

(19) World Intellectual Property Organization
International Bureau



(43) International Publication Date
13 August 2009 (13.08.2009)

PCT

(10) International Publication Number
WO 2009/099482 A1

(51) International Patent Classification:

C08J 9/00 (2006.01) *C08L 25/12* (2006.01)
C08J 9/12 (2006.01) *C08L 23/00* (2006.01)
C08L 25/04 (2006.01)

(21) International Application Number:

PCT/US2008/086797

(22) International Filing Date:

15 December 2008 (15.12.2008)

(25) Filing Language:

English

(26) Publication Language:

English

(30) Priority Data:

61/026,486 6 February 2008 (06.02.2008) US

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(81) Designated States (*unless otherwise indicated, for every kind of national protection available*): AE, AG, AL, AM, AO, AT, AU, AZ, BA, BB, BG, BH, BR, BW, BY, BZ, CA, CH, CN, CO, CR, CU, CZ, DE, DK, DM, DO, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, GT, HN, HR, HU, ID, IL, IN, IS, JP, KE, KG, KM, KN, KP, KR, KZ, LA, LC, LK, LR, LS, LT, LU, LY, MA, MD, ME, MG, MK, MN, MW, MX, MY, MZ, NA, NG, NI, NO, NZ, OM, PG, PH, PL, PT, RO, RS, RU, SC, SD, SE, SG, SK, SL, SM, ST, SV, SY, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, ZA, ZM, ZW.

(84) Designated States (*unless otherwise indicated, for every kind of regional protection available*): ARIPO (BW, GH, GM, KE, LS, MW, MZ, NA, SD, SL, SZ, TZ, UG, ZM, ZW), Eurasian (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European (AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HR, HU, IE, IS, IT, LT, LU, LV, MC, MT, NL, NO, PL, PT, RO, SE, SI, SK, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, ML, MR, NE, SN, TD, TG).

Published:

— with international search report (Art. 21(3))

(54) Title: ARTICLE AND METHOD OF PRODUCING A LOW DENSITY FOAM BLEND OF STYRENIC POLYMER AND POLYOLEFIN

(57) Abstract: An extruded polymeric foam comprising a styrenic polymer and an olefinic polymer, the olefinic polymer in an amount less than the amount of the styrenic polymer. The styrenic polymer and the olefinic polymer are present as a blend, preferably free of an interpolymer or compatibilizer. The foam has a density below about 36 kg/m³ (as measured by ASTM D1622-03), a vertical compressive strength greater than about 100 kPa (as measured by ASTM D1621 -04) and a compressive strength ratio greater than about 0.35.



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**ARTICLE AND METHOD OF PRODUCING A LOW DENSITY FOAM BLEND
OF STYRENIC POLYMER AND POLYOLEFIN**

CLAIM OF PRIORITY

The present application claims the benefit of the filing date of US Provisional Application No. 61/026486 (filed 06 February 2008) the contents of which are hereby incorporated by reference in their entirety.

FIELD OF THE INVENTION

[001] The present invention relates generally to thermoplastic polymeric foams, and particularly to low density extruded foam blends and processing thereof.

BACKGROUND OF THE INVENTION

[002] In the field of polymer processing, extruded polystyrene foams are generally produced with relatively high density, usually above 36 kilograms per cubic meter (kg/m^3). To reduce the foam density, a secondary post expansion such as steam or vacuum post expansion may be used to increase blowing agent efficiency. However, it is observed that with the use of secondary post expansion, the foam tends to expand in a direction generally parallel with the extrusion direction (commonly known as the "machine direction" and along the X-axis in a Cartesian system) and does not tend to expand in a direction generally perpendicular to the extrusion direction (commonly known as the "cross-machine direction" and along the Y-axis in a Cartesian system). As a consequence of machine direction expansion, a drop in the vertical compressive strength (as measured per American Society for Testing Materials ("ASTM") ASTM D1621-04) may occur, wherein vertical compressive strength would be the compressive strength in response to a load along the Z-axis in a Cartesian system (commonly referred to as the thickness dimension, which is mutually perpendicular to the X and Y axes). The vertical compressive strength influences a measure known as the "compressive strength ratio (R_c)". The compressive strength ratio " R_c " is the ratio of vertical compressive strength to total compressive strength of the foamed article. Total

compressive strength is a sum of compressive strength in the X, Y and Z axes of the foam.

