(51) International Patent Classification 5 : C08L 97/02

(11) International Publication Number: WO 94/21731

(21) International Application Number: PCT/US94/03165

(22) International Filing Date: 23 March 1994 (23.03.94)

(30) Priority Data:
08/035,436 24 March 1993 (24.03.93) US

(71) Applicant: ILLINOIS INSTITUTE OF TECHNOLOGY [US/US] ; 10 West 33rd Street, Chicago, IL 60616 (US).

(72) Inventors: SHUTOV, Fyodor; 755 74th Street, Downers Grove, IL 60516 (US). ARASTOOPOUR, Hamid; 1822 Gigi Lane, Darien, IL 60559 (US). IVANOV, George; Apartment 208, 2102 South Wabash Avenue, Chicago, IL 60616 (US).


Published
Without international search report and to be republished upon receipt of that report.

(54) Title: BIODEGRADABLE FILMS AND MOLDED PRODUCTS AND PROCESS FOR THEIR PRODUCTION

(57) Abstract

Biodegradable films and molded products are produced by molding natural particles and having less than ten percent synthetic binder. Preferred embodiments use particles of agricultural hulls having a range of sizes of less than 500µ and do not require any synthetic binder. The films and molded products which may be solid or foamed are especially useful for food packaging. Cups may be produced having a coffee aroma.
FOR THE PURPOSES OF INFORMATION ONLY

Codes used to identify States party to the PCT on the front pages of pamphlets publishing international applications under the PCT.

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>AT</td>
<td>Austria</td>
<td>GB</td>
<td>United Kingdom</td>
<td>MR</td>
<td>Mauritania</td>
</tr>
<tr>
<td>AU</td>
<td>Australia</td>
<td>GE</td>
<td>Georgia</td>
<td>MW</td>
<td>Malawi</td>
</tr>
<tr>
<td>BB</td>
<td>Barbados</td>
<td>GN</td>
<td>Guinea</td>
<td>NE</td>
<td>Niger</td>
</tr>
<tr>
<td>BE</td>
<td>Belgium</td>
<td>GR</td>
<td>Greece</td>
<td>NL</td>
<td>Netherlands</td>
</tr>
<tr>
<td>BF</td>
<td>Burkina Faso</td>
<td>HU</td>
<td>Hungary</td>
<td>NO</td>
<td>Norway</td>
</tr>
<tr>
<td>BG</td>
<td>Bulgaria</td>
<td>IE</td>
<td>Ireland</td>
<td>NZ</td>
<td>New Zealand</td>
</tr>
<tr>
<td>BJ</td>
<td>Benin</td>
<td>IT</td>
<td>Italy</td>
<td>PL</td>
<td>Poland</td>
</tr>
<tr>
<td>BR</td>
<td>Brazil</td>
<td>JP</td>
<td>Japan</td>
<td>PT</td>
<td>Portugal</td>
</tr>
<tr>
<td>BY</td>
<td>Belarus</td>
<td>KE</td>
<td>Kenya</td>
<td>RO</td>
<td>Romania</td>
</tr>
<tr>
<td>CA</td>
<td>Canada</td>
<td>KG</td>
<td>Kyrgyzstan</td>
<td>RU</td>
<td>Russian Federation</td>
</tr>
<tr>
<td>CF</td>
<td>Central African Republic</td>
<td>KP</td>
<td>Democratic People's Republic of Korea</td>
<td>SD</td>
<td>Sudan</td>
</tr>
<tr>
<td>CG</td>
<td>Congo</td>
<td>KR</td>
<td>Republic of Korea</td>
<td>SE</td>
<td>Sweden</td>
</tr>
<tr>
<td>CH</td>
<td>Switzerland</td>
<td>KZ</td>
<td>Kazakhstan</td>
<td>SI</td>
<td>Slovenia</td>
</tr>
<tr>
<td>CI</td>
<td>Côte d'Ivoire</td>
<td>LI</td>
<td>Liechtenstein</td>
<td>SK</td>
<td>Slovakia</td>
</tr>
<tr>
<td>CM</td>
<td>Cameroon</td>
<td>LK</td>
<td>Sri Lanka</td>
<td>SN</td>
<td>Senegal</td>
</tr>
<tr>
<td>CN</td>
<td>China</td>
<td>LU</td>
<td>Luxembourg</td>
<td>TD</td>
<td>Chad</td>
</tr>
<tr>
<td>CS</td>
<td>Czechoslovakia</td>
<td>LV</td>
<td>Latvia</td>
<td>TG</td>
<td>Togo</td>
</tr>
<tr>
<td>CZ</td>
<td>Czech Republic</td>
<td>MC</td>
<td>Monaco</td>
<td>TJ</td>
<td>Tajikistan</td>
</tr>
<tr>
<td>DE</td>
<td>Germany</td>
<td>MD</td>
<td>Republic of Moldova</td>
<td>TT</td>
<td>Trinidad and Tobago</td>
</tr>
<tr>
<td>DK</td>
<td>Denmark</td>
<td>MG</td>
<td>Madagascar</td>
<td>UA</td>
<td>Ukraine</td>
</tr>
<tr>
<td>ES</td>
<td>Spain</td>
<td>ML</td>
<td>Mali</td>
<td>US</td>
<td>United States of America</td>
</tr>
<tr>
<td>FI</td>
<td>Finland</td>
<td>MN</td>
<td>Mongolia</td>
<td>UZ</td>
<td>Uzbekistan</td>
</tr>
<tr>
<td>FR</td>
<td>France</td>
<td></td>
<td></td>
<td>VN</td>
<td>Viet Nam</td>
</tr>
</tbody>
</table>
BIODEGRADABLE FILMS AND MOLDED PRODUCTS
AND PROCESS FOR THEIR PRODUCTION

