much lesser degree when using corresponding amylopectin-potato starch products. In contrast to the waxy cereal starches, amylopectin-potato starch contains chemically bound phosphate groups. As a result, amylopectin-potato starch products in a dissolved state have a distinct polyelectrolyte character.
A few varieties of natural "partial waxy" wheat have been identified in which one or two of the iso-enzymes are inactive or missing. Starches derived from this wheat are also called "partial waxy". The concentration in amylose in these "partial waxy" starches is low and variable, but it is not zero, and in some cases the content of amylose remains almost unchanged by a compensation phenomenon.

Other variations in starches come from the structure of amylose and amyllopectin molecules. The length of the amylose molecules in a starch (known as its degree of polymerization) can also vary. In amyllopectin, the length and number of branches on the molecule are also variable.

Additional starch variations include granule size and shape. Starch is found in nature in the form of granules which are easily liberated from plant materials in known processes. The starch is found tightly and radially packed into dehydrated granules (about one water per glucose) with origin-specific shape and size. Sizes ranging from 3 microns to over 100 microns. Representative granule sizes: maize (2-30 µm), wheat (-45 µm) and potato (5-100 µm). For some starches the granule size is polymodal, meaning the granules can be grouped into more than one size range. Granule shape also can be diverse. Granule shapes include symmetrical spheres, asymmetrical spheres, symmetrical disks and asymmetrical disks. Some granules exhibit their shape smoothly, while others are polyhedrons with a faceted surface. The size distribution determines its swelling functionality with granules being generally either larger and lenticular (lens-like, A-starch) or smaller and spherical (B-starch) with less swelling power. Granules contain 'blocklets' of amyllopectin containing both crystalline (-30%) and amorphous areas. As they absorb water, they swell, lose crystallinity and leach amylose. The higher the amylose content, the lower is the swelling power and the smaller is the gel strength for the same starch concentration.

Starch in the native state is completely insoluble. In general, starch granules are insoluble in cold water; however, if the outer membrane has been broken by, e.g., by grinding, the granules can swell in cold water to form a gel. When the intact granules are treated with warm water, the granules swell and a portion of the soluble starch diffuses through the granule wall to form a paste. In hot water, the granules swell to such an extent that they burst,
resulting in gelation of the mixture. The exact temperature at which a starch swells and gelates depends on the type of starch.

In one embodiment, the sources of starch for pre-gels according to the present invention are high amylopectin-content starches. In a particular embodiment, the starch or flour contains at least about 85, 86, 87, 88, 89, 90, 91, 92, 93, 94, 95, 96, 97, 98, 99 or 100% by weight amylopectin. In one embodiment, the starch or flour contains at least about 90% by weight amylopectin. In another embodiment, the starch or flour contains at least 95% by weight amylopectin. In a preferred embodiment, the starch or flour contains about 99% by weight amylopectin. In another embodiment, the starch or flour contains about 100% amylopectin.

The starch component of the pre-gel may include any known starch material. The starch can be selected from natural starch, chemically and/or physically modified starch, biotechnologically produced and/or genetically modified starch and mixtures thereof.

In one embodiment, the starch is a waxy starch. In another embodiment, the waxy starch is a waxy tuber, such as a waxy potato. In yet another embodiment, the waxy starch is a waxy root, such as a waxy tapioca or waxy cassava. Other waxy starches suitable for use in the present invention include waxy wheat, waxy pea, waxy yam, waxy arrowroot, waxy manihot and waxy sweet potato.

In one embodiment, the starch used to form the pre-gel is a waxy potato starch. Examples of such a potato plant variety are the variety Apriori or Apropect, or varieties derived thereof.

The high amylopectin content starch used to form the pre-gel may be the produced by any means, including but without limitations, physical means of separation, classical plant breeding or genetic engineering of the plant source. In an exemplary embodiment, the high amylopectin content starch is produced by physical means of separation or classical plant breeding. In one embodiment, the high amylopectin starch is Eliane™, and more particularly Eliane 100™. In another embodiment, the high amylopectin content starch is a genetically modified potato, such as Amflora™.
The source of starch for pre-gels according to the present invention may include starch mixtures of waxy and non-waxy (native) starches. In an exemplary embodiment, the starch mixture includes a first waxy starch and a second, non-waxy (native) starch wherein the non-waxy starch contains less than about 85% by weight amyllopectin, and more preferably, lower than about 84, 83, 82, 81, 80, 79, 78, 77, 76, 75, 74, 73, 72, 71, 70, 69, 68, 67, 66, 65, 64, 63, 62, 61, 59, 58, 57, 56, 54, 53, 52, 51, or 50% by weight amyllopectin. For example, the starch mixture includes 100% amyllopectin potato starch (amylose-free) and non-waxy potato starch (e.g., 80% amyllopectin starch).

The starch mixture may contain the same or different botanical source of starch. For example, a starch mixture may include (i) waxy potato starch and non-waxy potato starch; (ii) waxy potato starch and non-waxy corn starch; or (iii) waxy potato starch and non-waxy tapioca starch. Starches suitable for combination include without limitation potato, tapioca, yam, cassava, corn, rice, wheat and barley starch.

Starch mixtures of waxy or non-waxy starches in proportions between about 0% and 99% can be used to form the pre-gel. In one embodiment, the waxy starch represents about 5, 10, 15, 20, 25, 30, 35, 40, 45, 50, 55, 60, 65, 70, 75, 80, 85, 90, 95, or 99% of the starch mixture.

In an exemplary embodiment, a potato starch mixture is used to form the pre-gel which includes a high amyllopectin content potato starch (e.g., Eliane 100™) and a non-waxy potato starch, i.e., about 80% amyllopectin. According to this embodiment, the starch mixture includes about 5, 15, 20, 25, 30, 35, 40, 45, 50, 55, 60, 65, 70, 75, 80, 85, 90, 95, or 99% of the waxy potato starch (e.g., Eliane 100™). In one embodiment, the starch mixture is 50% waxy potato starch (e.g., Eliane 100™) and 50% non-waxy potato starch.

In another embodiment, a potato starch and cellulose fiber or cellulose flour mixture is used to form the pre-gel which includes a high amyllopectin content potato starch (e.g. Eliane 100™).

In another embodiment, when the pre-gel is formed from a single starch source or waxy mixture, the source of starch is not a waxy corn or waxy rice.
In another embodiment a low amylopectin starch can be used. An example is, native pea starch which contains amylose levels between 30 and 70%.

In addition to starch and water, the pre-gel can optionally contain cellulose pulps or cellulose powders and certain plant derived oils.

Cellulose pulps that may be used in the pre-gel can be produced by any method known in the art. Cellulose pulp production is a process that utilizes mainly arboreal species from specialized cultivations. To produce the cellulose pulp, wood, typically reduced to dimensions of about 30-40 mm and a thickness of about 5-7 mm, is treated at high temperature and pressure with suitable mixes of chemical reagents that selectively attack lignin and hemicellulose macromolecules, rendering them soluble. Pulps coming from this first treatment, commonly called "cooking", are called "raw pulps"; they still contain partly modified lignin and are more or less Havana-brown colored. Raw pulps can be submitted to further chemical-physical treatments suitable to eliminate almost entire lignin molecules and colored molecules in general; this second operation is commonly referred to as "bleaching".

For this process, rapid growth ligneous plants are mainly used, which, with the help of chemical substances (alkali or acids), in condition of high pressure and temperature, are selectively delignified to obtain pulps containing cellulose and other components of lignocellulose. These pulps are then submitted to mechanical and chemical-physical treatments, in order to complete the removal of lignin and hemicellulose residual components, and utilized thereafter for paper production. Any of form of cellulose pulp can be used in the packaging materials described herein.

Cellulose pulps or powders for this invention, including virgin cellulose pulp and recycled cellulose pulp, can be derived from any of the wood fibers described herein. Aspect ratios for cellulose pulps are less than about 1:10, preferably less than 1:9, and more preferably less than about 1.8. In a particular embodiment, the aspect ratio for the cellulose pulp or powder is about 1:8. Virgin cellulose pulp can be provided, e.g., as large blocks of compressed pulp and is derived from managed forests. Alternatively, virgin cellulose pulp can be obtained as loose pulps. Representative, non-limiting examples of cellulose pulps or powders include, e.g., Arboceil™ 1000, IFCl 178 or similar cellulose products.
Wood represents one source of cellulose pulp. Other suitable sources of cellulose pulp are known and include, e.g., cotton, flax, and hemp, sugarcane, bagasse, etc.

In one embodiment, oils, such as but not limited to, soy, canola, peanut, or palm oil may be added to the pre-gelled suspensions. In one exemplary embodiment, the plant derive oil functions as a water repellant.

In one aspect of the present invention, the pre-gelled starch suspension is produced from approximately 3-10%, preferably, 3, 5, 7.5 or 10%, waxy starch by weight of the pre-gel and 90-97% water by weight of the pre-gel such that the pre-gelled suspension is maintained at low temperatures. In one embodiment, the pre-gelled starch solution can be maintained at all temperatures above freezing, O°C. In another embodiment, the pre-gelled starch solution can be maintained for greater that 24 hours, up to a few days, if stored refrigerated, for example, between 3-15°C.

In an exemplary embodiment, a pre-gelled paper starch suspension is produced from approximately 5-15%, preferably 10%, waxy starch (by weight of the pre-gel), preferably a waxy potato starch; 5-10% cellulose fiber or powder (by weight of the pre-gel), preferably 5.9-8%, more preferably, 7.3-7.5, 6.5-6.7, or 5.9-6.1%; and an amount of water sufficient to equal 100% of the total weight of the pre-gel such that the pre-gelled suspension is maintained at low temperatures. In one embodiment, the pre-gelled paper starch solution can be maintained at all temperatures above freezing, O°C. In another embodiment, the pre-gelled paper starch solution can be maintained for greater that 24 hours, up to a few days, if stored refrigerated, for example, between 3-15°C.

In another aspect of the present invention, a pre-gelled paper starch suspension is produced from approximately 5-15%, preferably 10%, waxy starch by weight of the pre-gel (by weight of the pre-gel); 5-10% paper pulp (by weight of the pre-gel), preferably 5.9-8%, more preferably, 7.3-7.5, 6.5-6.7, or 5.9-6.1%; and an amount of water sufficient to equal 100% of the total weight of the pre-gel such that the pre-gelled suspension is maintained at low temperatures. In one embodiment, the pre-gelled paper starch solution can be maintained at all temperatures above freezing, O°C. In another embodiment, the pre-gelled paper starch
solution can be maintained for greater than 24 hours, up to a few days, if stored refrigerated, for example, between 3-15°C.

In another exemplary embodiment, a pre-gelled fiber starch suspension is produced from approximately 5-15%, preferably 8.2%, waxy starch (by weight of the pre-gel), 5-10% wood fibers or wood flour (by weight of pre-gel) preferably 9.2%, and a percentage of water sufficient to equal 100% of the total weight of the pre-gel, such that the water is pre-heated to a range of 65-75°C, prior to addition of the waxy starch and wood fibers or wood flour.

In one embodiment, virgin cellulose pulp is added to the pre-gel in an amount from about 5-10% by weight. In a particular embodiment, virgin cellulose pulp is 5, 6, 7, 8, or 9% by weight of the pre-gel. In a preferred embodiment, virgin cellulose pulp is 5% by weight of the pre-gel.

In another embodiment, recycled cellulose pulp is added to the pre-gel is an amount from about 5-10% by weight. In a particular embodiment, recycled cellulose pulp is 5, 6, 7, 8 or 9% by weight of the pre-gel. In a preferred embodiment, recycled cellulose pulp is 5% by weight of the pre-gel.

In another embodiment, plant derived oils are added to the pre-gel in an amount from 0.01 to 2.0 % by weight of the pre-gel. In an exemplary embodiment, soy oil is add in amount of 0.7% by weight of the pre-gel.

The pre-gelled starch may be prepared by mixing the starch with water at about ambient temperature (approximately 25°C). The gel is formed by slowly heating the water-starch mixture with constant agitation until a gel forms. Continued heating will slowly degrade the gel, so the process should be stopped as soon as an appropriate level of gelation is achieved. Gels can be used cold. The gel is stable for a few days if refrigerated. For storage a biocide can be added, preferably at a concentration of about 10 to about 500 ppm.

Alternatively, the pre-gelled starch may be prepared using a "flash gel" method that allows for preparation of a pre-gel that, while useable in batch preparation, makes continuous preparation of the homogenous moldable compositions possible. Continuous preparation in turn allows for easier scale-up of production as well as increases in the speed and efficiency
with which the final moldable homogenous compositions can be prepared. In the flash gel method the pre-gel is formed by first pre-heating the water to a range of 65-85\( ^\circ C \) followed by addition of cellulose pulps or powders. In one embodiment, the water is heated to 70-80\( ^\circ C \). In another embodiment, the water is pre-heated to 72\( ^\circ C \). The mixing of the cellulose fibers or powders results in heat reduction of the water to temperatures in the range of 55-75\( ^\circ C \). At this point the waxy starch and any additional components (plant derived oils, etc) are added. In contrast to adding the starch and other components at ambient temperature and then heating the mixture to 0 to 6\( ^\circ C \), which can take 10-15 minutes, the flash-gel method results in a pre-gel that forms within 30-120 seconds. In one exemplary embodiment, gelling of the starch materials takes place at about 62-63\( ^\circ C \). The increased efficiency and speed with which a pre-gel can be formed along with the ability to prepare the moldable homogenous compositions using continuous preparation results in a dramatic reduction in time and overall increase in efficiency with which the moldable homogenous compositions may be formed. For example, a typical batch process that requires adding the ingredients at ambient temperature followed by heating until a gel forms can take up to 30 minutes to 1 hour, or longer to prepare the final moldable homogenous composition. In contrast, the flash-gel method allows for preparation of a final moldable homogenous compositions within 10 to 15 minutes. One non-limiting example of a continuous mixer that may be used in preparation of the homogenous moldable compositions by the flash-gel method is the ReadCo LLC 2" paddle mixer (Readco Kurimoto, LLC, York, PA).

