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(54) Title: METHOD FOR PREPARING POLYOLEFIN FOAMED PARTICLES HAVING EXCELLENT FORMABILITY

(57) Abstract: The present invention relates to a method of preparing polyolefin-type foamed particles having excellent formability by using a dispersion medium in which an organic compound containing fatty acids is dispersed. The method according to the present invention can prepare polyolefin-type foamed particles with excellent formability at a low temperature without the need for a washing step after foaming. Further, since it is possible to simply recover the dispersion medium used in the foaming step and reuse it in a new foaming process, the method of the present invention has the advantage of reducing the cost of wastewater treatment and preventing environmental contamination.



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## Description

# METHOD FOR PREPARING POLYOLEFIN FOAMED PARTICLES HAVING EXCELLENT FORMABILITY

### Technical Field

- [1] The present invention relates to an eco-friendly and economical method of preparing polyolefin-type foamed particles having excellent formability by foaming polyolefin-type resin particles in a recyclable dispersion medium in which organic compounds containing fatty acids are dispersed.

[2]

### Background Art

- [3] Molded products made of polyolefin-type foamed particles exhibit superior chemical resistance, impact resistance, heat resistance, and/or adiabatic property, while molded products made of polypropylene-type foamed particles exhibit superior heat resistance and can be used at high temperatures. As a result, these products have been widely used as paving materials having high load capacities or structural materials for automobiles having high mechanical strength.
- [4] Generally, methods of preparing polyolefin-type foamed particles are classified into two groups: methods of preparing foam in a stick form or a sheet form by using a common extruder and methods of preparing foam in a particle form by using an autoclave. The method using an extruder has the advantage of being capable of manufacturing a simple form of the product economically, but is problematic in that it is difficult to prepare a molded product in various forms required at the real industrial site. On the other hand, the method using an autoclave has the advantage of being capable of manufacturing a product in various forms by molding the prepared foamed particles and producing a product having physical properties more superior than those of an extrusion product and, as a result, has been widely used.
- [5] Recently, the demand for polyolefin foamed particles as structural material for automobiles has been greatly increasing, but the high-temperature steam required during the steps of preparing and molding the polyolefin-type foamed particles increases the production cost per unit, acting as a technical limitation on the use of polyolefin-type foamed particles. Accordingly, the development of a product which is moldable at a relatively low temperature, while reducing the unit production cost for the foamed particles, is needed. Further, the conventional method of preparing polyolefin-type foamed particles is problematic in that the water, typically used as a dispersion medium, cannot be recycled and, thus, must be disposed as wastewater, leading to environmental problems.

- [6] In order to overcome the above problems, efforts have been made to prepare foamed particles that are moldable at low temperatures or to minimize the generation of wastewater by reducing the amount of water for washing the prepared foamed particles.
- [7] For example, Japanese Patent No. 3560238 and Japanese Laid-Open Patent Publication No. 2003-201361 disclose methods of chemically modifying the surface of polyolefin-type resin particles by thermally degrading organic peroxides, which are added to a dispersion medium, and foaming the surface modified polyolefin-type resin particles, where the obtained polyolefin-type resin particles are moldable at low temperatures. In the above methods, however, the water, typically used as a dispersion medium, is inevitably contaminated with organic peroxides, and these organic peroxides are not reusable because they must be thermally degraded in order to carry out their function. Thus, this method is inappropriate for the purpose of reducing environmental contamination by preventing the generation of wastewater through the recovery and reuse of the dispersion medium.
- [8] Japanese Patent Laid-Open Publication Nos. 2000-290419 and 2001-164024 show efforts to reduce the generation of wastewater by decreasing the amount of wash water. Japanese Patent Laid-Open Publication No. 2000-290419 teaches a method of using hot water that contains, in a single molecule, functional groups capable of binding to the surface of inorganic compounds hardly soluble in water used as a dispersing agent, along with water-soluble compounds including hydrophilic functional groups, as wash water. While the above method adds to the wash water water-soluble compounds capable of binding to the dispersing agent and, as a result, does not demand a great quantity of wash water for removing the dispersing agent adhered to the surface of polyolefin-type foamed particles, it is not practically feasible to recycle the used wash water or dispersion medium. Further, Japanese Patent Laid-Open Publication No. 2001-164024 discloses a method which requires no washing step by using a dispersing agent together with a dispersion reinforcing agent. However, this means that it is possible to wash with water only, without using dilute hydrochloric acid or the like, indicating that the above method is inappropriate for reducing environmental contamination by preventing the generation of wastewater through the reuse of the recovered dispersion medium or for preparing foamed particles that are moldable without any washing step.
- [9] In view of the above, the present inventors have conducted extensive research to produce polyolefin-type foamed particles having excellent formability in a more economical manner while minimizing the generation of wastewater. As a result, the present inventors have found that, when the foaming is carried out in a dispersion medium in which an organic compound containing fatty acids is dispersed, it is