DESCRIPTION

Technical Field

This invention relates to biodegradable films
and molded products produced from agricultural products
using natural binders and processes for their production.
Fibers and molded products solid and/or foamed according
to this invention are especially suited for food
packaging and associated uses. Foams of materials of
this invention can be produced and are especially
suitable for use as containers for hot or cold beverages.
Products can be produced according to this invention
containing little or no synthetic materials.
Background Of The Invention

The use of cellulosic sugars and their hydrolysis products as natural adhesives in the manufacture of panel board is known. U. S. Patent No. 5,017,319 teaches production of a thermosetting binder by first decomposing and steam hydrolyzing the hemicellulosic portion of lignocellulose in materials such as rice husks followed by a second stage in which the water soluble resin is dried, and immediately in a third stage molded under heat and pressure and in the presence of other decomposition products from hemicellulose hydrolysis cross-links, polymerizes and thermosets. U.S. Patent No. 4,357,194 teaches a method similar to the 5,017,319 patent except that steam treatment during heating and pressing causes bonding of lignocellulose materials by hydrolysis of added sugars and/or starches together with lignin and/or other phenolic containing materials. U. S. Patent No. 4,627,951 teaches comminuting lignocellulosic material containing a high proportion of free sugars followed by drying to low moisture content, forming the dried mass and then molding it under high temperature and pressure utilizing the residual sugars from lignocellulosic materials as a bonding and bulking agent. U. S. Patent No. 4,933,125 teaches production of cellulosic fiber mat product by heat and pressure fusion of moisture containing fibers followed by purging moisture from cellulosic materials with no pressure and at below the critical temperature and then applying heat and pressure until the desired density and caliper of the fusion bonded product is achieved at above the critical temperature. A compressed band of wood fibers for fiber board production is obtained by screw pressing wood fiber having 18 to 20 percent moisture to liberate lignines which act as binder agent for the extruded piece is taught by U. S. Patent No. 3,903,229. Substitute wood products may be made according to U. S. Patent No. 4,496,718 by treatment of wet sawdust by a strong oxidizing agent to at least partially solubilize the
cellulosic material which with the addition of starches and sugars may be shaped and integrally bonded by the formed hydrophilic binder.

Products have been produced from natural materials using added materials as binders. U.S. Patent No. 4,810,446 teaches production of straw briquettes by mixing straw with up to 10 percent of a protein containing plant material as a binding agent which during compression at temperatures of 75° to 200°C causes coagulation of protein to bind the straw. U. S. Patent No. 3,686,384 teaches producing molded articles from coffee beans and rice hulls by dehydration followed by molding at temperatures of 232° to 260°C and pressure of 1/2 to 10 tons PSI for less than 30 seconds, whereby the resin component flows to form a resin based molded article. After dehydrating, 5 weight percent of a siliceous inhibitor and 5 weight percent of a synthetic cross-linking resin may be added for water proofing.