It is possible to reduce the amount of water in starch melts by replacing the water inherently found in starch with an appropriate low volatile plasticizer capable of causing starch to melt below its decomposition temperature, such as glycerin, polyalkylene oxides, mono-and diacetates of glycerin, sorbitol, other sugar alcohols, and citrates. This can allow for improved processability, greater mechanical strength, better dimensional stability over time, and greater ease in blending the starch melt with other polymers.

Water can be removed before processing by using starch that has been pre-dried so as to remove at least a portion of the natural water content. Alternatively, water can removed during processing by degassing or venting the molten mixture, such as by means of an extruder equipped with venting or degassing means. Native starch can also initially be
blended with a small quantity of water and glycerin in order to form starch melts that are subjected to a degassing procedure prior to cooling and solidification in order to remove substantially all of the water therefrom.

**Homogenous Mixture**

After formation of a pre-gel, dry or damp materials can be added (such as dry starches and optionally, fibers, flour, pulp, diatomaceous earth, lubricants, foaming agents, etc.) to produce the final moldable mixture. The dry or damp materials can be pre-mixed before addition to the pre-gel, to increase the homogeneity of the final product and increase the structural integrity of the final molded product. Preferably, the amount of pre-gel added to the final mixture is in the range of about 7-60% by weight of the homogenous moldable composition. Preferably, the pre-gel is about at least 7%, 8%, 9%, 10%, 11%, 12%, 16%, 16.3%, 25%, 33%, 42%, 47%, 54%, 50%, 52%, 55%, 56%, 60% or 60.4% by weight of the homogenous moldable composition.

The dry or damp materials are generally pre-mixed into a homogeneous mixture before being added to the pre-gel. The dry or damp materials can be mixed to form a homogeneous mixture using any suitable means, such as, for example, a Kitchen Aid® Commercial Mixer.

One component in the dry/damp material that can be added to the pre-gelled starch is a dry or damp starch binder component. This starch can be corn or other dry starch (for example potato or tapioca). In one embodiment, the starch is a waxy potato starch or other starch identified above in the section entitled "Pre-Gelled Starch Suspensions."

Pre-gelatinized starch-based binders can also be added to the moldable mixture. Pregelatinized starch-based binders are starches that have previously been gelated, dried, and ground back into a powder. Since pre-gelatinized starch-based binders gelate in cold water, such starch-based binders can be added to the moldable mixture to increase the mixture viscosity prior to being heated. The increased viscosity prevents settling and helps produce thicker cell walls. This starch component can be pre-gelled in a manner similar to that describes above. For example, the second starch component can be pre-gelled in a mixture of
between about 1 and 15% starch (for example 15% corn starch) and 85-99% water. In these cases additional dry starch can be added as necessary to the homogeneous mixture to adsorb excess water. If the pre-gelled second starch is still damp, the preferred amount to be added is in the range of 55-65% by weight of the homogenous moldable composition, most preferably about 57% or about 65%.

The concentration of the native or waxy starch binder within the moldable mixtures of the present invention are preferably in a range from about 5% to about 60% by weight of the homogenous moldable composition, more preferably in a range from about 15% to about 30%, and most preferably about at least 6%, 20%, 21%, 25%, 26%, 27%, 28%, 29%, 30%, 31%, or 34% by weight of the homogenous moldable composition. Furthermore, combinations of different starches can be employed to more carefully control the viscosity of the mixture throughout a range of temperatures, as well as to affect the structural properties of the final hardened article. For example, the mixture can consist of a mixture of dry or damp corn and potato starch (16-44% of corn and potato starch by weight of the homogenous moldable composition), such that the corn starch comprises between about 13-30%, preferably between about 13-18% or 28-30%, and the potato starch comprises between about 3-14%, preferably approximately 11-14% or 3-5% of the final homogenous moldable composition.

Preferred starches include waxy starches, such as waxy potato starch, or other suitable waxy tuber and root starches, as discussed above. Starch mixtures may also be utilized including, without limitation, waxy potato starch and corn starch (non-waxy).

In a particular embodiment, the dry or damp homogenous mixture added to the pre-gel includes waxy potato starch.

In another particular embodiment, the dry or damp homogenous mixture added to the pre-gel includes waxy potato starch, native tapioca starch and corn starch (non-waxy). In another embodiment the homogenous mixture comprises native corn starch, native tapioca starch and modified tapioca starch.
In yet another particular embodiment, the damp or dry homogenous mixture added to the pre-gel includes a waxy starch selected from the group including, without limitation, waxy potato, waxy tapioca, waxy wheat, waxy yam, waxy sweet potato and waxy arrowroot.

In one embodiment, diatomaceous earth can be used to replace substantial amounts of one or more starches in the homogenous mixture. The replacement of starch with diatomaceous earth results in unexpected improvements in the properties of the products made from the homogenous moldable compositions, such as but not limited to, increases in flexibility and strength. In one embodiment, diatomaceous earth is used to replace at least 50%, at least 75%, or 100% of one or more starches of the homogenous mixture. In one exemplary embodiment, the diatomaceous earth is ground, washed, dried and passed through a #40 sieve. Powders passing smaller sieves (#80 to #200) may also be used. In one non-limiting embodiment, the diatomaceous earth will pass a #100 sieve and be retained on a #200 sieve. In one embodiment, the size of the diatomaceous earth is, between 20 and 75 microns. In another embodiment the size of the diatomaceous earth is between 30 and 60 microns. In yet another embodiment, the diatomaceous earth is between 40 and 50 microns. In one embodiment, the diatomaceous earth is a fluxed calcined diatomaceous earth. A non-limiting example of fluxed calcined diatomaceous earth is Celite® (World Minerals, Santa Barbara, CA). In one embodiment the amount of diatomaceous used is in the range of 1-60% by weight of the homogenous moldable composition. In another embodiment, the amount of diatomaceous earth used is in the range of 30-60% by weight of the homogenous moldable composition. In yet another embodiment, the amount of diatomaceous earth used is 50% by weight of the homogenous moldable compositions.

In one embodiment of the present invention, additional fibers can be employed as part of the dry/damp material added to the pre-gelled starch, or as part of the pre-gel itself. Larger particles are considered to be fibers. The expression "fibers" refers to fine, thin objects restricted in their length, the length being greater than the width. They can be present as individual fibers or as fiber bundles. Such fibers can be produced in a manner known to those skilled in the art. Preferred fibers have a low length to diameter ratio and produce materials of excellent strength and light weight. In general, the fibers used in the invention will have an
aspect ratio of about between 1:2 and 1:10; 1:2 and 1:9; 1:2 and 1:8; 1:2 and 1:7; 1:2 and 1:6; 1:2 and 1:5; 1:2 and 1:4; 1:2 and 1:3; 1:2 and 1:2; or 1:2 and 1:9.9.

The fibers used are preferably organic, and most preferably cellulose-based materials, which are chemically similar to starches in that they comprise polymerized glucose molecules. "Cellulosic fibers" refers to fibers of any type which contain cellulose or consist of cellulose. Plant fibers preferred here are those of differing lengths typically in the range from 600 micron to 3000 micron, principally from hemp, cotton, plant leaves, sisal, abaca, bagasse, wood (both hard wood or soft wood, examples of which include southern hardwood and southern pine, respectively), or stems, or inorganic fibers made from glass, graphite, silica, ceramic, or metal materials. The cellulosic fibers include wood fibers and wood flour.

Wood flour and fibers are very much like rough tooth picks that have small barb like structures coming out from the main fiber to participate in the cross linkage process with the cooling starch melt. This property adds both strength and water resistance to the surface produced within the mold. The rapid grinding process to produce flour or short fibers bypasses the expensive and polluting processes that are used to manufacture pulp and paper. The wood flour can be a resinous wood flour. Preferably, the wood flour is softwood flour, which contains relatively large amounts of resin. Moreover, softwood is used industrially on a large scale, such as in the building trade, with the consequence that an abundance of wood flour from, for instance, saw mills, is available at a low price. Wood flours can be graded based on the mesh size the flour. In general, wood flour having a mesh size of 20 - 100 is suitable, and an aspect ratio or 1:8 or 1:9, or 1:10 or less.

In one aspect of the invention, at least about 1-50%, preferably 5-50% by weight of wood fibers or flour is added to the final mixture. In one embodiment, 11-24% by weight of wood fibers or flour is added to the final mixture. In a particular embodiment, wood fibers or flour comprise about at least about 1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19, 20, 21, 22, 23, 24, 25, 26, 27, 28, 29, 30, 31, 32, 33, 34, 35, 36, 37, 38, 39, 40, 41, 42, 43, 44, 45, 46, 47, 48, 49 or 50 by weight of the homogenous moldable composition.

It should also be understood that some fibers, such as southern pine and abaca, have high tear and burst strengths, while others, such as cotton, have lower strength but greater
flexibility. In the case where better placement, higher flexibility, and higher tear and burst strength are desired, a combination of fibers having varying aspect ratios and strength properties can be added to the mixture.

The dry or damp homogenous mixture may also include additional ingredients such as mold release agents/lubricants, foaming agents, and/or citric acid. Suitable lubricants include, but are not limited to, magnesium, calcium, or sodium stearate or oleates and similar lubricants. In one embodiment, the mold release agent is present in an amount between about 0.1% and 0.5% by weight of the homogenous moldable composition. A representative, non-limiting foaming agent is CT1480 (Clariant, Charlotte, NC). In one embodiment, the foaming agent is present in an amount between about 0.01 and 0.1%, preferably about 0.025% by weight of the homogenous moldable composition. In another embodiment citric acid is added to the homogenous mixture in an amount between about 0.001 to about 0.010% by weight of the homogenous moldable composition.

In an additional aspect of the present invention, it is recognized that to decrease the residual odor of the wood in the final product, the amount of paper pulp can be increased to 50%, or 30-50%, by weight of the final mixture, and the amount of wood flour or fiber can be decreased to 0%.

In one embodiment of the present invention, additional fibers (e.g., wood fibers or wood flour) are not employed as part of the dry/damp material added to the pre-gelled starch.

In addition to the components discussed above the homogenous mixture can also include one or more optional additional materials depending on desired characteristics of the final product. Fillers can be included for a stronger product. Suitable fillers include but are not limited to clays such as bentonite, amorphous raw products such as gypsum (calcium sulfate dehydrate) and calcium sulfate, minerals such as limestone and man made materials such as fly ash. These natural earth fillers are able to take part in the cross linking and binding that occurs during the molding process. Other examples of useful fillers include perlite, vermiculite, sand, gravel, rock, limestone, sandstone, glass beads, aerogel, xerogels, seagal, mica, clay, synthetic clay, alumina, silica, fused silica, tabular alumina, kaolin, microspheres, hollow glass spheres, porous ceramic spheres, calcium carbonate, calcium
aluminate, lightweight polymers, xonotlite (a crystalline calcium silicate gel), lightweight expanded clays, hydrated or unhydrated hydraulic cement particles, pumice, exfoliated rock, and other geologic materials. Partially hydrated and hydrated cement, as well as silica fume, have a high surface area and give excellent benefits such as high initial cohesiveness of the freshly formed article. Even discarded inorganically filled materials, such as discarded containers or other articles of the present invention can be employed as aggregate fillers and strengtheners. It will also be appreciated that the containers and other articles of the present invention can be easily and effectively recycled by simply adding them to fresh moldable mixtures as an aggregate filler. Hydraulic cement can also be added in either its hydrated or unhydrated form. Both clay and gypsum can be important aggregate materials because they are readily available, relatively inexpensive, workable, form easily, and can also provide a degree of binding and strength if added in high enough amounts (for example in the case of gypsum hemihydrate). Because gypsum hemihydrate can react with the water within the moldable mixture, it can be employed as a means for holding water internally within the molded article. Preferably, the inorganic materials are added in an amount from up to approximately 5%, 0-4%, 0-13%, 2-13% or 0-15% by weight of the weight of the final composition.

Because of the wide variety of agents that can be used as fillers, preferred concentration ranges are difficult to calculate. For bentonite clay a preferred range is from about 2.5-4% of the weight of the final mixture. The additional agents can be pre-dissolved or can be added dry. A preferred clay slurry is 20% bentonite clay in water.