possible to mold the polyolefin-type foamed particles at a low temperature with no washing step and reduce the generation of wastewater through the reuse of the dispersion medium.

[10]

## **Disclosure of Invention**

### **Technical Problem**

[11] Accordingly, the objective of the present invention is to provide an eco-friendly and economical method of preparing polyolefin-type foamed particles having excellent formability, where the polyolefin-type foamed particles are capable of being molded at a low temperature without a washing step and the generation of wastewater is minimized through the reuse of the dispersion medium.

[12]

### **Technical Solution**

[13] One aspect of the present invention relates to a method of preparing polyolefin-type foamed particles having excellent formability, which comprises the steps of:

[14] 1) dispersing an organic compound containing fatty acids in a dispersion medium;

[15] 2) adding polyolefin-type resin particles and a foaming agent to said dispersion medium and heating resultant mixture to raise temperature, thereby preparing polyolefin-type foamed particles;

[16] 3) recovering the dispersion medium from said mixture, thereby obtaining the polyolefin-type foamed particles; and

[17] 4) reusing said recovered dispersion medium for preparing polyolefin-type foamed particles in step 2).

[18]

### **Advantageous Effects**

[19] The method of the present invention is capable of effectively preparing polyolefin-type foamed particles with excellent formability even if they are foamed at a low temperature or not subjected to a washing process after foaming, due to the use of a dispersion medium in which an organic compound containing fatty acids is dispersed. Further, in the method of the present invention, it is possible to reuse the dispersion medium used in the foaming process, preventing the generation of wastewater and environmental contamination, while reducing the production cost. As a result, the method of the present invention can be effectively used for economically producing polyolefin-type foamed particles in an environmentally favorable manner.

[20]

### **Best Mode for Carrying Out the Invention**

[21] The method of preparing polyolefin-type foamed particles having excellent

formability according to the present invention comprises the steps of:

- [22] 1) dispersing an organic compound containing fatty acids in a dispersion medium;
- [23] 2) adding polyolefin-type resin particles and a foaming agent to said dispersion medium and heating resultant mixture to raise temperature, thereby preparing polyolefin-type foamed particles;
- [24] 3) recovering the dispersion medium from said mixture, thereby obtaining the polyolefin-type foamed particles; and
- [25] 4) reusing said recovered dispersion medium for preparing polyolefin-type foamed particles in step 2).

[26] Each step of the above method is described in detail as follows.

[27] Step 1) is a step where an organic compound containing fatty acids is added to a dispersion medium and stirred for uniform dispersion (hereinafter referred to as the "dispersion step"), which is a technical feature of the present invention that has not yet been reported or attempted in the prior art. Because the fatty acids, which are uniformly dispersed in the dispersion medium, only affect the surface of the polyolefin-type resin particles used as a foaming raw material and do not change the unique properties of the particles, the polyolefin-type foamed particles are capable of being molded at a lower temperature, while still maintaining their superior properties, such as high chemical resistance, impact resistance, heat resistance, adiabatic property, and the like.

[28] In the conventional foaming method, when the surfactant, used as a dispersing agent and a dispersion-aiding agent, is added to the dispersion medium and foamed, the foamed particles need to be washed after the foaming process, with a large quantity of wash water, in particular, in order to improve the formability of the foamed particles. In the method of the present invention, however, the organic compound containing fatty acids that is dispersed in the dispersion medium has little influence on the molding of the foamed particles, and there is no need to carry out a washing step after the foaming process. Accordingly, the present invention has the advantage of being able to avoid environmental contamination due to the use of excessive wash water and also avoid wasting time and costs for carrying out the additional wastewater treatment.