Attempts to obtain biodegradable containers and dishes having a sand base and a decomposable binder of, preferably, 20 to 60 percent grain fiber, 50 to 30 percent salt, 1 to 5 percent petroleum distillate and water is taught by U. S. Patent No. 5,108,677. The 5,108,677 patent teaches that water resistance is obtained by coating with a water resistant wax-like sealant.

Biodegradable blown films of gelatized starch and ethylene acrylic acid copolymer for packaging are taught by U. S. Patent No. 4,337,181.

Extraction of oils from vegetable oil bearing seeds and plants is known from U. S. Patent No. 5,077,071 which teaches mixing extracted oil with a second oil bearing material to form a slurry, adding water to the slurry and extracting product oil corresponding to the oil in the second oil bearing material.
Summary Of The Invention

The food industry, particularly carry-out fast food establishments, have the desire to substitute natural biodegradable containers for the synthetic polymer containers now used to avoid, or significantly reduce, the nuisance and unsightly appearance of such containers along roadsides and their long life in landfills. Prior attempts to reduce the synthetic binder contents of such containers below about fifteen percent, even when using natural powders as the main constituent, has not been successful.

This invention provides truly biodegradable solid or foamed molded products, films and various shapes such as cups, food containers, and the like. The biodegradable products of this invention have less than ten percent synthetic binder, preferred embodiments having no synthetic binder. Other preferred embodiments include biodegradable products which are also edible, particularly suitable for packaging agricultural products, such as animal feed. Still other embodiments include foamed biodegradable products particularly well suited for hot or cold cups or containers. Yet other embodiments include biodegradable products with a controlled aroma.

The process of this invention produces molded products based upon natural particles with less than ten percent and down to no synthetic binder, and preferable no synthetic binder. Natural oils originally contained in the natural particles or added separately serve to facilitate uniform distribution of particles and to enhance flow of particles into the mold as well as for binders in the products and process of this invention. Important aspects of the process of this invention are the use natural materials, such as agricultural wastes, particularly agricultural hulls, having a maximum particle size of about 500 μm and a broad particle size distribution. The natural particles are placed into a mold of the desired shape and heated to temperatures greater than about 80° C and subjected to mold pressures
of greater than about 0.25 metric tons per sq. cm. for a
time greater than about 2 minutes. Foamed products,
providing about 10 to about 50 percent lower density,
according to this invention may be obtained using no less
than 3 percent sodium and/or ammonium bicarbonate alone
or no less than 3 percent in admixture with citric acid
may be used as a blowing agent. The color and aroma of
the materials of this invention may be modified by
incorporation of natural colored and/or aromatic
particles, such as ground coffee beans, vanilla,
cinnamon, saffron, and the like.
Detailed Description Of The Preferred Embodiment

Natural particles from agricultural products and wastes are suitable for use in this invention. Suitable materials include hulls from a wide variety of agricultural products, such as corn, wheat, rice, soy and oat. The solid natural materials should be of small size, having a maximum particle size of 500 μ, and preferably have a particle size range from about 1 to about 250 μ. The particles should have a wide size distribution such that smaller particles occupy the space between larger particles, making compact packing of the particles. Furthermore, irregular shaped particles are preferred, especially fiber type particle shapes. The desired particle size and size distribution can be obtained by methods well known in the art, such as by grinding and screening.

The natural particles of desired particle size and size distribution are then placed in a heatable mold of the desired shape. It is helpful to add natural oil, in the amount of about 1 to about 5 weight percent, to the natural particles before or upon their introduction into the mold to aid in their flow and uniform distribution in the mold. The amount of oil needed to be added is dependent upon the amount of oil originally contained in the natural particles. Suitable natural oils include vegetable oils, such as corn and soy derived oils. The addition of natural oils also enhance the binding of the natural particles achieved under high temperature in the mold. Production of products of high strength may be achieved by varying the amount and type of natural oil added. It is especially desirably to utilize edible oils in the process and products of this invention since this will assure no detrimental effect upon humans and will enable and encourage consumption of such materials by animals. For example, animal feed may by packaged in containers of products according to this invention and the entire container consumed in a beneficial manner.
We have found it necessary to heat the mold to produce satisfactory products according to this invention. Preferred mold temperatures are about 120° to about 230° C. Mold pressures of about 0.25 to about 2.5 metric tons per sq. cm. are suitable to produce the fully biodegradable products of this invention, about 0.5 to about 1.25 metric tons per sq. cm. being preferred. Suitable times for application of the temperature and pressure in the mold depend upon the thickness of the film or molded article. The mold is allowed to cool and the product removed in finished form.