In addition, further cellulose-based thickening agents can be added, which can include a wide variety of cellulosic ethers, such as methylhydroxyethylcellulose, hydroxymethylcellulose, carboxymethylcellulose, methylcellulose, ethylcellulose, hydroxyethylcellulose, hydroxypropylcellulose, hydroxypropylmethylcellulose, and the like. Other natural polysaccharide-based thickening agents include, for example, alginic acid, phycocolloids, agar, gum arabic, guar gum, locust bean gum, gum karaya, xanthan gum, and gum tragacanth. Suitable protein-based thickening agents include, for example, Zein®. (a prolamine derived from corn), collagen (derivatives extracted from animal connective tissue such as gelatin and glue), and casein (derived from cow's milk). Suitable synthetic organic
thickening agents include, for example, polyvinyl pyrrolidone, polyethylene glycol, polyvinyl alcohol, polyvinylmethyl ether, polyacrylic acids, polyacrylic acid salts, polyvinyl acryl
cids, polyvinyl acryl acid salts, polyacrylamides, ethylene oxide polymers, polyylactic acid, and latex. Latex is a broad category that includes a variety of polymerizable substances
formed in a water emulsion. An example is styrene-butadiene copolymer. Additional
copolymers include: vinyl acetate, acrylate copolymers, butadiene copolymers with styrene
and acetonitrile, methacrylates, vinyl chloride, acrylamide, fluorinated ethylenes.
Hydrophilic monomers can be selected from the following group: N-(2-hydroxypropyl)methacrylamide, N-isopropyl acrylamide, N,N-diethylacrylamide, N-
ethylmethacrylamide, 2-hydroxyethyl methacrylate, acrylic acid 2-(2-hydroxyethoxy)ethyl
methacrylate, methacrylic acid, and others, and can be used for the preparation of hydrolytically degradable polymeric gels. Suitable hydrophobic monomers can be selected
from the 2-acetoxyethyl methacrylate group of monomers comprising dimethylaminoethyl
methacrylate, n-butyl methacrylate, \( \text{tert} \)-butylacrylamide, n-butyl acrylate, methyl
methacrylate, and hexyl acrylate. The polymerization can be carried out in solvents, e.g. in
dimethylsulfoxide, dimethylformamide, water, alcohols as methanol and ethanol, using
common initiators of the radical polymerization. The hydrophilic gels are stable in an acidic
environment at pH 1-5. Under neutral or weak alkaline conditions at pH above 6.5, the gels
degrad. The gels mentioned above are nontoxic as well as the products of their
biodegradation.

Other co-polymers include: aliphatic polyester, polycaprolactone, poly-3-
hydroxybutyric acid, poly-3-hydroxyvaleric acid, polyglycolic acid, copolymers of glycolic
acid and lactic acid, and polylactide, PVS, SAN, ABS, phenoxy, polycarbonate,
nitrocellulose, polyvinylidene chloride, a styrene/allyl alcohol copolymer, polyethylene,
polypropylene, natural rubber, a sytrene/butadiene elastomer and block copolymer,
polyvinylacetate, polybutadiene, ethylene/propylene rubber, starch, and thermoplastic
segmented polyurethane, homopolymers or copolymers of polyesters, polyorthoesters,
polylactides, polyglycolides, polycaprolactones, polyhydroxybutyrates, polyhydroxyvalerates,
porno acids, pseudopolyamino acids, polyamides and polyanhydrides, homopolymers and
copolymers of polylactic acid, polyglycolic acid, polycaprolactone (PCL), polyanhydrides,
polyorthoesters, polyaminoacids, pseudopolyaminoacids, polyhydroxybutyrates, polyhydroxyvalerates, polyphosphazenes, and polyalkylcyanoacrylates.

Additional polymers that can be added include: citrates, diethyl citrate (DEC), triethyl citrate (TEC), acetyl triethyl citrate (ATEC), tributyl citrate (TBC), acetyl tributyl citrate (ATBC), phthalates such as dimethyl phthalate (DMP), diethyl phthalate (DEP), triethyl phthalate (TEP), dibutyl phthalate (DBP), dioctyl phthalate, glycol ethers such as ethylene glycol diethyl ether, propylene glycol monomethyl ether, ethylene glycol monoethyl ether, diethylene glycol monoethyl ether (Transcutol™), propylene glycol monotertiary butyl ether, dipropylene glycol monomethyl ether, n-methyl pyrrolidone, 2 pyrrolidone (2-Pyrrol™), propylene glycol, glycerol, glyceryl dioleate, ethyl oleate, benzylbenzoate, glycofurol sorbitol sucrose acetate isobutyrate, butyryltri-n-hexyl-citrate, acetyltri-n-hexyl citrate, sebacates such as dibutyl sebacate, tributyl sebacate, dipropylene glycol methyl ether acetate (DPM acetate), propylene carbonate, propylene glycol laurate, propylene glycol caprylate/caprate, caprylic/capric triglyceride, gamma butyrolactone, polyethylene glycols (PEG), glycerol and PEG esters of acids and fatty acids (Gelucires™, Labrafils™ and Labrasol™) such as PEG-6 glycerol monooleate, PEG-6 glycerol linoleate, PEG-8 glycerol linoleate, PEG-4 glyceryl caprylate/caprate, PEG-8 glyceryl caprylate/caprate, polyglyceryl-3-oleate, polyglyceryl-6-dioleate, polyglyceryl-3-isostearate, PEG-32 glycerol laurate (Gelucire 44/1™), PEG-32 glyceryl palmitostearate (Gelucire 50/13™), PEG-32 glyceryl stearate (Gelucire 53/10™), glyceryl behenate, cetyl palmitate, glyceryl di and tri stearate, glyceryl palmitostearate, and glyceryl triacetate (Triacetin™), vegetable oils obtained from seeds, flowers, fruits, leaves, stem or any part of a plant or tree including cotton seed oil, soy bean oil almond oil, sunflower oil, peanut oil, sesame oil. The use of two or more plasticizers in a combination or blend of varying ratios and hydrophilicity or hydrophobicity is also encompassed by the present invention. Plasticizers also include: phthalates, glycol ethers, n-methyl pyrrolidone, 2 pyrrolidone, propylene glycol, glycerol, glyceryl dioleate, ethyl oleate, benzylbenzoate, glycofurol sorbitol sucrose acetate isobutyrate, butyryltri-n-hexyl-citrate, acetyltri-n-hexyl citrate, sebacates, dipropylene glycol methyl ether acetate (DPM acetate), propylene carbonate, propylene glycol laurate, propylene glycol caprylate/caprate, caprylic/capric triglyceride, gamma butyrolactone, polyethylene glycols (PECs), vegetable oils obtained from seeds, flowers, fruits, leaves, stem or any part of a plant or tree including cotton seed oil, soy
bean oil, almond oil, sunflower oil peanut oil, sesame oil, glycerol and PEG esters of acids and fatty acids, polyglyceryl-3-oleate, polyglyceryl-6-dioleate, polyglyceryl-3-isostearate, PEG-32 glyceryl laurate, PEG-32 glyceryl palmitostearate, PEG-32 glyceryl stearate, glyceryl behenate, cetyl palmitate, glyceryl di and tri stearate, glyceryl palmitostearate, and glyceryl triacetate. These materials can also be added in combination with other polymers to improve flexibility.

In a particular embodiment, the homogenous mixture does not include a thickening agent.

The addition of these items will increase the efficiency of production of the product on an item basis. Baking powder and other materials, such as leavening agents, which release gases, (e.g., sodium or calcium bicarbonates or carbonates) can be included in the compositions of the invention to elevate the number of open cells in the final structure by introducing a source of carbon dioxide gas which is released in the mold.

Glycerol, microcrystalline wax, fatty alcohols and other similar organic molecules can be added as a mold release agent, and to produce a smoother surface on the finished product. Examples of agents that can be added, either as plasticizers or as mold releasing agents are ethylene glycol, propylene glycol, glycerin, 1,3-propanediol, 1,2-butandiol, 1,3-butandiol, 1,4-butanediol, 1,5-pentandiol, 1,5-hexandiol, 1,6-hexandiol, 1,2,6-hexantriol, 1,3,5-hexantriol, neopentylglycol, sorbitol acetate, sorbitol diacetate, sorbitol monoethoxylate, sorbitol diethoxylate, sorbitol hexaethoxylate, sorbitol dipropoxylate, arrunosorbitol, trihydroxymethylaminomethane, glucose/PEG, the reaction product of ethylene oxidewith glucose, trimethylolpropane monoethoxylate, mannitol monoacetate, mannitol monoethoxylate, butyl glucoside, glucose monoethoxylate, a-methyl glucoside, the sodium salt of carboxymethylsorbitol, polyglycerol monoethoxylate, erythritol, pentaerythritol, arabitol, adonitol, xylitol, mannitol, iditol, galactitol, allitol, sorbitol, polyhydric alcohols generally, esters of glycerin, formarnide, N-methylformamide, DMSO, mono- and diglycerides, alkylarruides, polyols, trimethylpropane, polyvinylalcohol with from 3 to 20 repeating units, polyglycerols with from 2 to 10 repeating units, and derivatives of the foregoing. Examples of derivatives include ethers, thiethers, inorganic and organic esters,
acetals, oxidation products, amides, and anilines. These agents can be added from 0-10%, preferably 3-4% (w/w). A consideration of the inventive mixture should be that the composition preferably contains at least 75%, more preferably at least 95% of natural or organic-derived materials by weight of the homogenous moldable composition.

Preparation of molded articles

The starch, and optionally, wood flour mixture, with any included additives, is added to the pre-gelled starch and mixed (for example with a Kitchen Aid® Commercial Mixer, a Hobart mixer, or a ReadCo 2" paddle mixer) until a homogeneous moldable mixture is generated. The mixture can be as thick as peanut butter or as thin as a pancake batter. Varying amounts of additional water can by added to facilitate different types of molding, since the form of the pre-molded [green] product is dependent on the mold, heating rate and drying/melt time. If the product is to be molded by classic injection methods the material is thinner, if the material is molded on the equipment described below the mixture is thicker. The material can also be rolled into green sheets and molded, extruded and made into dry pellets for other processes. The means of production for the product could be created from any of several possible process approaches. One specific methodology is described below, but this description is intended only to describe one possible means of production, and shall not be construed in any way to represent a limitation to the outlined approach. While the compression molding process detailed herein is useful, other types of compression molding, injection molding, extrusion, casting, pneumatic shaping, vacuum molding, etc can be used. One embodiment involves a means of production incorporating moving upper and lower continuous track assemblies each with an upper and lower substantially elongated horizontal section, and with a curved portion of track joining the upper and lower horizontal section for each of the upper and lower tracks. Riding in each of the track assemblies is a linked belt made from any material or combination of materials that allows the belt or belt assembly to be in constant or intermittent motion about the tracks. The track assemblies are located vertically such that the upper portion of the lower track and the lower portion of the upper track are in close proximity such that the belts of each track move at a synchronized speed and in a
common direction. In this embodiment, the male mold portion is mounted to the belt following the upper track, and the female portion of the mold is mounted to the belt following the lower track, with the tracks synchronized in a fashion that causes the mold halves to join and close as they merge between the upper and lower tracks. In this embodiment, the material to be processed is deposited into the female mold half prior to the mold halves closing, or is injected into the mold after it has been closed. The track and belt assemblies hold the mold halves together during drying by any of a number of, or combination of, methods including without limitation spring force, pneumatic force, or mechanical compression. Other forcing methods are possible. One possible arrangement of the curved end of the tracks aligns them such that the lower tracks' upper horizontal section are located to start before the upper tracks' lower horizontal section to allow the female mold half on the upper section of the lower track to assume a substantially horizontal orientation prior to the male mold half attached to upper track, thereby allowing the female mold half to receive deposited material before it engages the corresponding male mold half merging from the upper track and belt assembly. Other aspects that can be incorporated in this embodiment include, removable cavity inserts and or multiple cavities in the molds; heating of the molds or product to speed drying by electric, microwave, hot gas, friction, ultrasonic, or any other means: on the fly cleaning of the molds, on the fly coating of product with any of a number of coating agents.

In another embodiment, once the moldable mixture has been prepared, it is positioned within a heated mold cavity. The heated mold cavity can comprise many different embodiments, including molds typically used in conventional injection molding processes and die-press molds brought together after placing the inorganically filled mixture into the female mold. In one preferred embodiment, for example, the moldable mixture is placed inside a heated female mold. Thereafter, a heated male mold is complementarily mated with the heated female mold, thereby positioning the mixture between the molds. As the mixture is heated, the starch-based binder gelates, increasing the viscosity of the mixture. Simultaneously, the mixture increases in volume within the heated molds cavity as a result of the formation of gas bubbles from the evaporating solvent, which are initially trapped within the viscous matrix. By selectively controlling the thermodynamic parameters applied to the mixture (e.g., pressure, temperature, and time), as well as the viscosity and solvent content,
the mixture can be formed into a form-stable article having a selectively designed cellular structural matrix.

In a non-limiting embodiment, a temperature between 195-225°C, preferably 200°C is used for baking for a time period of 60-90 seconds, preferably 75 seconds. Temperatures can vary based on the article bring manufactured, for example, 200°C is preferred for the rapid production of thin-walled articles, such as cups. Thicker articles require a longer time to remove the solvent and are preferably heated at lower temperatures to reduce the propensity of burning the starch-based binder and fiber. Leaving the articles within the locked molds too long can also result in cracking or deformation of the articles.