[29] Suitable organic compounds for the above dispersion step may include unsaturated fatty acids such as linoleic acid and glycerides such as lecithin. Specifically, vegetable oils which are rich in linoleic acid and have a boiling point of 150°C or higher can be used, including, but not limited to, linseed oil, tung oil, safflower oil, soybean oil, castor oil, cottonseed oil, peanut oil, rapeseed oil, coconut oil, palm oil, olive oil, corn oil, corn germ oil, sesame oil, peach seed oil, peanut oil, soybean lecithin, and egg yolk lecithin. The suitable amount of the organic compound used in the above dispersion

step specifically ranges from 0.05% to 1% based on the weight of the dispersion medium, more specifically 0.1% to 0.5%.

[30] Typically, a suitable dispersion medium for the dispersion step is water, but a hydrophilic dispersion medium such as ethanol may also be used on its own or, alternatively, a mixture of water and a hydrophilic dispersion medium can be used in order to maintain equal densities of the dispersion medium and polyolefin resin particles and facilitate the stirring of the two.

[31] In Step 2), the dispersion medium prepared in the dispersion step where an organic compound containing fatty acids is dispersed, polyolefin-type resin particles, and a foaming agent are put into an autoclave and heated to achieve high temperature and high pressure, after which an outlet valve of the autoclave is opened to discharge the mixture of the dispersion medium, polyolefin-type resin particles, and foaming agent to outside of the autoclave where the pressure is lower than the inside of the autoclave, thereby preparing polyolefin-type foamed particles (hereinafter referred to as the "foaming step").

[32] In the method of the present invention, it is not necessary to carry out the dispersion step followed by the foaming step in order, and the two steps can be carried out at the same time. That is, the organic compound containing fatty acids, polyolefin-type resin particles, a foaming agent, and/or a dispersing agent are added to a dispersion medium at once and stirred, thereby simultaneously dispersing the above ingredients in the dispersion medium and carrying out the foaming step by heating.

[33] There is no limitation to the kind of polyolefin-type resins that can be used in the above foaming step, where any polyolefin-type resin well-known in the art can be used. Examples of well-known polyolefin-type resins may include, but are not limited to, polyethylenes, such as high density polyethylene, medium density polyethylene, low density polyethylene, linear low density polyethylene, linear ultra-low density polyethylene, and branched low density polyethylene; polypropylenes; polybutenes; and ethylene-propylene copolymers. Further, ethylenes, such as ethylene-propylene-1-butene tertiary copolymers and ethylene-propylene-butadiene copolymers; olefin monomers excluding propylene; and copolymers of ethylene and propylene can be used, while copolymers of non-olefin monomers, such as vinylacetate and styrene, and olefin monomers can also be used. The above copolymers include all types of copolymers, such as random copolymers, block copolymers, graft copolymers, and the like. Ideally, the copolymers contain more than 70% of propylene.

[34] Considering the period of time it takes for the foaming agent to penetrate into the resin and the size of the foamed particles, the polyolefin-type resin particles used for foaming should ideally have a diameter of 0.1 to 5  $\mu$ m. There is no limitation to the shape of the foamed particles, but ideally the foamed particles should be prepared to have a

spherical shape, since the foamed particles need to be molded in order to manufacture the desired product.

[35] Suitable foaming agents for the present invention may include, but are not limited to, volatile hydrocarbon foaming agents, such as propane, n-butane, iso-butane, and n-pentane, which can be used on their own or in admixture; inorganic gas foaming agents, such as carbon dioxide and nitrogen, which are free from the risk of an explosion; and mixed foaming agents of the above volatile foaming agents and inorganic foaming agents.

[36] Further, since the foaming step is carried out under conditions of high temperature and high pressure, a dispersing agent may be further added so as to prevent the polyolefin-type resin particles from aggregating. Suitable dispersing agents for the present invention include all types of organic and inorganic dispersing agents well-known in the art, as long as they are not dissolved in the dispersion medium and melt at high temperatures. Generally, it is ideal to use inorganic dispersion agents. Examples of such inorganic dispersing agents may include, but are not limited to, aluminum oxide, calcium carbonate, magnesium carbonate, potassium carbonate, zinc carbonate, calcium hydroxide, tricalcium phosphate, talc, and the like.