In a preferred embodiment, a first stage of high pressure and low temperature is applied to release the oil contained in the natural particles and uniformly distribute the particles in the mold and a second stage of continuously decreasing the pressure and at an increased temperature is applied for binding of the particles.

By the terminology "molding" as used herein, we mean to include pressure molding, injection molding and extrusion molding. Conventional machinery as known to the art for these types of molding may be used in the process of this invention.

The appearance and aroma of products produced according to this invention may be modified by addition of natural materials for such modification without affecting the desired full biodegradability of the products. For example, we have found that additional of ground coffee beans to coffee cups produced according to this invention provides the pleasant aroma of gourmet coffee. Likewise, other spices, such as ground cinnamon, vanilla, saffron or sugar, powdered milk and the like may be added. Less than about 30 volume percent, and preferably less than about 15 volume percent, of aroma and/or color modifiers should be used. The particle size of such additives should be in the same range as the base natural products used.

Foamed products which are fully biodegradable may be produced by the process of this invention as
described above by use of not less than about 3 and not more than about 15 weight percent sodium bicarbonate or ammonium bicarbonate or a mixture of sodium bicarbonate and/or ammonium bicarbonate and citric acid. Foamed products can be produced according to this invention having a density of about 10 to about 50 percent less than the non-foamed product. The foamed fully biodegradable products according to this invention having lower thermal conductivity are especially suited for use as containers for high temperature food or beverages.

The following examples are set forth using specific materials and process conditions to aid in the understanding of this invention and should not be considered to limit it in any way.

Example I
Thin bars, 62.0 x 12.5 x 2.0 mm, were produced from ground soy hulls. The natural particles were sized from 180μ to 425μ were fed to a mold heated to 120°C. A force of 7 metric tons was applied to the mold for 30 seconds following which the force was decreased to 2 metric tons and the temperature increased to 145°C and held at that temperature and pressure for 10 minutes. The mold was cooled to 60°C and the molded product removed. The molded product had a brown color with a smooth, hard surface and had sufficient strength to resist breaking between the fingers.

Example II
Similar bars to those described in Example I were produced from ground hominy particles sized to 180μ to 425μ. The natural particles were fed to a mold heated to 100°C and a force of 5.5 metric tons applied to the mold for 30 seconds. The force was then decreased to 3 metric tons and the temperature raised to 160°C and temperature and pressure maintained for 16 minutes. The mold was cooled to 55°C and the molded product removed. The molded product had a brown color with a smooth, hard
surface and had sufficient strength to resist breaking between the fingers.

Example III

Similar bars to those described in Example I were produced from ground rice hulls sized to 180μ to 425μ. The natural particles were fed to a mold heated to 130°C and a force of 8 metric tons was applied to the mold for 2 seconds. The force was decreased to 4 metric tons and the temperature increased to 165°C and temperature and pressure maintained for 10 minutes. The mold was cooled to 55°C and the molded product removed. The product had a brown color with a smooth, hard surface and had sufficient strength to resist breaking between the fingers.

Example IV

Similar bars to those described in Example I were produced from ground wheat hulls sized to 250μ to 425μ. The natural particles were fed to a mold at room temperature. The mold was heated to 130°C and a force of 10 metric tons was applied to the mold for 2 seconds. The force was decreased to 5 metric tons and the temperature increased to 180°C and held at that temperature and pressure for 21 minutes. The mold was cooled to 65°C and the product removed. The product had a brown color with a smooth, hard surface and had sufficient strength to resist breaking between the fingers.

Example V

The rate of water absorption by the bars produced according to Examples I - IV was determined by two testing procedures as set forth in ASTM D570. In the first test, the bars were placed in boiling water at 100°C for 30 minutes and were then placed in water at room temperature, 23°C, for an additional 15 minutes. In a second test, the bars were placed in water at about 90°C and allowed to gradually cool by surrounding air,
dropping to about 45°C after about 30 minutes. The second test closely simulates the conditions experienced by a container for hot beverages, such as coffee. The results are shown in Table 1.