[003] In many applications for extruded foams, such as energy absorbing articles or safety barriers, it is often desired that the foam have a relatively low density (*e.g.*, less than about 36 kg/m³ as measured by ASTM D1622-03), relatively high Rc (*e.g.*, greater than about 0.35) or both. Foams with a vertical compressive strength of preferably greater than about 100 kilo-Pascal (kPa) and less than about 300 kPa per ASTM D1621-04) may also be desired. Good surface finish (*e.g.*, substantially free of cracking) and good cell structure (*e.g.*, foam cells containing pin holes less than about 5 percent (%) of the time in the case of a closed cell foam) may also be desirable. It is also attractive to produce such foam with relatively benign environmental consequences.

[004] Generally, prior to the present invention, the art sought to achieve the above mentioned foam characteristics through the use of blends of polystyrene and polyolefin resin and expensive compatibilizing ingredients. For example, one approach has been to employ polymeric compatibilizers (*e.g.*, hydrogenated styrene-conjugated diene rubber, ethylene-styrene interpolymer, styrene-isoprene-styrene triblock copolymer, or the like), permeability modifiers (*e.g.*, glycerol monostearate, monostearic acid glyceride, monostearin, or the like), or a cross linking agent. By way of illustration, U.S. Patent No. 6,048,909 discloses a foam with attractive characteristics in which an interpolymer is employed in a blend.

[005] From the processing side, the use of limonene as foaming aids, emulsion polymerisation or the use of an environmentally unfriendly blowing agents (*e.g.*, mixture of chlorine and dichlorodifluoromethane "CFC-12") have been used. By way of illustration, U.S. Patent No. 4,515,907 and Japanese Patent 61101538A disclose a foam manufactured with one or both of the above referenced environmentally unfriendly blowing agents.

[006] Among the literature that may pertain to this technology include the following patent documents: United States Published Patent Application ("USPub"); United States Patent ("US"); International Application Published Under The Patent Cooperation Treaty ("WO"); Japan Patent ("JP"); German Patent ("DE"); European Patent ("EP"); USPub2002111389A1; US5,290,822; US4,652,590; USPub2004152795A1; US4,515,907; USPub2005154115A1; US4,692,471; US4,605,682; US5,693,687;

US6,225,363; US5,137,933; US6,048,909; US5,591,778; WO2004087798A1; WO2000053669A1; WO9951667A1; WO8808864A1; WO9114724A2; JP06049256A; JP04089846A; JP2004352927A; JP62174237A; JP03081347A; JP02232240A; JP61101538A; JP59105036A; JP58191727; JP62280237A; JP2004323635A; DE102004042297A1; DE102004042297A1; EP1095969A2; and EP1847566A1, all incorporated herein by reference for all purposes.

SUMMARY OF THE INVENTION

[007] The present invention meets the above needs by providing an extruded styrenic foam and method of producing it that affords attractive density, compressive ratios, surface quality, cell structure characteristics or any combination thereof with a minimal amount of or without substantially any compatibilizing substance. Additives that function as compatibilizing substances or permeability modifiers generally are preferably present in an amount less than about 5% of the total polymer weight, more preferably less than about 2%, more preferably less than about 1%, and most preferably absent entirely from the foamed article.

[008] Accordingly, pursuant to one aspect of the present invention, there is contemplated a styrenic polymer having a glass transition temperature (T_g ; the temperature below which the physical properties of amorphous materials vary in a manner similar to those of a solid phase (glassy state), and above which amorphous materials behave like liquids); an olefinic polymer having a crystallization temperature (T_c), present in an amount by weight less than the amount of the styrenic polymer; wherein the styrenic polymer and the olefinic polymer are present as a blend including less than about 5% interpolymer or less than about 2% of any other compatibilizers by total polymer weight and wherein the foam has a resultant density below about 36 kg/m³ (as measured by ASTM D1622-03), a vertical compressive strength greater than about 100 kPa and optionally less than about 300 kPa (as measured by ASTM D1621-04), and a compressive strength ratio greater than about 0.35.