The temperature of the mold can also effect the surface texture of the molds. Once the outside skin is formed, the solvent remaining within the interior section of the mixture escapes by passing through minute openings in the outside skin and then traveling between the skin and the mold surface to the vent holes. If one mold is hotter than the other, the laws of thermodynamics would predict, and it has been empirically found, that the steam will tend to travel to the cooler mold. As a result, the surface of the article against the hotter mold will have a smoother and more uniform surface than the surface against the cooler mold.

A variety of articles can be produced from the processes and compositions of the present invention. The terms "article" and "article of manufacture" as used herein are intended to include all goods that can be formed using the disclosed process.

Coating of molded article

Before, during, or after any of the molding processes, coatings can be applied to the surface of a substantially dried article for any desired purpose, such as to make the article more waterproof, grease and food product proof, more flexible, or to give it a glossier surface. Coatings can be used to alter the surface characteristics including sealing and protecting the article made therefrom. Coatings can provide protection against moisture, base, acid, grease, and organic solvents. They can provide a smoother, glossier, or scuff-resistant surface, they can help reinforce the article and coatings can also provide reflective, electrically conductive or insulative properties.
Water resistance can be achieved through the use of a water resistant layer applied on one or both sides of the product. There are many currently available coatings that can be used to coat this product. Some of these are: commercial water and/or solvent based coatings used in the paper industry, Zein® - a biodegradable material isolated from corn; poly lactic acid [PLA] - a polymer of lactic acid from fermentation feed stock; polyhydroxyalkanoates [PHA] from microbial fermentation; bacterial cellulose; chitosan based polymers - from shell fish wastes; natural waxes and oil based coatings.

Appropriate organic coatings include edible oils, melamine, polyvinyl chloride, polyvinyl alcohol, polyvinyl acetate, polyacrylates, polyamides, hydroxypropylmethylcellulose, polyethylene glycol, acrylics, polyurethane, polyethylene, polylactic acid, Biopol™ (a polyhydroxybutyrate-hydroxyvalerate copolymer), starches, soybean protein, polyethylene, and synthetic polymers including biodegradable polymers, waxes (such as beeswax or petroleum based wax), elastomers, edible oils, fatty alcohols, phospholipids and other high molecular weight biochemicals, and mixtures or derivatives thereof. Biopol® is manufactured by ICI in the United Kingdom. Elastomer, plastic, or paper coatings can aid in preserving the integrity of the article. Appropriate inorganic coatings include sodium silicate, calcium carbonate, aluminum oxide, silicon oxide, kaolin, clay, ceramic and mixtures thereof. The inorganic coatings can also be mixed with one or more of the organic coatings set forth above. Coatings based upon materials such as soybean oil or Methocel®. (available from Dow Chemical), either alone or in combination with polyethylene glycol, can be applied to the surface in order to permanently soften the article or a hinge area within the article.

The coating can be applied either during the forming process or after the article is formed. The coating can be formed during the forming process by adding a coating material that has approximately the same melting temperature as the peak temperature of the mixture. As the mixture is heated, the coating material melts and moves with the vaporized solvent to the surface of the article where it coats the surface.

The coatings can be applied to the shaped articles using any coating means known in the art of manufacturing paper, paperboard plastic, polystyrene, sheet metal, or other packaging materials, including blade, puddle, air-knife, printing, Dahlgren, gravure, and
powder coating. Coatings can also be applied by spraying the article with any of the coating materials listed below or by dipping the article into a vat containing an appropriate coating material. These materials can be applied either as a thin film or can be sprayed/dipped onto the product. The apparatus used for coating will depend on the shape of the article. For example, cups will usually be coated differently than flat plates. Bonding processes for application of thin films of water-resistant material are known in the art. Each of these coatings is biodegradable and should not significantly impact the compostability of the product. The second method of improving the water resistance of the product is to add one or more biodegradable materials to the material either before molding or as part of the molding process. In each of these cases the basic composition of the product will remain fairly constant.

A waterproof coating is desirable for articles intended to be in contact with water. As the articles having a starch-based binder have a high affinity for water, the preferred coatings are non-aqueous and have a low polarity. Appropriate coatings include paraffin (synthetic wax); shellac; xylene-formaldehyde resins condensed with 4,4’-isopropylidenephenolepichlorohydrin epoxy resins; drying oils; reconstituted oils from triglycerides or fatty acids from the drying oils to form esters with various glycols (butylene glycol, ethylene glycol), sorbitol, and trimethylol ethane or propane; synthetic drying oils including polybutadiene resin; natural fossil resins including copal (tropical tree resins, fossil and modern), damar, elemi, gilsonite (a black, shiny asphaltitic, soluble in turpentine), glycol ester of damar, copal, elemi, and sandarac (a brittle, faintly aromatic translucent resin derived from the sandarac pine of Africa), shellac, Utah coal resin; rosins and rosin derivatives including rosin (gum rosin, tall oil rosin, and wood rosin), rosin esters formed by reaction with specific glycols or alcohols, rosin esters formed by reaction formaldehydes, and rosin salts (calcium resinate and zinc resinate); phenolic resins formed by reaction of phenols with formaldehyde; polyester resins; epoxy resins, catalysts, and adjuncts; coumarone-indene resin; petroleum hydrocarbon resin (cyclopentadiene type); terpene resins; urea-formaldehyde resins and their curing catalyst; triazine-formaldehyde resins and their curing catalyst; modifiers (for oils and alkyds, including polyesters); vinyl resinous substances (polyvinyl chloride, polyvinyl acetate, polyvinyl alcohol, etc.); cellulosic materials (carboxymethylcellulose, cellulose acetate, ethylhydroxyethylcellulose, etc.); styrene polymers; polyethylene and its
copolymers; acrylics and their copolymers; methyl methacrylate; ethyl methacrylate; waxes (paraffin type I, paraffin type II, polyethylene, sperm oil, bees, and spermaceti); melamine; polyamides; polylactic acid; Biopol® (a polyhydroxybutyrate-hydroxyvalerate copolymer); soybean protein; other synthetic polymers including biodegradable polymers; and elastomers and mixtures thereof. Biopol® is manufactured by ICI in the United Kingdom. Appropriate inorganic coatings include sodium silicate, calcium carbonate, aluminum oxide, silicon oxide, kaolin, day, ceramic and mixtures thereof. The inorganic coatings can also be mixed with one or more of the organic coatings set forth above.

If the articles are used as containers or for other products intended to come into contact with foodstuffs, the coating material will preferably include an FDA-approved coating. An example of a particularly useful coating is sodium silicate, which is acid resistant. Resistance to acidity is important, for example, where the article is a container exposed to foods or drinks having a high acid content, such as soft drinks or juices. It is generally unnecessary to protect the article from basic substances, but increased resistance to basic substances can be provided by an appropriate polymer or wax coating, such as those used to coat paper containers.

Polymeric coatings, such as polyethylene, are useful in forming generally thin layers having low density. Low density polyethylene is especially useful in creating containers which are liquid-tight and even pressure-tight to a certain extent. Polymeric coatings can also be utilized as an adhesive when heat sealed.

Aluminum oxide and silicon oxide are useful coatings, particularly as a barrier to oxygen and moisture. The coatings can be applied to the article by any means known in the art, including the use of a high energy electron beam evaporation process, chemical plasma deposition and sputtering. Another method of forming an aluminum oxide or silicon oxide coating involves treating article with an aqueous solution having an appropriate pH level to cause the formation of aluminum oxide or silicon oxide on the article due to the composition of the article.

Waxes and wax blends, particularly petroleum and synthetic waxes, provide a barrier to moisture, oxygen, and some organic liquids, such as grease or oils. They also allow an
article such as a container to be heat sealed. Petroleum waxes are a particularly useful group of waxes in food and beverage packaging and include paraffin waxes and microcrystalline waxes.

In some cases, it can be preferable for the coating to be elastomeric or deformable. Some coatings can also be used to strengthen places where the articles are severely bent. In such cases, a pliable, possibly elastomeric, coating can be preferred.

Of course, it should be understood that the starch compositions of the present invention can themselves be used as coating materials in order to form a synergistic composite with, or otherwise improve the properties of, any number of other materials. Such disparate materials such as paper, paperboard, molded starch-bound articles such as starch-based foams, metals, plastics, concrete, plaster, ceramics, and the like can be coated with starch composition.

It can be desirable to apply print or other indicia, such as trademarks, product information, container specifications, or logos, on the surface of the article. This can be accomplished using any conventional printing means or processes known in the art of printing paper or cardboard products, including planographic, relief, intaglio, porous, and impactless printing. Conventional printers include offset, Van Dam, laser, direct transfer contact, and thermographic printers. However, essentially any manual or mechanical means can be used.

In a further aspect of the present invention, it is recognized that to facilitate the coating of the molded article, as well as for other specific indications, the amount of paper pulp can be increased to 50%, or 30-50%, by weight of the final mixture, and the amount of wood flour or fiber can be decreased to 0%.

When using a vacuum to form a film around the molded article, increasing the levels of wood flour/fiber and/or paper pulp can facilitate the vacuuming process. For example, wood flour/fiber and/or paper pulp levels can be increased to 30, 40 or 50% by weight of the final mixture.

The articles of the present invention can be, in one embodiment, coated according to the methods and utilizing the coating compositions as described in U.S. Patent Application
No. 11/605,169, which describes methods for filming biodegradable or compostable containers by applying a heated film to a heated biodegradable or compostable container, wherein the temperature of the container is approximately the temperature of the biodegradable film.

Types of articles produced

Materials capable of holding dry, damp and wet products have diverse uses. Containers suitable for holding dry materials can be used to hold dried fruit, or raw nuts such as almonds. Containers suitable for holding damp materials can be used to hold fresh mushrooms or tomatoes (for example in groups of 4 or 6) and should be able to perform this function for a period of at least about two to three weeks since normal packing to use time is about 14 days. Damp food packing can also be used with a hot fast food item such as French fries or hamburger, in which case the container needs to last for only a short time, for example about one hour after addition of the damp food. Damp food packing could also be used, in combination with an adsorbent pad, to package raw meat. In this case, the container needs to withstand exposure to the meat for a period of seven days or longer and desirably can stand at least one cycle of freeze and thaw. If possible this package should be able to withstand a microwave signal. When formulated for holding wet foods, the containers of the invention will suitably have the ability to hold a hot liquid, such as a bowl of soup, a cup of coffee or other food item for a period of time sufficient to allow consumption before cooling, for example within one hour of purchase. Such containers can also be used to hold a dry product that will be re-hydrated with hot water such as the soup-in-a-cup products.

Articles made from the present invention to can be manufactured into a wide variety of finished articles that can presently be made plastics, paper, paperboard, polystyrene, metals, ceramics, and other materials. Merely by way of example, it is possible to manufacture the following exemplary articles: films, bags, containers, including disposable and non-disposable food or beverage containers, cereal boxes, sandwich containers, "clam shell" containers (including, but not limited to, hinged containers used with fast-food sandwiches such as hamburgers), drinking straws, baggies, golf tees, buttons, pens, pencils, rulers, business cards, toys, tools, Halloween masks, building products, frozen food boxes,
milk cartons, fruit juice containers, yoghurt containers, beverage carriers (including, but not limited to, wraparound basket-style carriers, and "six pack" ring-style carriers), ice cream cartons, cups, French fry containers, fast food carryout boxes, packaging materials such as wrapping paper, spacing material, flexible packaging such as bags for snack foods, bags with an open end such as grocery bags, bags within cartons such as a dry cereal box, multiwell bags, sacks, wraparound casing, support cards for products which are displayed with a cover (particularly plastic covers disposed over food products such as lunch meats, office products, cosmetics, hardware items, and toys), computer chip boards, support trays for supporting products (such as cookies and candy bars), cans, tape, and wraps (including, but not limited to, freezer wraps, tire wraps, butcher wraps, meat wraps, and sausage wraps); a variety of cartons and boxes such as corrugated boxes, cigar boxes, confectionery boxes, and boxes for cosmetics-, convoluted or spiral wound containers for various products (such as frozen juice concentrate, oatmeal, potato chips, ice cream, salt, detergent, and motor oil), mailing tubes, sheet tubes for rolling materials (such as wrapping paper, cloth materials, paper towels and toilet paper), and sleeves; printed materials and office supplies such as books, magazines, brochures, envelopes, gummed tape, postcards, three-ring binders, book covers, folders, and pencils-, various eating utensils and storage containers such as dishes, lids, straws, cutlery, knives, forks, spoons, bottles, jars, cases, crates, trays, baking trays, bowls, microwaveable dinner trays, "TV" dinner trays, egg cartons, meat packaging platters, disposable plates, vending plates, pie plates, and breakfast plates, emergency emesis receptacles (i.e., "barf bags"), substantially spherical objects, toys, medicine vials, ampules, animal cages, firework shells, model rocket engine shells, model rockets, coatings, laminates, and an endless variety of other objects.

The container should be capable of holding its contents, whether stationary or in movement or handling, while maintaining its structural integrity and that of the materials contained therein or thereon. This does not mean that the container is required to withstand strong or even minimal external forces. In fact, it can be desirable in some cases for a particular container to be extremely fragile or perishable. The container should, however, be capable of performing the function for which it was intended. The necessary properties can always be designed into the material and structure of the container beforehand.
The container should also be capable of containing its goods and maintaining its integrity for a sufficient period of time to satisfy its intended use. It will be appreciated that, under certain circumstances, the container can seal the contents from the external environments, and in other circumstances can merely hold or retain the contents.