[37] Step 3) is a step where the dispersion medium and the polyolefin-type foamed particles are separated from the mixture of the dispersion medium, polyolefin-type foamed particles, foaming agent, and/or dispersion agent obtained in the foaming step, and the dispersion medium is recovered for reuse, thereby obtaining the polyolefin foamed particles (hereinafter referred to as the "recovery step"). The dispersion medium can be easily separated from the polyolefin-type foamed particles by means of the gravity difference, while the foaming agent included in the dispersion medium can be naturally removed, due to the decrease in solubility caused by the drastic change in pressure during the foaming step, and the dispersing agent can be easily removed by filtration using a suitable filter. Such filtration only removes the dispersion agent, where the organic compound containing fatty acids dispersed in the dispersion medium still remains, enabling the subsequent reuse of the dispersion medium in a new process of preparing foamed particles.

[38] The polyolefin-type foamed particles separated from the dispersion medium may be washed, dried, and molded, or alternatively, directly dried and molded without undergoing a washing step.

[39] Step 4) is a step where the dispersion medium recovered in the step 3) is reused without any further treatment in a new foaming process of preparing polyolefin-type foamed particles (hereinafter referred to as the "reuse step"), which is a technical feature of the present invention that has not yet been reported or attempted in the prior art. Since the organic compound containing fatty acids still remains in the dispersion

medium recovered in step 3), as long as the insufficient amounts of the dispersion medium and the corresponding organic compound containing fatty acids is supplemented, the recovered dispersion medium can be used in a new foaming step.

[40] In the method of preparing polyolefin-type foamed particles according to the present invention, the foaming step is carried out by using a dispersion medium in which an organic compound containing fatty acids is dispersed, making it possible to mold the foamed particles without a washing step and mold the foamed particles at a lower temperature than that used for foaming particles according to conventional methods. Further, it is possible to recover the dispersion medium used in the foaming process and reuse it in a new foaming process, preventing the generation of wastewater and environmental contamination, while reducing the cost of production. Therefore, the method of the present invention can be effectively used for economically producing polyolefin-type foamed particles in an environmentally favorable manner.

[41]

[42] The following examples are provided to illustrate the embodiments of the present invention in more detail, but are by no means intended to limit its scope.

[43]

[44] **Example 1**

[45] Ethylene-propylene random copolymer particles (melting point 143°C) were used as the polyolefin-type resin, iso-butane as the foaming agent, water as the dispersion medium, and lecithin as the organic compound containing fatty acids. The dispersion and foaming steps were simultaneously carried out by dispersing the rest of the ingredients in water and foaming.

[46]

[47] **<1-1> Dispersion and Foaming Steps**

[48] After 7  $\mu$ l of water, 7 g of lecithin, 0.7 g of basic magnesium carbonate and 3  $\mu$ l of the above ethylene-propylene random copolymer particles were added into a reactor, the air in the reactor was removed by a vacuum pump and 330 g of iso-butane was added as a foaming agent. The above mixture was stirred with a magnetic drive and heated up to 140°C so as to raise the pressure inside the reactor to 14.7  $\mu$ f/ $\mu$ . Under such conditions, the dispersion medium and the resin particles were discharged to the outside of the reactor under atmospheric pressure by opening an outlet valve of the reactor, thereby producing polyolefin-type foamed particles.

[49]

[50] **<1-2> Recovery Step**

[51] After the dispersion and foaming steps were completed, the polyolefin-type foamed particles and the dispersion medium in which lecithin was dispersed were separated from the reaction mixture, where the dispersion medium was used again at a

subsequent reuse step and the separated polyolefin-type foamed particles were washed with water and then dried in an oven at 60°C for 24 hours. In order to examine whether the dried polyolefin-type foamed particles were fused to each other, about 1 g of the foamed particles were randomly taken, and the number of particles that were attached to each other was counted with the naked eye. No attached particles were found, indicating that fusion between the foamed particles did not occur.

- [52] After keeping the dried polyolefin-type foamed particles at room temperature and under atmospheric pressure for 48 hours, they were pressurized under 3 MPa of pressure at room temperature for 24 hours, filled in a circular mold having a diameter of 9 mm and a depth of 2.5 mm, and molded at 135°C by heating with steam. After the molded product was dried in an oven at 60°C for 24 hours, the molded product was cut where the cross-section was observed in order to check the fused state of the foamed particles in the molded product. As a result, it was found that the foamed particles themselves were cut in 90% or more of the cross section, indicating that the foamed particles were successively fused to each other within the molded product.