[009] The invention may be further characterized by one or any combination of the features described herein, such as the foam is essentially free of any compatibilizers; the styrenic polymer consists essentially of a styrene-acrylonitrile copolymer; the glass transition temperature T_g of the styrenic polymer and the crystallization temperature T_c

of the polyolefin, measured in °C, are within about 30°C of each other, and more preferably within 20°C of each other; the olefinic polymer consists essentially of polyethylene, polypropylene, ethylenic copolymer, or any combination thereof; the styrene-acrylonitrile copolymer has an acrylonitrile content of about 5% to 25% by weight of resulting material; the extruded low density polymeric foam includes at least one additive selected from a fire retardant, a colorant, a nucleator, a clay, an ultraviolet stabilizer, or an IR blocker; the extruded polymeric foam wherein the T_g is in the range of about 90 to 120°C, the T_c is in the range of about 70 to 130°C; the extruded polymeric foam consists essentially of at least 50% styrene-acrylonitrile copolymer and at least 5% linear low density polyethylene by weight; the extruded low density polymeric foam has a compressive strength ratio of greater than about 0.35 (as measured per ASTM D1621-04); the extruded low density polymeric foam has a resultant density is below about 32 kg/m³ (as measured by ASTM D1622-03); the extruded low density polymeric foam is an open cell foam, a closed cell foam, or any combination thereof (as used herein, "closed cell" foam structures refer to foams having an open cell content of less than 30%, as determined by ASTM D6226-05, while "open cell" foam structures refer to an open cell content greater than or equal to 30%, as determined by ASTM D6226-05); the extruded polymeric foam wherein the foam vertical compressive strength less than about 300 kPa; or any combination of the foregoing.

[0010] Another aspect of the present invention contemplates a process of forming an extruded polymeric foam article, comprising the steps of: blending a styrenic polymer with an olefinic polymer having a crystallization temperature greater than 25°C in an amount less than the amount of the styrenic polymer to form a polymeric blend including less than about 5% interpolymers or less than about 2% of any other compatibilizer by total polymer weight; introducing a blowing agent including H₂O, CO₂, hydrocarbons, hydrofluorocarbons, or any combination of the preceding to the polymeric blend; and foaming the polymeric blend at a temperature above the crystallization temperature of the olefinic polymer to a resulting density below about 28 kg/m³ (as measured by ASTM D-1622-03) and a compressive strength ratio greater than about 0.35.

[0011] This aspect of the invention may be further characterized by one or any combination of the features described herein, such as the styrenic polymer consists essentially of polystyrene, styrene-acrylonitrile copolymer, or any combination thereof;

the polyolefin consists essentially of polyethylene, polypropylene, ethylenic copolymer, olefin block copolymer or any combination thereof; the styrene-acrylonitrile copolymer has an acrylonitrile component of about 1% to 35%; the process includes a step of introducing at least one additive to the polymeric blend selected from a fire retardant, a colorant, a nucleator, a clay, an ultraviolet stabilizer, an Infrared (IR) blocker, or any combination thereof; the polymeric blend consists essentially of at least 50% by weight of the polymeric blend styrene-acrylonitrile copolymer and at least 2% linear low density polyethylene by weight of the polymeric blend; the foam is an open cell foam or the foam is a closed cell foam, or any combination of the foregoing.

[0012] Among the advantages obtainable from the aspects of the present invention are that relatively low density foam (e.g., less than ($<$) 36 kg/m³) with high compressive strength ratio (R_c greater than ($>$) 0.35, preferably $>$ 0.40) can be efficiently and reproducibly extruded online using an environmentally acceptable foaming agent and little or no compatibilizers. The foam generally will have a regular cell structure (that is, the cell structure is relatively equalized and is generally consistent and uniform throughout the foam). The resulting properties and characteristics should allow the foam to function well for applications requiring energy absorption. In particular, the foam may help meet the market requirements for Thorax impacts (e.g., impacts that effect vehicle passengers in a side impact) and Safety barrier impacts (e.g., vehicle impacts to roadway barriers) applications. For these energy absorbing applications, the vertical compressive strength of the foam should be less than 300 kPa, preferably less than 250 kPa and most preferably less than 200 kPa.

DETAILED DESCRIPTION

[0013] The present invention is directed at an improved polymeric foam article, and preferably one made from a thermoplastic foam (e.g., a polymeric foam material including an alkenyl aromatic polymer, an olefinic polymer, such as α -olefinic polymer, and more preferably a combination thereof). The present invention is particularly suited for polymeric foams made by an extrusion foaming process. By way of example, in a preferred aspect of the present invention, polymeric ingredients are converted into a polymer melt and a blowing agent is incorporated into the polymer melt to form a foamable gel. The foamable gel is then extruded through a die and appropriately cooled

to form a desired product. Depending upon the die and operating conditions, the product may vary from a coalesced foam strand product, such as an extruded foam plank or rod, through foam beads and eventually to chopped strands of foamable beads. Preferably, the product is a relatively thick plank or rod (e.g., greater than about 15 millimeters (mm)). More detailed processing parameters and steps are discussed below.