<table>
<thead>
<tr>
<th>Material</th>
<th>Weight Percent Test 1</th>
<th>Water Absorbed Test 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>Soy</td>
<td>91</td>
<td>40</td>
</tr>
<tr>
<td>Hominy</td>
<td>89</td>
<td>40</td>
</tr>
<tr>
<td>Rice</td>
<td>90</td>
<td>59</td>
</tr>
<tr>
<td>Wheat</td>
<td>70</td>
<td>35</td>
</tr>
</tbody>
</table>

Example VI

The rate of heat transfer through the bars made in accordance with Examples I – IV was obtained by measuring the surface temperature rise for the samples when the opposite surface was in contact with a heating element maintained at 100°C. The results, together with comparison to polystyrene and polystyrene foam, are shown in Table 2.

<table>
<thead>
<tr>
<th>Material</th>
<th>Temperature °C after 5 Min.</th>
<th>10 Min.</th>
<th>15 Min.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rice</td>
<td>69</td>
<td>70</td>
<td>70</td>
</tr>
<tr>
<td>Wheat</td>
<td>66</td>
<td>67</td>
<td>67</td>
</tr>
<tr>
<td>Soy bean</td>
<td>65</td>
<td>66</td>
<td>66</td>
</tr>
<tr>
<td>Hominy</td>
<td>63</td>
<td>65</td>
<td>65</td>
</tr>
<tr>
<td>Polystyrene (solid)</td>
<td>54</td>
<td>55</td>
<td>55</td>
</tr>
<tr>
<td>Polystyrene Foam</td>
<td>46</td>
<td>47</td>
<td>48</td>
</tr>
</tbody>
</table>

It is seen from Table 2 that the foamed product can be expected to exhibit about 15 percent less heat transfer than the solid material.

Example VII

The break flexibility strength of the bars made in Examples I – IV was tested according to ASTM Method D790, Text Method 2, Procedure A and the results are shown in Table 3.
Table 3

<table>
<thead>
<tr>
<th>Material</th>
<th>Stress (psi)</th>
<th>Displacement (in)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hominy</td>
<td>1117</td>
<td>0.02652</td>
</tr>
<tr>
<td>Wheat</td>
<td>3478</td>
<td>0.04124</td>
</tr>
<tr>
<td>Rice</td>
<td>2190</td>
<td>0.04579</td>
</tr>
<tr>
<td>Soy</td>
<td>1797</td>
<td>0.0461</td>
</tr>
</tbody>
</table>

Example VIII

The shear modulus of the bars made in Examples I, II and IV was measured at 25°C and 100°C with the results shown in Table 4.

Table 4

<table>
<thead>
<tr>
<th>Material</th>
<th>Shear Modulus G (psi x 10^5)</th>
<th>G (100°C)</th>
<th>G (25°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>25°C</td>
<td>100°C</td>
<td></td>
</tr>
<tr>
<td>Wheat</td>
<td>1.90</td>
<td>1.10</td>
<td>0.58</td>
</tr>
<tr>
<td>Hominy</td>
<td>1.40</td>
<td>0.92</td>
<td>0.66</td>
</tr>
<tr>
<td>Soy</td>
<td>1.20</td>
<td>0.45</td>
<td>0.38</td>
</tr>
</tbody>
</table>

Example IX

Ground corn hulls (hominy flour) average sized 180μ were the only material placed in an "Engle" type injection molding machine with a heatable mold and subjected to a injection pressure of 1000Kg/cm² and a temperature of 180°C for a duration of injection molding of 8 minutes, producing discs having diameters of 90mm and 1 - 2 mm thickness. The product discs had a smooth, mirror-like surface and high shape stability.

Example X

A foamed product was produced using wheat hull particles sized to 250 to 425μ which were mixed in a ball mill with 12 weight percent powdered sodium bicarbonate for 30 minutes. The mixture was fed to a mold heated to 195°C. A force of 8 metric tons was applied to the mold for 2 minutes following which the force on the mold was released and the mold cavity increased by 1 mm by opening to atmospheric pressure. The mold was held in this condition for 16 minutes at 195°C. The mold was then cooled to 65°C and the molded product removed. The
molded foam product had a density of 15 percent less than the corresponding solid molded product.

In the foregoing specification this invention has been described in relation to certain preferred embodiments thereof, and many details have been set forth for the purpose of illustration, it will be apparent to those skilled in the art that the invention is susceptible to additional embodiments and that certain of the details described herein can be varied considerably without departing from the basic principles of the invention.
We claim:

1. A biodegradable film or molded product comprising less than 10 percent synthetic binder and natural particles having a range of sizes less than 500μ.

2. A biodegradable product according to Claim 1 wherein said natural particles are agricultural hulls selected from the group consisting of corn, wheat, rice, soy and mixtures thereof.