[53]

[54] **<1-3> Reuse Step**

- [55] To the dispersion medium collected in the recovery step, the insufficient amounts of water and lecithin were added, where the resulting dispersion medium was reused for a new process for preparing polyolefin foamed particles.

[56]

[57] **Example 2**

- [58] Ethylene-propylene random copolymer particles (melting point 143°C) were used as the polyolefin-type resin, iso-butane as the foaming agent, water as the dispersion medium, and lecithin as the organic compound containing fatty acids. The dispersion and foaming steps were simultaneously carried out by dispersing the rest of the ingredients in water and foaming, according to the same procedure as described in Example <1-1>.

[59]

[60] **<2-1> Recovery Step**

- [61] After the dispersion and foaming steps were completed, the polyolefin-type foamed particles and dispersion medium in which lecithin was dispersed were separated from the reaction mixture, where the dispersion medium was used again at a subsequent reuse step and the separated polyolefin-type foamed particles were dried in an oven at 60°C for 24 hours, without washing. In order to examine whether the dried polyolefin-type foamed particles were fused to each other, about 1 g of the foamed particles were randomly taken, and the number of particles that were attached to each other was counted with the naked eye. No attached particles were found, indicating that fusion

between the foamed particles did not occur.

- [62] After keeping the dried polyolefin-type foamed particles at room temperature and under atmospheric pressure for 48 hours, they were pressurized under 3 MPa of pressure at room temperature for 24 hours, filled in a circular mold having a diameter of 9 mm and a depth of 2.5 mm, and molded at 135°C by heating with steam. After the molded product was dried in an oven at 60°C for 24 hours, the molded product was cut where the cross-section was observed in order to check the fused state of the foamed particles in the molded product. As a result, it was found that the foamed particles themselves were cut in 90% or more of the cross section, indicating that the foamed particles were successively fused to each other within the molded product.

[63]

[64] **<2-2> Reuse Step**

- [65] To the dispersion medium collected in the recovery step, the insufficient amounts of water and lecithin were added, where the resulting dispersion medium was reused for a new process for preparing polyolefin foamed particles.

[66]

[67] **Example 3**

- [68] Ethylene-propylene random copolymer particles (melting point 143°C) were used as the polyolefin-type resin, iso-butane as the foaming agent, water as the dispersion medium, and lecithin as the organic compound containing fatty acids. The dispersion and foaming steps were simultaneously carried out by dispersing the rest of the ingredients in water and foaming, as follows.

[69]

[70] **<3-1> Dispersion and Foaming Steps**

- [71] After 7 mm of the dispersion medium recovered from the <1-2> recovery step of Example 1, 0.7 g of basic magnesium carbonate and 3 mm of the above ethylene-propylene random copolymer particles were added into a reactor, the air in the reactor was removed by a vacuum pump and 330 g of iso-butane was added as a foaming agent. The above mixture was stirred with a magnetic drive and heated up to 140°C so as to raise the pressure inside the reactor to 14.7 MPa. Under such conditions, the dispersion medium and the resin particles were discharged to the outside of the reactor under atmospheric pressure by opening an outlet valve of the reactor, thereby producing polyolefin-type foamed particles.

[72]

[73] **<3-2> Recovery Step**

- [74] After the dispersion and foaming steps were completed, the polyolefin-type foamed particles and dispersion medium in which lecithin was dispersed were separated from the reaction mixture, where the dispersion medium was used again at a subsequent

reuse step and the foamed particles were washed with water and then dried in an oven at 60°C for 24 hours. In order to examine whether the dried polyolefin-type foamed particles were fused to each other, about 1 g of the foamed particles were randomly taken, and the number of particles that were attached to each other was counted with the naked eye. No attached particles were found, indicating that fusion between the foamed particles did not occur.

- [75] After keeping the dried polyolefin-type foamed particles at room temperature and under atmospheric pressure for 48 hours, they were pressurized under 3 MPa of pressure at room temperature for 24 hours, filled in a circular mold having a diameter of 9 mm and a depth of 2.5 mm, and molded at 135°C by heating with steam. After the molded product was dried in an oven at 60°C for 24 hours, the molded product was cut where the cross-section was observed in order to check the fused state of the foamed particles in the molded product. As a result, it was found that the foamed particles themselves were cut in 90% or more of the cross section, indicating that the foamed particles were successively fused to each other within the molded product.