The terms "container" or "containers" as used herein, are intended to include any receptacle or vessel utilized for, e.g., packaging, storing, shipping, serving, portioning, or dispensing various types of products or objects (including both solids and liquids), whether such use is intended to be for a short-term or a long-term duration of time.

Containment products used in conjunction with the containers are also intended to be included within the term "containers." Such products include, for example, lids, straws, interior packaging, such as partitions, liners, anchor pads, corner braces, corner protectors, clearance pads, hinged sheets, trays, funnels, cushioning materials, and other object used in packaging, storing, shipping, portioning, serving, or dispensing an object within a container.

The containers within the purview of the present invention can or can not be classified as being disposable. In some cases, where a stronger, more durable construction is required, the container might be capable of repeated use. On the other hand, the container might be manufactured in such a way so as to be economical for it to be used only once and then discarded. The present containers have a composition such that they can be readily discarded or thrown away in conventional waste landfill areas as an environmentally neutral material.

The articles within the scope of the present invention can have greatly varying thicknesses depending on the particular application for which the article is intended. They can be as thin as about 1 mm for uses such as in a cup. In contrast, they can be as thick as needed where strength, durability, and or bulk are important considerations. For example, the article can be up to about 10 cm thick or more to act as a specialized packing container or cooler. The preferred thickness for most articles is in a range from about 1.5 mm to about 1 cm, with about 2 mm to about 6 mm preferred.

Using a microstructural engineering approach, the present invention can produce a variety of articles, including plates, cups, cartons, and other types of containers and articles
having mechanical properties substantially similar or even superior to their counterparts made from conventional materials, such as paper, polystyrene foam, plastic, metal and glass. The inventive articles can also be made at a fraction of the cost of their conventional counterparts. The minimal cost is a result of the relatively inexpensive aggregate which typically comprises a large percentage of the mixture and the minimum processing energy required.

The method of the present invention provides basic methodologies which can be utilized with little modification and a basic material from which product items can be produced by tailoring of the additives and additional processing steps employed. The composition preferably contains at least 75%, at least 85% or at least 95% or more of natural or organic-derived materials by weight of the homogenous moldable composition.

EXAMPLES

The following examples are presented in order to more specifically teach compositions and process conditions for forming the thermoplastic starch compositions according to the present invention, as well as articles therefrom. The examples include various mix designs, as well as various processes for manufacturing thermoplastic starch compositions, including sheets, films, pellets, containers, and other articles of manufacture.

Examples are as follows:

**Example 1** [100% waxy potato starch]

1. Form a pre-gelled cellulose paper-waxy potato starch suspension.

   5.8 g waxy potato starch [Eliane 100]: 8.3%

   6.5 g virgin cellulose pulp: 9.2%

   58 g Water: 82.5%

   Add components, heat to 60-70°C with mixing on high speed with a wire whisk to form the gel. Alternatively, as the paper pulp is dispersed, and as the temperature begins to rise (above
30°C), the RPM of the mixer is increased until the maximum RPM is reached. The heating
continues until the temp reaches 65°C. At this time, the mixture is a homogeneous gel
suspension. The heat is turned off and beater heads changed to classic dough hook and speed
is lowered to 10% of maximum (KitchenAid®). Once gelled, the gel may be cooled or
refrigerated (but not frozen) until used.

2. Premix the following materials:

   32.2 g waxy potato starch [Eliane 100]
   0.25 g magnesium stearate
   0.05 g foaming agent

   to form a homogenous mixture

3. Add homogenous mixture to the pre-gelled paper potato starch, mix with a dough hook
mixture on low speed. This mixture is stable and can be cooled or refrigerated, but not frozen.

4. Place mixture into mold (about 50-55 g) and bake at 195-225°C (ideal 215°C) for 60-90
seconds (ideal 75°C)

Example 1 is preferably used for flatter trays.

   Example 2 [90% waxy potato + 10 corn starch]

1. Form a pre-gelled cellulose paper-waxy potato starch suspension.

   5.8 g waxy potato starch [Eliane 100]: 8.3%
   6.5 g virgin cellulose pulp: 9.2%
   58 g water: 82.5%

2. Premix the following materials:

   29 g waxy potato starch [Eliane 100]
3.2 g corn starch

0.125 g magnesium stearate

0.025 g foaming agent
to form a homogeneous mixture

3. Add homogenous mixture to the pre-gelled paper potato starch, mix with a dough hook mixer on low speed. This mixture is stable and can be cooled or refrigerated, but not frozen.

4. Place mixture into mold (about 50-55 g) and bake at 195-225°C (ideal 215°C) for 60-90 seconds (ideal 75°C)

Example 2 is preferably used for deeper trays that require high edge strength.

Example 3 [80% waxy potato + 20% corn starch]

1. Form a pre-gelled cellulose paper-waxy potato starch suspension.

5.8 g waxy potato starch [Eliane 100]: 8.3%

6.5 g virgin cellulose pulp: 9.2%

58 g water: 82.5%

2. Premix the following materials:

22.5 g waxy potato starch [Eliane 100]

9.7 g corn starch

0.125 g magnesium stearate

0.025 g foaming agent
to form a homogenous mixture
3. Add homogenous mixture to the pre-gelled paper potato starch, mix with a dough hook mixer on low speed. This mixture is stable and can be cooled or refrigerated, but not frozen.

4. Place mixture into mold (about 50-55 g) and bake at 195-225°C (ideal 215°C) for 60-90 seconds (ideal 75°C).

Example 3 is preferably used for trays that need to be over wrapped or edge sealed.

Example 4 [70% waxy potato + 30% corn starch]

1. Form a pre-gelled cellulose paper-waxy potato starch suspension.
   
   5.8 g waxy potato starch [Eliane 100]: 8.3%
   6.5 g pre-paper cellulose pulp: 9.2%
   58 g water: 82.5%

2. Premix the following materials:
   
   19.4 g of waxy potato starch [Eliane 100]
   13.1 g of corn starch
   0.125 g magnesium stearate
   0.025 g foaming agent

   to form a homogenous mixture

3. Add homogenous mixture to the pre-gelled paper potato starch, mix with a dough hook mixer on low speed. This mixture is stable and can be cooled or refrigerated, but not frozen.

4. Place mixture into mold (about 50-55 g) and bake at 195-225°C (ideal 215°C) for 60-90 seconds (ideal 75°C).
Example 4 is preferably used for deeper trays or salad/soup type bowls requiring high edge strength.

Example 5 [wood fiber formula]

1. Form a pregelled cellulose paper-waxy potato starch suspension.

   58 g waxy potato starch [Eliane 100]
   65 g virgin cellulose pulp
   580 g water

2. Premix the following materials:

   163 g waxy potato starch [Eliane 100]
   159 g corn starch
   26 g wood flour #4025
   0.5 g magnesium stearate
   0.05 g foaming agent

   to form a homogenous mixture

3. Add homogenous mixture to the pregelled paper potato starch, mix with a dough hook mixer on low speed. This mixture is stable and can be cooled or refrigerated, but not frozen.

4. Place mixture into mold (about 50-55 g) and bake at 195-225°C (ideal 215°C) for 60-90 seconds (ideal 75°C)

Example 6 [20% cornstarch using powdered cellulose formula 1]
1. Form a pregelled cellulose paper-waxy potato starch suspension.

   21.75 g waxy potato starch [Eliane 100]

   24.37 g International Fiber 1178 cellulose pulp

   217.5 g water

2. Premix the following materials:

   96.75 g waxy potato starch [Eliane 100]

   24.0 g corn starch

   0.9375 g magnesium stearate

   0.1875 g foaming agent

   to form a homogenous mixture

3. Add homogenous mixture to the pregelled paper potato starch, mix with a dough hook mixer on low speed. This mixture is stable and can be cooled or refrigerated, but not frozen.

4. Place mixture into mold (about 50-55 g) and bake at 195-225°C (ideal 215°C) for 60-90 seconds (ideal 75°C)

Example 7 [Arbocell 1000 cellulose powder formula 2]

1. Form a pregelled cellulose paper-waxy potato starch suspension.

   58 g waxy potato starch [Eliane 100]

   65 g Arbocell 1000 cellulose powder

   580 g water

2. Premix the following materials:
322 g waxy potato starch [Eliane 100]

0.5 g magnesium stearate

0.05 g foaming agent

to form a homogenous

3. Add homogenous mixture to the pregelled paper potato starch, mix with a dough hook mixer on low speed. This mixture is stable and can be cooled or refrigerated, but not frozen.

4. Place mixture into mold (about 50-55 g) and bake at 195-225°C (ideal 215°C) for 60-90 seconds (ideal 75°C)

Example 8 [flash gel preparation of a small batch of dough made from 100% amylopectin]:

Pre-gel

1160 gm of water at 68 deg C

130 gm of native cellulose fiber [International Fiber Corp IFCT 1178 or equivalent]

116 gm of waxy potato starch [Eliane 100 starch, Avebe Veendam, Holland]

Add to pre-gel to form homogenous moldable composition

644 gm waxy potato starch [Eliane 100 starch, Avebe Veendam, Holland]

5 gm of mold release [Magnesium Sterate]

1 gm of foaming agent [Clariant Masterbatches CT 1480]

Step 1 - The water is introduced into the preheated [to 68 deg C] mixer such as a Kitchen Aid Classic fitted with the paddle blade in a stainless steel bowl. The fiber is quickly added with mixing [setting # 1]. As soon as the fiber is well wetted the gelling starch is added. The gel will form within 60 seconds. Continue mixing for an additional 120 seconds.
Step 2 - Now add the mold release and the foaming agent [pre blended], with mixing at setting #1. The balance of the starch is added slowly until the mixture is homogenous. Continue mixing for an additional four minutes. At this time the dough will be smooth and slightly sticky. The mixed dough is transferred into a sealed transport container and product produced.

Example 9. [flash gel preparation of a small batch of dough made from amylopectin starch and corn starch]

Pre-gel

1160 gm of water at 65 deg C

130 gm of native cellulose fiber [International Fiber Corp IFCT 1178 or equivalent]

116 gm of amylopectin [Eliane 100 starch, Avebe Veendam, Holland]

Add to pre-gel to form homogenous moldable composition

516 gm amylopectin [Eliane 100 starch, Avebe Veendam, Holland]

128 gm native corn starch

5 gm of mold release [Magnesium Sterate]

1 gm of foaming agent [Clariant Masterbatches CT 1480]

Step 1 - The water is introduced into the preheated [to 65 deg C] mixer such as a Kitchen Aid Classic fitted with the paddle blade in a stainless steel bowl. The fiber is quickly added with mixing [setting #1]. As soon as the fiber is well wetted the gelling starch is added. The gel will form within 60 seconds. Continue mixing for an additional 120 seconds.

Step 2 - Now add the mold release and the foaming agent [pre blended], with mixing at setting #1. The balance of the starches are added [pre blended] slowly until the mixture is homogenous. Continue mixing for an additional four minutes. At this time the dough will be
smooth and slightly sticky. The mixed dough is transferred into a sealed transport container and product produced.

**Example 10** [flash gel preparation of a large batch of dough made from 100% amyllopectin starch]

**Pre-gel**

1.16 kg of water at 68 deg C

1.30 kg of native cellulose fiber [International Fiber Corp IFCT 1178 or equivalent]

1.16 kg of amyllopectin [Eliane 100 starch, Avebe Veendam, Holland]

Add to pre-gel to form homogenous mixture

6.44 kg amyllopectin [Eliane 100 starch, Avebe Veendam, Holland]

50gm of mold release [Magnesium Sterate]

10 gm of foaming agent [Clariant Masterbatches CT 1480]

Step 1 - The water is introduced into the preheated [to 68 deg C] large capacity mixer such as a Varimixer Bear AR40 [A/S Wodscow & Co of Denmark] fitted with a twin fin paddle blade and scraper in a stainless steel bowl. The fiber is quickly added with mixing [setting # 1.5]. As soon as the fiber is well wetted the gelling starch is added. The gel will form in 60 seconds. Continue mixing for an additional 120 seconds.

Step 2 - Now add the mold release and the foaming agent [pre blended], with mixing at setting # 1.5. The balance of the starch is added slowly until the mixture is homogenous. Continue mixing for an additional four minutes. At this time the dough will be smooth and slightly sticky. The mixed dough is transferred into a sealed transport container and product produced.
Example 1 [flesh gel preparation of a large batch of dough made from amylopectin starch and corn starch]

Pre-gel

1.16 kg of water at 65 deg C

1.30 kg of native cellulose fiber [International Fiber Corp IFCT 1178 or equivalent]

1.16 kg of amylopectin [Eliane 100 starch, Avebe Veendam, Holland]

Add to pre-gel to form homogenous moldable composition

5.16 kg amylopectin [Eliane 100 starch, Avebe Veendam, Holland]

1.28 kg of native corn starch

50 gm of mold release [Magnesium Sterate]

10 gm of foaming agent [Clariant Masterbatches CT 1480]

Step 1 - The water is introduced into the preheated [to 65 deg C] mixer such as a Varimixer Bear AR40 [A/S Wodscow & Co of Denmark] fitted with a twin fin paddle blade and scraper in a stainless steel bowl. The fiber is quickly added with mixing [setting # 1.5]. As soon as the fiber is well wetted the gelling starch is added. The gel will form within 60 seconds. Continue mixing for an additional 120 seconds.