3. A biodegradable product according to Claim 1 additionally comprising about 0.5 to about 3 weight percent vegetable oil.

4. A biodegradable product according to Claim 3 wherein said oil is an edible oil selected from the group consisting of corn, soy, wheat, canola and mixtures thereof.

5. A biodegradable product according to Claim 1 wherein said particles have a size range within about 1 to about 250μ.

6. A biodegradable product according to Claim 1 which is edible.

7. A biodegradable product according to Claim 1 which is foamed.

8. A biodegradable product according to Claim 1 additionally comprising a natural color or aroma modifying substance.

9. A biodegradable product according to Claim 1 containing no synthetic binder.
10. A biodegradable product according to Claim 9 wherein said natural particles are agricultural hulls selected from the group consisting of corn, wheat, rice, soy and mixtures thereof.

11. A biodegradable product according to Claim 9 additionally comprising about 0.5 to about 3 weight percent vegetable oil.

12. A biodegradable product according to Claim 11 wherein said oil is an edible oil selected from the group consisting of corn, soy, wheat, canola and mixtures thereof.

13. A biodegradable product according to Claim 9 wherein said particles have a size range within about 1 to about 250μ.

14. A biodegradable product according to Claim 9 which is edible.

15. A biodegradable product according to Claim 9 which is foamed and has a density of about 10 to about 50 percent less than the corresponding solid material thereby providing a lower rate of thermal conductivity.

16. A biodegradable product according to Claim 9 additionally comprising a natural color or aroma modifying substance.

17. A biodegradable cup for hot or cold liquids consisting of natural materials comprising particles having a range of sizes less than about 500μ.

18. A biodegradable cup according to Claim 17 wherein said particles are agricultural hulls selected from the group consisting of corn, wheat, rice, soy and mixtures thereof.
19. A biodegradable cup according to Claim 17 additionally comprising about 0.5 to about 3 weight percent vegetable oil.

20. A biodegradable cup according to Claim 19 wherein said oil is an edible oil selected from the group consisting of corn, soy, wheat, canola and mixtures thereof.

21. A biodegradable cup according to Claim 17 wherein said particles have a size range within about 1 to about 250μ.

22. A biodegradable cup according to Claim 17 which is edible.

23. A biodegradable cup according to Claim 17 which is foamed.

24. A biodegradable cup according to Claim 23 additionally comprising a natural color or aroma modifying substance.

25. A biodegradable cup according to Claim 24 comprising a coffee aroma.

26. A process for producing a biodegradable film or molded product comprising, feeding natural particles having a range of sizes less than about 500μ to a mold, heating said mold to a temperature greater than about 80°C, applying mold pressure of greater than about 0.25 metric tons per square centimeter for a time greater than about 2 minutes while maintaining said temperature.

27. A process according to Claim 26 wherein said range of sizes is about 1 to about 250μ.
28. A process according to Claim 26 wherein said natural particles are agricultural hulls selected from the group consisting of corn, wheat, rice, soy and mixtures thereof.

29. A process according to Claim 26 wherein about 0.5 to about 3 weight percent vegetable oil is added to said film or mold.

30. A process according to Claim 26 wherein said oil is an edible oil selected from the group consisting of corn, soy, wheat, canola and mixtures thereof.

31. A process according to Claim 26 wherein said pressure is about 0.25 to about 2.5 metric tons per square centimeter.

32. A process according to Claim 26 wherein said pressure is about 0.5 to about 1.25 metric tons per square centimeter.

33. A process according to Claim 26 wherein said temperature is about 120°C to about 230°C.

34. A process according to Claim 26 wherein said heating and applying mold pressure is performed in two stages, a first stage at a high pressure and low temperature and a second stage at a lowered pressure and raised temperature as compared to said first stage.

35. A process according to Claim 26 wherein no synthetic binder is added to said film or mold.

36. A process according to Claim 26 wherein a foaming agent is added to said film or mold.
37. A process according to Claim 36 wherein said foaming agent is selected from the group consisting of sodium bicarbonate, ammonium bicarbonate, mixtures thereof and in combination with citric acid.

38. A process according to Claim 26 wherein a natural color or aroma modifying agent is added to said film or mold.

39. A process according to Claim 38 wherein said color or aroma modifying agent comprises coffee particles.

40. A biodegradable product according to Claim 1 in the form of a film.

41. A biodegradable product according to Claim 1 in the form of a molded article.