[76]

[77] **<3-3> Reuse Step**

- [78] To the dispersion medium collected in the recovery step, the insufficient amounts of water and lecithin were added, where the resulting dispersion medium was reused for a new process for preparing polyolefin foamed particles.

[79]

[80] **Example 4**

- [81] Ethylene-propylene random copolymer particles (melting point 143°C) were used as the polyolefin-type resin, iso-butane as the foaming agent, water as the dispersion medium, and lecithin as the organic compound containing fatty acids. The dispersion and foaming steps were simultaneously carried out by dispersing the rest of the ingredients in water and foaming, as follows.

[82]

[83] **<4-1> Dispersion and Foaming Steps**

- [84] After 7 ml of the dispersion medium recovered from the <2-1> recovery step of Example 2, 27 g of calcium phosphate, and 3 ml of the above ethylene-propylene random copolymer particles were added into a reactor, the air in the reactor was removed by a vacuum pump and 330 g of iso-butane was added as a foaming agent. The above mixture was stirred with a magnetic drive and heated up to 140°C so as to raise the pressure inside the reactor to 15.4 MPa. Under such conditions, the dispersion medium and the resin particles were discharged to the outside of the reactor under atmospheric pressure by opening an outlet valve of the reactor, thereby producing polyolefin-type foamed particles.

[85]

[86]       **<4-2> Recovery Step**

[87]       After the foaming step was completed, the polyolefin-type foamed particles and dispersion medium in which lecithin was dispersed were separated from the reaction mixture, where the dispersion medium was used again at a subsequent reuse step and the foamed particles were washed with water and then dried in an oven at 60°C for 24 hours. In order to examine whether the dried polyolefin-type foamed particles were fused to each other, about 1 g of the foamed particles were randomly taken, and the number of particles that were attached to each other was counted with the naked eye. No attached particles were found, indicating that fusion between the foamed particles did not occur.

[88]       After keeping the dried polyolefin-type foamed particles at room temperature and under atmospheric pressure for 48 hours, they were pressurized under 3 MPa of pressure at room temperature for 24 hours, filled in a circular mold having a diameter of 9 mm and a depth of 2.5 mm, and molded at 135°C by heating with steam. After the molded product was dried in an oven at 60°C for 24 hours, the molded product was cut where the cross-section was observed in order to check the fused state of the foamed particles in the molded product. As a result, it was found that the foamed particles themselves were cut in 90% or more of the cross section, indicating that the foamed particles were successively fused to each other within the molded product.

[89]

[90]       **<4-3> Reuse Step**

[91]       To the dispersion medium collected in the recovery step, the insufficient amounts of water and lecithin were added, where the resulting dispersion medium was reused for a new process for preparing polyolefin foamed particles.

[92]

[93]       **Comparative Example 1**

[94]       Ethylene-propylene random copolymer particles (melting point 143°C) were used as the polyolefin-type resin, iso-butane as the foaming agent, water as the dispersion medium, where only a foaming step was carried out without going through any dispersion, recovery, and reuse steps, as follows.

[95]       After 7 mm of water, 5 g of sodium dodecylbenzene sulfonate, 21 g of calcium phosphate, and 3 mm of the above ethylene-propylene random copolymer particles were added into a reactor, the air in the reactor was removed by a vacuum pump and 340 g of iso-butane was added as a foaming agent. The above mixture was stirred with a magnetic drive and heated up to 144.3°C so as to raise the pressure inside the reactor to 22.3 MPa. Under such conditions, the dispersion medium and the resin particles were discharged to the outside of the reactor under atmospheric pressure by opening an

outlet valve of the reactor, thereby producing polyolefin-type foamed particles.

[96] Thus prepared polyolefin-type foamed particles were washed with water and then dried in an oven at 60°C for 24 hours. In order to examine whether the dried polyolefin-type foamed particles were fused to each other, about 1 g of the foamed particles were randomly taken, and the number of particles that were attached to each other was counted with the naked eye. No attached particles were found, indicating that fusion between the foamed particles did not occur.

[97] After keeping the dried polyolefin-type foamed particles at room temperature and under atmospheric pressure for 48 hours, they were pressurized under 3 MPa of pressure at room temperature for 24 hours, filled in a circular mold having a diameter of 9 mm and a depth of 2.5 mm, and molded at 140°C by heating with steam. After the molded product was dried in an oven at 60°C for 24 hours, the molded product was cut where the cross-section was observed in order to check the fused state of the foamed particles in the molded product. As a result, it was found that the foamed particles themselves were cut in 90% or more of the cross section, indicating that the foamed particles were successively fused to each other within the molded product.