Step 2 - Now add the mold release and the foaming agent [pre blended], with mixing at setting #1.5. The balance of the starches are added [pre blended] slowly until the mixture is homogenous. Continue mixing for an additional four minutes. At this time the dough will be smooth and slightly sticky. The mixed dough is transferred into a sealed transport container and product produced.
Example 12 [High % Tapioca starch]

1. Form a pre-gelled cellulose fiber-waxy potato starch suspension using the Flash Gel method.

   51 g waxy potato starch [Eliane 100]: 8.1%

   65 g virgin cellulose pulp: 10.4.2%

   510 g Water: 81.5%

Mix fiber into preheated water, at 70-85 deg C and mix high speed with a paddle blade. When the fiber is well dispersed add gelling starch and mix for 1-3 min. The final temperature will be 64-66 deg C. Alternatively, the paper pulp and gelling starch are dispersed into warm water. The mixing bowl is heated. The RPM of the mixer is set at the first setting (KitchenAid®). The heating continues until the temp reaches 650C. At this time, the mixture is a homogeneous gel suspension. Once gelled, the gel may be used hot, cooled or refrigerated (but not frozen) until used.

2. Premix the following materials:

   163 grams of native tapioca starch

   159 grams of native corn starch

   2.5 g magnesium stearate

   0.5 g foaming agent

   to form a homogenous mixture

3. Add homogenous mixture to the pre-gelled fiber-starch, mix low speed until blended and for ten additional min to condition the dough mix. This mixture is stable and can be molded hot or stored by refrigeration, but not frozen.

4. Place mixture into mold (about 45-50 g) and bake at 195-225°C (ideal 215°C) for 60-90 seconds (ideal 70 seconds).
Example 12 is preferably used for flatter trays with good flexibility and moderate crush strength.

Example 13 [Tapioca starch-diatomaceous earth mix]

1. Form a pre-gelled cellulose fiber-waxy potato starch suspension using the Flash Gel method.

   51 g waxy potato starch [Eliane 100]: or native potato starch 8.1%

   65 g virgin cellulose pulp: 10.4.2%

   510 g Water: 81.5%

2. Mix fiber into preheated water, at 70-85 deg C and mix high speed with a paddle blade. When the fiber is well dispersed add gelling starch and mix for 1-3 min. The final temperature will be 64-66 deg C. Alternatively, the paper pulp and gelling starch are dispersed into warm water. The mixing bowl is heated. The RPM of the mixer is set at the first setting (KitchenAid®). The heating continues until the temp reaches 65°C. At this time, the mixture is a homogeneous gel suspension. Once gelled, the gel may be used hot, cooled or refrigerated (but not frozen) until used.

2. Premix the following materials:

   163 grams of native tapioca starch

   55 grams diatomaceous earth [Celite]

   2.5 g magnesium stearate

   0.5 g foaming agent

   to form a homogenous mixture
3. Add homogenous mixture to the pre-gelled fiber-starch, mix low speed until blended and for ten additional min to condition the dough mix. This mixture is stable and can be molded hot or stored by refrigeration, but not frozen.

4. Place mixture into mold (about 45-50 g) and bake at 195-225°C (ideal 215°C) for 60-90 seconds (ideal 65 seconds).

Example 13 is preferably used for flatter trays with high flexibility and moderate crush strength.

Example 14 [Tapioca starch-diatomaceous earth mix]

1. Form a pre-gelled cellulose fiber-waxy potato starch suspension using the Flash Gel method.

   51 g native potato starch: 7.6%

   65 g virgin cellulose pulp: 9.7%

   4.75 g soy oil: 0.7%

   550 g Water: 82%

Mix fiber and oil into preheated water, at 70-85 deg C and mix high speed with a paddle blade. When the fiber is well dispersed add gelling starch and mix for 1-3 min. The final temperature will be 64-66 deg C. Alternatively, the paper pulp and gelling starch are dispersed into warm water. The mixing bowl is heated. The RPM of the mixer is set at the first setting (KitchenAid®). The heating continues until the temp reaches 65°C. At this time, the mixture is a homogeneous gel suspension. Once gelled, the gel may be used hot, cooled or refrigerated (but not frozen) until used.

2. Premix the following materials:

   163 grams of native tapioca starch
55 grams diatomaceous earth [Celite]

0.37 grams citric acid

2.5 g magnesium stearate

0.5 g foaming agent
to form a homogenous mixture

3. Add homogenous mixture to the pre-gelled fiber-starch, mix low speed until blended and for ten additional min to condition the dough mix. This mixture is stable and can be molded hot or stored by refrigeration, but not frozen.

4. Place mixture into mold (about 45-50 g) and bake at 195-225°C (ideal 215°C) for 60-90 seconds (ideal 65 seconds).

Example 14 is preferably used for flatter trays with high flexibility and moderate crush strength with a smooth finish.

Example 15 [Potato starch-diatomaceous earth mix]

1. Form a pre-gelled cellulose fiber-waxy potato starch suspension using the Flash Gel method.

<table>
<thead>
<tr>
<th>Component</th>
<th>Percentage</th>
</tr>
</thead>
<tbody>
<tr>
<td>51 g native potato starch</td>
<td>7.6%</td>
</tr>
<tr>
<td>65 g virgin cellulose pulp</td>
<td>9.7%</td>
</tr>
<tr>
<td>4.75 g soy oil</td>
<td>0.7%</td>
</tr>
<tr>
<td>550 g Water</td>
<td>82%</td>
</tr>
</tbody>
</table>

Mix fiber and oil into preheated water, at 70-85 deg C and mix high speed with a paddle blade. When the fiber is well dispersed add gelling starch and mix for 1-3 min. The final
The temperature will be 64-66 deg C. Alternatively, the paper pulp and gelling starch are dispersed into warm water. The mixing bowl is heated. The RPM of the mixer is set at the first setting (KitchenAid®). The heating continues until the temperature reaches 65°C. At this time, the mixture is a homogeneous gel suspension. Once gelled, the gel may be used hot, cooled or refrigerated (but not frozen) until used.

2. Premix the following materials:

- 178 grams of native potato starch
- 55 grams diatomaceous earth [Celite]
- 0.37 grams citric acid
- 2.5 g magnesium stearate
- 0.5 g foaming agent
to form a homogenous mixture

3. Add homogenous mixture to the pre-gelled fiber-starch, mix low speed until blended and for ten additional min to condition the dough mix. This mixture is stable and can be molded hot or stored by refrigeration, but not frozen.

4. Place mixture into mold (about 45-50 g) and bake at 195-225°C (ideal 215°C) for 60-90 seconds (ideal 65 seconds).

Example 15 is preferably used for flatter trays with high flexibility and moderate crush strength with a smooth finish.

Example 16 [Tapioca starch-diatomaceous earth mix with added water repellant]

1. Form a pre-gelled cellulose fiber-waxy potato starch suspension using the Flash Gel method.
51 g native potato starch: 7.6%

65 g virgin cellulose pulp: 9.7%

4.75 g soy oil: 0.7%

550 g Water: 82%

5 Mix fiber and oil into preheated water, at 70-85 deg C and mix high speed with a paddle blade. When the fiber is well dispersed add gelling starch and mix for 1-3 min. The final temperature will be 64-66 deg C. Alternatively, the paper pulp and gelling starch are dispersed into warm water. The mixing bowl is heated. The RPM of the mixer is set at the first setting (KitchenAid®). The heating continues until the temp reaches 65°C. At this time, the mixture is a homogeneous gel suspension. Once gelled, the gel may be used hot, cooled or refrigerated (but not frozen) until used.

2. Premix the following materials:

   163 grams of native tapioca starch

   55 grams diatomaceous earth [Celite]

   10 grams calcium stearate

   0.37 grams citric acid

   2.5 g magnesium stearate

   0.5 g foaming agent

   to form a homogenous mixture

3. Add homogenous mixture to the pre-gelled fiber-starch, mix low speed until blended and for ten additional min to condition the dough mix. This mixture is stable and can be molded hot or stored by refrigeration, but not frozen.
4. Place mixture into mold (about 45-50 g) and bake at 195-225°C (ideal 215°C) for 60-90 seconds (ideal 65 seconds).

Example 16 is preferably used for flatter trays with high flexibility and moderate crush strength with a smooth more water repellant finish.

Example 17 [Diatomaceous earth-tapioca starch mix]

1. Form a pre-gelled cellulose fiber-waxy potato starch suspension using the Flash Gel method.

   51 g native potato starch: 7.6%

   65 g virgin cellulose pulp: 9.7%

   4.75 g soy oil: 0.7%

   550 g Water: 82%

Mix fiber and oil into preheated water, at 70-85 deg C and mix high speed with a paddle blade. When the fiber is well dispersed add gelling starch and mix for 1-3 min. The final temperature will be 64-66 deg C. Alternatively, the paper pulp and gelling starch are dispersed into warm water. The mixing bowl is heated. The RPM of the mixer is set at the first setting (KitchenAid®). The heating continues until the temp reaches 65°C. At this time, the mixture is a homogeneous gel suspension. Once gelled, the gel may be used hot, cooled or refrigerated (but not frozen) until used.

2. Premix the following materials:

   85 grams diatomaceous earth [Celite]

   81.5 grams of native tapioca starch

   0.37 grams citric acid
2.5 g magnesium stearate

0.5 g foaming agent
to form a homogenous mixture

3. Add homogenous mixture to the pre-gelled fiber-starch, mix low speed until blended and for ten additional min to condition the dough mix. This mixture is stable and can be molded hot or stored by refrigeration, but not frozen.

4. Place mixture into mold (about 55-60 g) and bake at 195-225°C (ideal 215°C) for 60-90 seconds (ideal 70 seconds).

Example 17 is preferably used for flatter trays which are highly flexible and dense exhibiting a very smooth finish.

Example 18 [Pea starch gelling mix]

1. Form a pre-gelled cellulose fiber-waxy pea starch suspension using the Flash Gel method.

5.1 g native pea starch: 7.6%

65 g virgin cellulose pulp: 9.7%

4.75 g soy oil: 0.7%

550 g Water: 82%

Mix fiber and oil into preheated water, at 70-85 deg C and mix high speed with a paddle blade. When the fiber is well dispersed add gelling starch and mix for 1-3 min. The final temperature will be 64-66 deg C. Alternatively, the paper pulp and gelling starch are dispersed into warm water. The mixing bowl is heated. The RPM of the mixer is set at the first setting (KitchenAid®). The heating continues until the temp reaches 65°C. At this time, the mixture is a homogeneous gel suspension. Once gelled, the gel may be used hot, cooled or refrigerated (but not frozen) until used.
2. Premix the following materials:

- 85 grams diatomaceous earth [Celite]
- 81.5 grams of native tapioca starch
- 0.37 grams citric acid
- 2.5 g magnesium stearate
- 0.5 g foaming agent

to form a homogenous mixture

3. Add homogenous mixture to the pre-gelled fiber-starch, mix low speed until blended and for ten additional min to condition the dough mix. This mixture is stable and can be molded hot or stored by refrigeration, but not frozen.

4. Place mixture into mold (about 55-60 g) and bake at 195-225°C (ideal 215°C) for 60-90 seconds (ideal 70 seconds).

Example 18 is preferably used for flatter trays which have high crush strength, good flexibility and a smooth finish.

Example 19 [304]

1. Form a pre-gelled cellulose fiber-waxy potato starch suspension using the Flash Gel method comprising the following:

- 58 grams waxy potato starch: 8.7%
- 26 grams wood flour: 3.9%
- 580 grams water: 87.3%

Mix flour into preheated water, at 70-85 deg C and mix high speed with a paddle blade. When the flour is well dispersed add gelling starch and mix for 1-3 min. The final temperature will be 64-66 deg C. Alternatively, the wood flour and gelling starch are dispersed into warm water. The mixing bowl is heated. The RPM of the mixer is set at the
first setting (KitchenAid®). The heating continues until the temp reaches 65°C. At this time, the mixture is a homogeneous gel suspension. Once gelled, the gel may be used hot, cooled or refrigerated (but not frozen) until used.

2. Premix the following materials:

- 159 grams native corn starch;
- 0.5 grams magnesium stearate; and
- 0.05 grams foaming agent

to form a homogenous mixture.

3. Add homogenous mixture to the pre-gelled flour-starch, mix low speed until blended and for ten additional min to condition the dough mix. This mixture is stable and can be molded hot or stored by refrigeration, but not frozen.

4. Place mixture into mold (about 55-60 g) and bake at 195-225°C (ideal 215°C) for 60-90 seconds (ideal 70 seconds).

Example 20 [3051]

1. Form a pre-gelled cellulose fiber-waxy potato starch suspension using the Flash Gel method comprising the following:

- 51 grams waxy potato starch
- 65 grams cellulose fiber
- 4.75 grams soy oil
- 560 grams water
Mix fiber and oil into preheated water, at 70-85 deg C and mix high speed with a paddle blade. When the fiber is well dispersed add gelling starch and mix for 1-3 min. The final temperature will be 64-66 deg C. Alternatively, the cellulose fiber, oil and gelling starch are dispersed into warm water. The mixing bowl is heated. The RPM of the mixer is set at the first setting (KitchenAid®). The heating continues until the temp reaches 650C. At this time, the mixture is a homogeneous gel suspension. Once gelled, the gel may be used hot, cooled or refrigerated (but not frozen) until used.