[98] However, when compared with Examples 1 to 4, the foaming step in the above preparation process is carried out at a higher temperature and a higher pressure, and the molding step is also conducted at a temperature higher by about 5°C.

[99]

[100] **Comparative Example 2**

[101] Ethylene-propylene random copolymer particles (melting point 143°C) were used as the polyolefin-type resin, iso-butane as the foaming agent, water as the dispersion medium, where only a foaming step was carried out without going through any dispersion, recovery, and reuse steps, as follows.

[102] After 7 ml of water, 5 g of sodium dodecylbenzene sulfonate, 21 g of calcium phosphate, and 3 ml of the above ethylene-propylene random copolymer particles were added into a reactor, the air in the reactor was removed by a vacuum pump and 340 g of iso-butane was added as a foaming agent. The above mixture was stirred with a magnetic drive and heated up to 140°C so as to raise the pressure inside the reactor to 15 MPa. Under such conditions, the dispersion medium and the resin particles were discharged to the outside of the reactor under atmospheric pressure by opening an outlet valve of the reactor, thereby producing polyolefin-type foamed particles.

[103] Thus prepared polyolefin-type foamed particles were washed with water and then dried in an oven at 60°C for 24 hours. In order to examine whether the dried polyolefin-type foamed particles were fused to each other, about 1 g of the foamed particles were randomly taken, and the number of particles that were attached to each other was counted with the naked eye. No attached particles were found, indicating

that fusion between the foamed particles did not occur.

[104] After keeping the dried polyolefin-type foamed particles at room temperature and under atmospheric pressure for 48 hours, they were pressurized under 3 MPa of pressure at room temperature for 24 hours, filled in a circular mold having a diameter of 9 mm and a depth of 2.5 mm, and molded at 140°C by heating with steam. After the molded product was dried in an oven at 60°C for 24 hours, the molded product was cut where the cross section was observed in order to check the fused state of the foamed particles in the molded product. As a result, it was found that the foamed particles themselves were cut in only 30% of the cross section and the rest still remained in their original particle shape, indicating that the foamed particles were not successively fused to each other in the molded product.

[105] Accordingly, it was found that even if the mold temperature was raised to 140°C or higher, the polyolefin-type foamed particles prepared according to the conventional foaming process were not moldable into a desired shape, indicating extremely low formability.

[106]

[107] **Comparative Example 3**

[108] Ethylene-propylene random copolymer particles (melting point 143°C) were used as the polyolefin-type resin, iso-butane as the foaming agent, water as the dispersion medium, where only a foaming step was carried out without going through any dispersion, recovery, and reuse steps, as follows.

[109] After 7 ml of water, 5 g of sodium dodecylbenzene sulfonate, 21 g of calcium phosphate, and 3 ml of the above ethylene-propylene random copolymer particles were added into a reactor, the air in the reactor was removed by a vacuum pump and 340 g of iso-butane was added as a foaming agent. The above mixture was stirred with a magnetic drive and heated up to 144.5°C so as to raise the pressure inside the reactor to 15.8 MPa. Under such conditions, the dispersion medium and the resin particles were discharged to the outside of the reactor under atmospheric pressure by opening an outlet valve of the reactor, thereby producing polyolefin-type foamed particles.

[110] Thus prepared polyolefin-type foamed particles were washed with water and then dried in an oven at 60°C for 24 hours. In order to examine whether the dried polyolefin-type foamed particles were fused to each other, about 1 g of the foamed particles were randomly taken, and the number of particles that were attached to each other was counted with the naked eye. No attached particles were found, indicating that fusion between the foamed particles did not occur.

[111] After keeping the dried polyolefin-type foamed particles at room temperature and under atmospheric pressure for 48 hours, they were pressurized under 3 MPa of pressure at room temperature for 24 hours, filled in a circular mold having a diameter of 9 mm and

a depth of 2.5 mm, and molded at 140°C by heating with steam. After the molded product was dried in an oven at 60°C for 24 hours, the molded product was cut where the cross section was observed in order to check the fused state of the foamed particles in the molded product. As a result, it was found that the foamed particles themselves were cut in only 10% of the cross section and the rest still remained in their original particle shape, indicating that the foamed particles were not successively fused to each other in the molded product.

[112] From the above described Examples and Comparative Examples, it was confirmed that the method of preparing polyolefin-type foamed particles according to the present invention can mold the polyolefin foamed particles into a desired shape without a washing step even if the molding process is carried out at a lower temperature than that used in the conventional methods, and can reuse the dispersion medium recovered from the preceding foaming process without a reprocessing step.

[113] Although the invention has been described in detail for the purpose of illustration, it is understood that such detail is solely for that purpose, and variations can be made therein by those skilled in the art without departing from the spirit and scope of the invention which is defined by the following claims.

## Claims

- [1] A method of preparing polyolefin-type foamed particles having excellent formability which comprises the steps of:
- 1) dispersing an organic compound containing fatty acids in a dispersion medium;
  - 2) adding polyolefin-type resin particles and a foaming agent to said dispersion medium and heating resultant mixture to raise temperature, thereby preparing polyolefin-type foamed particles;
  - 3) recovering the dispersion medium from said mixture, thereby obtaining the polyolefin-type foamed particles; and
  - 4) reusing said recovered dispersion medium for preparing polyolefin-type foamed particles in step 2).
- [2] The method according to Claim 1, wherein the organic compound containing fatty acids in step 1) is a linoleic acid, a linoleic acid-containing vegetable oil, or lecithin.
- [3] The method according to Claim 2, wherein the organic compound containing fatty acids in step 1) is selected from the group consisting of linoleic acid, linseed oil, tung oil, safflower oil, soybean oil, castor oil, cottonseed oil, peanut oil, rapeseed oil, coconut oil, palm oil, olive oil, corn oil, corn germ oil, sesame oil, peach seed oil, peanut oil, soybean lecithin, and egg yolk lecithin.
- [4] The method according to Claim 1, wherein the dispersion medium in step 1) is water, ethanol, or a mixture thereof.
- [5] The method according to Claim 1, wherein the polyolefin resin in step 2) is selected from the group consisting of high density polyethylene, medium density polyethylene, low density polyethylene, linear low density polyethylene, linear ultra-low density polyethylene, branched low density polyethylene, polypropylene, polybutene, ethylene-propylene copolymers, copolymers of olefin monomer, ethylene, and propylene, and copolymers of non-olefin monomer and olefin monomer.
- [6] The method according to Claim 5, wherein the amount of propylene in the copolymers is 70% or more.
- [7] The method according to Claim 1, wherein the foaming agent in step 2) is a volatile hydrocarbon foaming agent selected from the group consisting of propane, n-butane, iso-butane, and n-pentane; an inorganic gas foaming agent selected from the group consisting of carbon dioxide and nitrogen; or a mixed foaming agent of the volatile foaming agent and inorganic foaming agent.
- [8] The method according to Claim 1, wherein step 2) further comprises adding a

dispersing agent to said dispersion medium.

- [9] The method according to Claim 8, wherein the dispersing agent is an organic dispersing agent or an inorganic dispersing agent.
- [10] The method according to Claim 9, wherein the inorganic dispersing agent is selected from the group consisting of aluminum oxide, calcium carbonate, magnesium carbonate, potassium carbonate, zinc carbonate, calcium hydroxide, tricalcium phosphate, and talc.
- [11] The method according to Claim 1, wherein step 1) is carried out simultaneously with step 2).
- [12] The method according to Claim 1, wherein the polyolefin-type foamed particles obtained in step 3) are dried and molded without undergoing a washing step.

**A. CLASSIFICATION OF SUBJECT MATTER***C08L 23/00(2006.01)i, C08L 23/26(2006.01)i*

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 23/00, C08L 23/26

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

eKIPASS; PAJ

**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

| Category* | Citation of document, with indication, where appropriate, of the relevant passages                            | Relevant to claim No. |
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| A         | JP 14-167460 A (JSP CORP) 11 June 2002<br>See the abstract; and the claims.                                   | 1-12                  |
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| A         | KR 10-2003-0083270 A (HOWTECH CORPORATION.,LTD.) 30 October 2003<br>See the abstract; and examples 1-5.       | 1-12                  |
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Further documents are listed in the continuation of Box C.



See patent family annex.

\* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

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"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"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

"&amp;" document member of the same patent family

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09 JUNE 2008 (09.06.2008)

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**INTERNATIONAL SEARCH REPORT**

Information on patent family members

International application No.

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