2. Premix the following materials:

172 grams native tapioca;

50 grams modified tapioca starch;

100 grams native potato starch;

2.5 grams magnesium stearate;

0.5 grams foaming agent; and

0.5 grams citric acid

to form a homogenous mixture.

3. Add homogenous mixture to the pre-gelled fiber-starch, mix low speed until blended and for ten additional min to condition the dough mix. This mixture is stable and can be molded hot or stored by refrigeration, but not frozen.

4. Place mixture into mold (about 55-60 g) and bake at 195-225°C (ideal 215°C) for 60-90 seconds (ideal 70 seconds).
Example 21 [306]

1. Form a pre-gelled cellulose fiber-waxy potato starch suspension using the Flash Gel method comprising the following:

   58 grams waxy potato starch
   65 grams cellulose fiber
   4.75 grams soy oil
   540 grams water

Mix fiber and oil into preheated water, at 70-85 deg C and mix high speed with a paddle blade. When the fiber is well dispersed add gelling starch and mix for 1-3 min. The final temperature will be 64-66 deg C. Alternatively, the cellulose fiber, oil and gelling starch are dispersed into warm water. The mixing bowl is heated. The RPM of the mixer is set at the first setting (KitchenAid®). The heating continues until the temp reaches 650C. At this time, the mixture is a homogeneous gel suspension. Once gelled, the gel may be used hot, cooled or refrigerated (but not frozen) until used.

2. Premix the following materials:

   107 grams native corn starch;
   107 grams native potato starch;
   42 grams diatomaceous earth;
   2.5 grams magnesium stearate;
   0.5 grams foaming agent; and

   to form a homogenous mixture.
3. Add homogenous mixture to the pre-gelled fiber-starch, mix low speed until blended and for ten additional min to condition the dough mix. This mixture is stable and can be molded hot or stored by refrigeration, but not frozen.

4. Place mixture into mold (about 55-60 g) and bake at 195-225°C (ideal 215°C) for 60-90 seconds (ideal 70 seconds).

Example 22 [307]

1. Form a pre-gelled cellulose fiber-waxy potato starch suspension using the Flash Gel method comprising the following:

- 51 grams waxy potato starch
- 65 grams cellulose fiber
- 525 grams water

Mix fiber into preheated water, at 70-85 deg C and mix high speed with a paddle blade. When the fiber is well dispersed add gelling starch and mix for 1-3 min. The final temperature will be 64-66 deg C. Alternatively, the cellulose fiber and gelling starch are dispersed into warm water. The mixing bowl is heated. The RPM of the mixer is set at the first setting (KitchenAid®). The heating continues until the temp reaches 65°C. At this time, the mixture is a homogeneous gel suspension. Once gelled, the gel may be used hot, cooled or refrigerated (but not frozen) until used.

2. Premix the following materials:

- 163 grams native tapioca;
- 50 grams diatomaceous earth;
- 2.5 grams magnesium stearate;
- 0.5 grams foaming agent; and
to form a homogenous mixture.

3. Add homogenous mixture to the pre-gelled fiber-starch, mix low speed until blended and for ten additional min to condition the dough mix. This mixture is stable and can be molded hot or stored by refrigeration, but not frozen.

4. Place mixture into mold (about 55-60 g) and bake at 195-225°C (ideal 215°C) for 60-90 seconds (ideal 70 seconds).

   Example 23 [308]

1. Form a pre-gelled cellulose fiber-waxy potato starch suspension using the Flash Gel method comprising the following:

   51 grams waxy potato starch

   65 grams cellulose fiber

   525 grams water

Mix fiber into preheated water, at 70-85 deg C and mix high speed with a paddle blade. When the fiber is well dispersed add gelling starch and mix for 1-3 min. The final temperature will be 64-66 deg C. Alternatively, the cellulose fiber and gelling starch are dispersed into warm water. The mixing bowl is heated. The RPM of the mixer is set at the first setting (KitchenAid®). The heating continues until the temp reaches 65°C. At this time, the mixture is a homogeneous gel suspension. Once gelled, the gel may be used hot, cooled or refrigerated (but not frozen) until used.

2. Premix the following materials:

   130 grams native tapioca;

   75 grams native potato starch;

   45 grams diatomaceous earth
5.0 grams magnesium stearate;

0.5 grams foaming agent; and

to form a homogenous mixture.

3. Add homogenous mixture to the pre-gelled fiber-starch, mix low speed until blended and for ten additional min to condition the dough mix. This mixture is stable and can be molded hot or stored by refrigeration, but not frozen.

4. Place mixture into mold (about 55-60 g) and bake at 195-225°C (ideal 215°C) for 60-90 seconds (ideal 70 seconds).

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Example 24 [309]

1. Form a pre-gelled cellulose fiber-waxy potato starch suspension using the Flash Gel method comprising the following:

   58 grams waxy potato starch

   65 grams cellulose fiber

   4.75 grams soy oil

   560 grams water

Mix fiber and oil into preheated water, at 70-85 deg C and mix high speed with a paddle blade. When the fiber is well dispersed add gelling starch and mix for 1-3 min. The final temperature will be 64-66 deg C. Alternatively, the cellulose fiber, oil and gelling starch are dispersed into warm water. The mixing bowl is heated. The RPM of the mixer is set at the first setting (KitchenAid®). The heating continues until the temp reaches 65°C. At this time, the mixture is a homogeneous gel suspension. Once gelled, the gel may be used hot, cooled or refrigerated (but not frozen) until used.

2. Premix the following materials:
50 grams modified tapioca starch;
215 grams native potato starch;
20 grams diatomaceous earth;
2.5 grams magnesium stearate;
0.5 grams foaming agent; and
0.23 grams citric acid
to form a homogenous mixture.

3. Add homogenous mixture to the pre-gelled fiber-starch, mix low speed until blended and for ten additional min to condition the dough mix. This mixture is stable and can be molded hot or stored by refrigeration, but not frozen.

4. Place mixture into mold (about 55-60 g) and bake at 195-225°C (ideal 215°C) for 60-90 seconds (ideal 70 seconds).

Example 25 [3101]

1. Form a pre-gelled cellulose fiber-waxy potato starch suspension using the Flash Gel method comprising the following:

   58 grams waxy potato starch
   100 grams cellulose fiber
   650 grams water

20 Mix fiber into preheated water, at 70-85 deg C and mix high speed with a paddle blade. When the fiber is well dispersed add gelling starch and mix for 1-3 min. The final temperature will be 64-66 deg C. Alternatively, the cellulose fiber gelling starch are
dispersed into warm water. The mixing bowl is heated. The RPM of the mixer is set at the first setting (KitchenAid®). The heating continues until the temp reaches 65°C. At this time, the mixture is a homogeneous gel suspension. Once gelled, the gel may be used hot, cooled or refrigerated (but not frozen) until used.

2. Premix the following materials:

- 300 grams native potato starch;
- 2.5 grams magnesium stearate; and
- 0.5 grams foaming agent;

to form a homogenous mixture.

3. Add homogenous mixture to the pre-gelled fiber-starch, mix low speed until blended and for ten additional min to condition the dough mix. This mixture is stable and can be molded hot or stored by refrigeration, but not frozen.

4. Place mixture into mold (about 55-60 g) and bake at 195-225°C (ideal 215°C) for 60-90 seconds (ideal 70 seconds).

Example 26 [311]

1. Form a pre-gelled cellulose fiber-waxy potato starch suspension using the Flash Gel method comprising the following:

- 51 grams waxy potato starch
- 65 grams cellulose fiber
- 560 grams water

Mix fiber into preheated water, at 70-85 deg C and mix high speed with a paddle blade. When the fiber is well dispersed add gelling starch and mix for 1-3 min. The final
temperature will be 64-66 deg C. Alternatively, the cellulose fiber and gelling starch are dispersed into warm water. The mixing bowl is heated. The RPM of the mixer is set at the first setting (KitchenAid®). The heating continues until the temp reaches 65°C. At this time, the mixture is a homogeneous gel suspension. Once gelled, the gel may be used hot, cooled or refrigerated (but not frozen) until used.

2. Premix the following materials:

- 56 grams modified tapioca starch;
- 159 grams native corn starch;
- 107 grams native potato starch; and
- 2.5 grams magnesium stearate;

...to form a homogenous mixture.

3. Add homogenous mixture to the pre-gelled fiber-starch, mix low speed until blended and for ten additional min to condition the dough mix. This mixture is stable and can be molded hot or stored by refrigeration, but not frozen.

4. Place mixture into mold (about 55-60 g) and bake at 195-225°C (ideal 215°C) for 60-90 seconds (ideal 70 seconds).

Example 27 [3121]

1. Form a pre-gelled cellulose fiber-waxy potato starch suspension using the Flash Gel method comprising the following:

- 51 grams waxy potato starch
- 65 grams cellulose fiber
- 4.75 grams soy oil
510 grams water

Mix fiber and oil into preheated water, at 70-85 deg C and mix high speed with a paddle blade. When the fiber is well dispersed add gelling starch and mix for 1-3 min. The final temperature will be 64-66 deg C. Alternatively, the cellulose fiber, oil and gelling starch are dispersed into warm water. The mixing bowl is heated. The RPM of the mixer is set at the first setting (KitchenAid®). The heating continues until the temp reaches 65°C. At this time, the mixture is a homogeneous gel suspension. Once gelled, the gel may be used hot, cooled or refrigerated (but not frozen) until used.

2. Premix the following materials:

- 322 grams native tapioca;
- 2.5 grams magnesium stearate; and
- 0.5 grams foaming agent;

...to form a homogenous mixture.

3. Add homogenous mixture to the pre-gelled fiber-starch, mix low speed until blended and for ten additional min to condition the dough mix. This mixture is stable and can be molded hot or stored by refrigeration, but not frozen.

4. Place mixture into mold (about 55-60 g) and bake at 195-225°C (ideal 215°C) for 60-90 seconds (ideal 70 seconds).

Example 28
Scale-up for Continuous Mixing

Methods for scaling up the above dough recipes for use with continuous mixers were developed. Scale-up, using continuous mixing can be achieved using, for example, the ReadCo LLC 2" paddle mixer (Readco Kurimoto, LLC, York, PA), which can produce 100 pounds of dough per hour. The 5" model is capable of producing up to 750 pounds per hour.
Continuous Production using a ReadCo 5" unit

Stage One - Pre-gel addition:

- Waxy potato starch at a rate of 17.34 kg/hour
- Cellulose fiber at a rate of 22.1 kg/hour
- Water at 69 °C +/- 1 °C at a rate of 190.4 kg/hour

Stage Two - Homogenous mixture addition:

- Tapioca starch at a rate of 55.42 kg/hour
- Corn starch at a rate of 54.1 kg/hour
- A mixture of magnesium stearate, foaming agent and citric acid at a rate of 1.15 kg/hour

The temperature of the produced dough is between 50 and 55 °C and is ready for molding or direct injection. The rate of dough produced is 340 +/- 5 kg/hour.

Example 29

Scale-up For Batch Processing

The scaled-up batch process can be achieved using, for example, the Hobart 140 quart mixer which can mix up to 300 kg of dough per hour using the flash-gel method.

Tables 1 and 2 list formulations prepared for patch processing using the Hobart 140 quart mixer.
Table 1 (Pre-Gel).

<table>
<thead>
<tr>
<th></th>
<th>299</th>
<th>300</th>
<th>301</th>
<th>302</th>
<th>303</th>
</tr>
</thead>
<tbody>
<tr>
<td>Waxy potato starch</td>
<td>55.42kg</td>
<td>5.1 kg</td>
<td>5.1 kg</td>
<td>5.1 kg</td>
<td>5.1 kg</td>
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<tr>
<td>cellulose fiber</td>
<td>54.06 kg</td>
<td>6.5 kg</td>
<td>6.5 kg</td>
<td>6.5 kg</td>
<td>6.5 kg</td>
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<tr>
<td>water</td>
<td>190.4 kg</td>
<td>56 kg</td>
<td>56 kg</td>
<td>56 kg</td>
<td>56 kg</td>
</tr>
<tr>
<td>soy oil</td>
<td>0</td>
<td>475 g</td>
<td>475 g</td>
<td>475 g</td>
<td>475 g</td>
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Table 2 (Homogenous Mixture)

<table>
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<th>Homogenous Mix</th>
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</thead>
<tbody>
<tr>
<td>native tapioca</td>
<td>16.3 kg</td>
<td>17.2 kg</td>
<td>16.3 kg</td>
<td>0</td>
<td>16.3 kg</td>
</tr>
<tr>
<td>modified tapioca</td>
<td>0</td>
<td>5.0 kg</td>
<td>0</td>
<td>5.6 kg</td>
<td>0</td>
</tr>
<tr>
<td>native corn</td>
<td>15.9 kg</td>
<td>0</td>
<td>15.9 kg</td>
<td>16.9 kg</td>
<td>0</td>
</tr>
<tr>
<td>native potato</td>
<td>0</td>
<td>10 kg</td>
<td>0</td>
<td>10.7 kg</td>
<td>0</td>
</tr>
<tr>
<td>diatomaceous earth</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>5.5 kg</td>
</tr>
<tr>
<td>magnesium stearate</td>
<td>250 g</td>
<td>250 g</td>
<td>250 g</td>
<td>250 g</td>
<td>250 g</td>
</tr>
<tr>
<td>foaming agent</td>
<td>50 g</td>
<td>50 g</td>
<td>50 g</td>
<td>50 g</td>
<td>50 g</td>
</tr>
<tr>
<td>citric acid</td>
<td>37 g</td>
<td>23 g</td>
<td>37 g</td>
<td>0</td>
<td>37 g</td>
</tr>
</tbody>
</table>
Homogenous moldable compositions were prepared according to the following steps:

1. Form a pre-gelled cellulose fiber-waxy potato starch suspension using the Flash Gel method. Mix fiber and oil into preheated water, at 70-85 deg C and mix high speed with a paddle blade. When the fiber is well dispersed add gelling starch and mix for 1-3 min. The final temperature will be 64-66 deg C. At this time, the mixture is a homogeneous gel suspension. Once gelled, the gel may be used hot, cooled or refrigerated (but not frozen) until used.

2. Add homogenous mixture to the pre-gelled fiber-starch, mix low speed until blended and for ten additional min to condition the dough mix. This mixture is stable and can be molded hot or stored by refrigeration, but not frozen.

3. Place mixture into mold (about 55-60 g) and bake at 195-225°C (ideal 215°C) for 60-90 seconds (ideal 70 seconds).
Example 30

Example 30 - Admet Testing of Formulas with and without Diatomaceous Earth

Table 3

<table>
<thead>
<tr>
<th>Ingredient</th>
<th>Example →</th>
<th>352</th>
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<tbody>
<tr>
<td>Flash Gel step one</td>
<td>Gm</td>
<td>Gm</td>
<td>Gm</td>
<td></td>
</tr>
<tr>
<td>Waxy potato starch</td>
<td>51</td>
<td>51</td>
<td>58</td>
<td></td>
</tr>
<tr>
<td>Cellulose fiber</td>
<td>65</td>
<td>65</td>
<td>65</td>
<td></td>
</tr>
<tr>
<td>Wood flour</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td></td>
</tr>
<tr>
<td>Water at 72deg C</td>
<td>550</td>
<td>510</td>
<td>560</td>
<td></td>
</tr>
<tr>
<td>Soy oil</td>
<td>4.75</td>
<td>0</td>
<td>4.75</td>
<td></td>
</tr>
</tbody>
</table>

Step two second addition

<table>
<thead>
<tr>
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<th>Example →</th>
<th>163</th>
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</tr>
</thead>
<tbody>
<tr>
<td>Native Tapioca starch</td>
<td>163</td>
<td>163</td>
<td>0</td>
<td></td>
</tr>
<tr>
<td>Modified Tapioca starch</td>
<td>0</td>
<td>0</td>
<td>32 3</td>
<td></td>
</tr>
<tr>
<td>Native corn starch</td>
<td>0</td>
<td>159</td>
<td>159</td>
<td></td>
</tr>
<tr>
<td>Native potato starch</td>
<td>0</td>
<td>0</td>
<td>131</td>
<td></td>
</tr>
<tr>
<td>Diatomaceous earth</td>
<td>55 1</td>
<td>0</td>
<td>0</td>
<td></td>
</tr>
<tr>
<td>Magnesium Stearate</td>
<td>2.5</td>
<td>2.5</td>
<td>2.5</td>
<td></td>
</tr>
<tr>
<td>Foaming agent 2</td>
<td>0.5</td>
<td>0.05</td>
<td>0.5</td>
<td></td>
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<tr>
<td>Citric Acid</td>
<td>0.37</td>
<td>0</td>
<td>0</td>
<td></td>
</tr>
</tbody>
</table>

1 = Celite® 545 (World Minerals); 3 = CT 1480 (Clariant Corp); 3 = P450 cross-linked Tapioca Starch (Mulit-Kem Ridgefield, NJ).

The above formulas were prepared according to the flash gel method and subjected to Admet testing to determine break strength and flexibility.

Test one is a break test for a tray clamped at 2/3 of its length and pressed down with the Admet Model 5600 Universal Tester until the tray fails. The computer calculates the peak load (in lbs) to break the tray, the amount of stress required to break the tray (in PSI) and
the amount of deflection before breakage (in inches deflected). The Peak Stress is indicative of strength and Deflection in indicative of flexibility.

### Table 4

<table>
<thead>
<tr>
<th>Example #</th>
<th>Peak Load (lbs)</th>
<th>Peak Stress (psi)</th>
<th>Deflection at break (ins)</th>
</tr>
</thead>
<tbody>
<tr>
<td>353 (base formula)</td>
<td>4.26</td>
<td>12.6</td>
<td>0.611</td>
</tr>
<tr>
<td>354 (add modified starch)</td>
<td>6.78</td>
<td>20.4</td>
<td>0.423</td>
</tr>
<tr>
<td>352 (with DE)</td>
<td>7.06</td>
<td>21</td>
<td>1.104</td>
</tr>
</tbody>
</table>

This data demonstrates that the base formula, #353 has moderate strength and flexibility. The addition of a modified (cross-linked) starch, #354, increases the strength, but reduces the flexibility. The formula with added diatomaceous earth (DE) and reduced starch content increases the strength and significantly increases the flexibility of the tray.

Test two is a break test for a tray supported upside down. A 2 inch disk is pressed down on the bottom of the upside down tray by the Admet Model 5600 Universal Tester until the tray bottom fails. The computer calculates the peak load (in lbs) to break the tray, the amount of stress required to break the tray (in PSI) and the amount of deflection before breakage (in inches deflected). The Peak Stress is indicative of strength and Deflection in indicative of flexibility.

### Table 5

<table>
<thead>
<tr>
<th>Example #</th>
<th>Peak load (lbs)</th>
<th>Peak Stress (psi)</th>
<th>Deflection at break (in)</th>
</tr>
</thead>
<tbody>
<tr>
<td>353 (base formula)</td>
<td>12.4</td>
<td>37</td>
<td>0.542</td>
</tr>
<tr>
<td>352 (with DE)</td>
<td>19.7</td>
<td>59</td>
<td>0.822</td>
</tr>
</tbody>
</table>

This data again demonstrates that the base formula, #353 has moderate strength and flexibility. The formula with added DE and reduced starch content increase the strength and significantly increases the flexibility of the tray.

This invention has been described with reference to various specific and exemplary embodiments and techniques. However, it should be understood that many variations and modifications will be obvious to those skilled in the art from the foregoing detailed
description of the invention and made be made while remaining within the spirit and scope of the invention.
CLAIMS

We claim:

1. A process for forming a biodegradable material comprising:
   a) forming a pre-gelled starch suspension by heating an amount of water to a temperature between 65-80°C and adding at least one starch, wherein the total amount of starch is 5-15% by weight of the pre-gel and wherein the remainder of the pre-gel comprises water;
   b) forming a homogenous mixture comprising one or more starches, a mold release agent and a foaming agent;
   c) adding to the pre-gel, the homogenous mixture to form a final homogenous moldable composition; and
   d) molding the homogenous moldable composition with heat to form the biodegradable material.

2. The process of claim 1, wherein the pre-gel comprises waxy potato starch.

3. The process of claim 1, wherein the pre-gel comprises native pea starch.

4. The process of claim 1, wherein the pre-gel further comprises 5-15% cellulose fibers or pulp.

5. The process of claim 1, wherein the pre-gel further comprises 5-10% paper pulp.

6. The process of claim 1, wherein the pre-gel further comprises 5-10% wood fibers or wood flour.

7. The process of claim 1, wherein the pre-gel further comprises a plant derived oil.

8. The process of claim 7, wherein the plant derived oil is soy oil.
9. The process of claim 1, wherein the homogenous mixture comprises one or more of potato starch, corn starch, or tapioca starch.

10. The process of claim 1, wherein the homogenous mixture comprises a native tapioca starch, a modified tapioca starch, and a native corn starch.

11. The process of claim 1, wherein the homogenous mixture further comprises diatomaceous earth.

12. The process of claim 4, wherein the diatomaceous earth is 1-60% by weight of the homogenous moldable composition.

13. The process of claim 1, wherein the homogenous mixture further comprises citric acid.

14. The process of claim 1, wherein the homogenous mixture further comprises wood fibers or wood flour having an aspect ratio between 1:2 and 1:8.

15. The process of claim 1, wherein the homogenous mixture further comprises one or more materials selected from the following:
   i) waxes, fatty alcohols, phospholipids or glycerol;
   ii) baking powder;
   iii) glycerol, clay , bentonite, kaolin, amorphous raw products, gypsum, calcium sulfate, minerals, limestone, or man made inert fillers; and
   iv) cellulose-based materials.

16. The process or claim 1 further comprising coating the biodegradable material with a liquid resistant coating.
17. A process for forming a biodegradable material comprising:
   a) forming a pre-gelled suspension comprising 5-15% starch by weight of the pre-
gelled suspension and wherein the remainder of the pre-gelled suspension is water;
   b) forming a homogenous mixture comprising tapioca starch and optionally an
   additional waxy or native starch, a mold release agent, and a foaming agent;
   c) adding the homogenous mixture to the pre-gelled suspension to form a final
   moldable homogenous compositions; and
   d) molding the homogenous moldable composition with heat to form a biodegradable
   material.

18. The process of claim 19, wherein the starch is a native potato starch, a waxy potato
    starch, or native pea starch.

19. The process of claim 19, wherein the pre-gelled suspension further comprises 5-10%
cellulose pulp or powder.

20. The process of claim 19, wherein the pre-gelled suspension further comprises a plant
    derived oil.

21. The process of claim 22, wherein the plant derived oil is soy oil.

22. The process of claim 19, wherein the additional native starch in the homogenous
    mixture is corn starch, potato starch, or a mixture thereof.

23. The process of claim 19, wherein the homogenous mixture further comprises diatomaceous earth.

24. The process of claim 19, wherein the homogenous mixture further comprises citric
    acid.
25. The process of claim 19, wherein the homogenous mixture further comprises wood fibers or wood flour having an aspect ratio between 1:2 and 1:8.

26. The process or claim 19 further comprising coating the biodegradable material with a liquid resistant coating.

27. A biodegradable, compostable material made according to the process of claim 1 or 21.

28. The material of claim 29, wherein the material is in the form of a cup, a tray, a bowl, a plate, a utensil, a coffee cup, a microwave dinner tray, or a "TV" dinner tray.
INTERNATIONAL SEARCH REPORT

International application No
PCT/US 09/34216

A CLASSIFICATION OF SUBJECT MATTER

<table>
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<th>IPC(8)</th>
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USPC - 106/162.5, 162.51

According to International Patent Classification (IPC) or to both national classification and IPC

B FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC(8) - C08L 300, C08L 5000 (2009.01) and USPC - 106/162 5, 162 51

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched
IPC(8) -C08S, B32B$ (2009 01) and USPC -- 264/1 01,232,300,31 9,330

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)
PubWest (PQP:BUSPT.USOCEPAB.JPAB), USPTO, Espacenet, Google Patents, Google Scholar, Google-please see extra sheet for Search Terms Used

C DOCUMENTS CONSIDERED TO BE RELEVANT

<table>
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<th>Citation of document, with indication, where appropriate, of the relevant passages</th>
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<td>US 2006/0255507 A1 (Bowden et al.) 16 November 2006 (16 11 2006) Abstract, para [0052], [0065], [0066], [0076], [0079], [0082], [0083], [0085], [0135],[0142], [0148], [0149],[0151], [0156], [0160], [0161],[0162],[0163],[0184]</td>
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D

[Box C continued]

* Special categories of cited documents
  *A* document defining the general state of the art which is not considered to be of particular relevance
  *E* earlier application or patent but published on or after the international filing date
  *L* document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
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Y document of particular relevance, the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

K document member of the same patent family

Date of the actual completion of the international search
20 March 2009 (20 03 2009)

Date of mailing of the international search report
2 MAR 2009

Name and mailing address of the ISA/US
Mail Stop PCT, Attn ISA/US, Commissioner for Patents
P O Box 1450, Alexandria, Virginia 22313-1450
Facsimile No 571-273-3201

Authorizing officer
Lee W Young

PCT Helpdesk 571-272-4300
PCT/ISP 571-272 7774

Form PCT/ISA/2 10 (second sheet) (April 2007)
Search Terms Used

ADDING STARCH, AQUEOUS, BIODEGRAD$, CELLULOSE, CENTIGRADE, CITRIC ACID, COMPOSTABS, DEGREE, FOAMING AGENT, GEL, GELLED, HEATED WATER, HEATING WATER, HOMOGENOUS, HOT WATER, INTRODUCING STARCH, MIX, MIXED, MIXING, MOLD, NATIVE, NATURAL, PEA, POTATO, PREGELS, PRE-GELS, REHEATING WATER, SOLUTION, STARCH ADDED, STARCH INTRODUCED, SOY, TAPIOCA, WATER TEMPERATURE, WAXY, WOOD FIBER, WOOD FLOUR