[0014] Any numerical values recited herein include all values from the lower value to the upper value in increments of one unit provided that there is a separation of at least 2 units between any lower value and any higher value. As an example, if it is stated that the amount of a component or a value of a process variable such as, for example, temperature, pressure, time and the like is, for example, from 1 to 90, preferably from 20 to 80, more preferably from 30 to 70, it is intended that values such as 15 to 85, 22 to 68, 43 to 51, 30 to 32 etc. are expressly enumerated in this specification. For values which are less than one, one unit is considered to be 0.0001, 0.001, 0.01 or 0.1 as appropriate. These are only examples of what is specifically intended and all possible combinations of numerical values between the lowest value and the highest value enumerated are to be considered to be expressly stated in this application in a similar manner.

[0015] Unless otherwise stated, all ranges include both endpoints and all numbers between the endpoints. The use of "about" or "approximately" in connection with a range applies to both ends of the range. Thus, "about 20 to 30" is intended to cover "about 20 to about 30", inclusive of at least the specified endpoints. "About" may also encompass the end point (e.g. "about 35") and plus or minus as much as 10% to 15% of the endpoint value (e.g. 35 could be 35+/-10%).

[0016] The disclosures of all articles and references, including patent applications and publications, are incorporated by reference for all purposes.

[0017] References to an acrylic or (meth)acrylic (or derivative terms such as "acrylate") contemplate meth-acrylics and acrylics (and corresponding derivative terms).

[0018] The term "consisting essentially of" to describe a combination shall include the elements, ingredients, components or steps identified, and such other elements ingredients, components or steps that do not materially affect the basic and novel characteristics of the combination.

[0019] The use of the terms "comprising" or "including" to describe combinations of elements, ingredients, components or steps herein also contemplates embodiments that consist essentially of the elements, ingredients, components or steps.

[0020] Plural elements, ingredients, components or steps can be provided by a single integrated element, ingredient, component or step. Alternatively, a single integrated element, ingredient, component or step might be divided into separate plural elements, ingredients, components or steps. The disclosure of "a" or "one" to describe an element, ingredient, component or step is not intended to foreclose additional elements, ingredients, components or steps. All references herein to elements or metals belonging to a certain Group refer to the Periodic Table of the Elements published and copyrighted by CRC Press, Inc., 1989. Any reference to the Group or Groups shall be to the Group or Groups as reflected in this Periodic Table of the Elements using the IUPAC system for numbering groups.

[0021] One particular preferred application of the aspects of the present invention is a closed cell low density extruded polymeric foam (e.g., including a polystyrenic material) with a density below about 36 kg/m^3 (as measured by ASTM D1622-03). It also exhibits a vertical compressive strength greater than about 100 kPa and less than about 300 kPa (as measured by ASTM D1622-03), good surface finish (e.g., visually substantially free of cracking) and good closed cell structure (e.g., closed foam cells as defined and measured by ASTM D6226-05 in an amount less than about 5% of total cells throughout substantially all of its volume). In another aspect, the foam article could exhibit the above characteristics, but will include an open cell foam. Closed cell foams of the present invention may have up to 30% open cell content, but preferably have 10% or less, more preferably 5% or less and most preferably 0% open cell content as determined by ASTM D6226-05. A particularly useful application of the teachings of the present invention herein is in the manufacture of an extruded foam, which may include or consist essentially of a polystyrene-including material such as a blend or copolymer thereof (e.g., a styrene-acrylonitrile copolymer) and an olefinic polymer. In a particularly preferred embodiment, the T_g of the styrenic polymer and the T_c of the olefinic polymer, are preferably within about 30°C and more preferably within about 20°C of each other.

[0022] The T_c is measured using a differential scanning calorimeter (DSC) according to ASTM D3418-03. It is obtained from the cooling curve and is the peak melting

temperature. The glass transition temperature is obtained using the half-height method from the second DSC melting curve (also called second heat) according to ASTM E1356-03. The procedure consists of heating rapidly a 5 gram sample in a sealed aluminum pan from ambient temperature to 180°C (at a rate of 10°C per minute); kept at 180°C for 4 minutes to ensure complete melting; cooled at 10°C/min to about 40°C below the expected T_g ; kept at this temperature for 4 minutes for DSC stabilization; and heated again to 180°C at 10°C/min.

[0023] One illustrative article of the present invention includes at least about 50% polystyrene, at least about 1% styrene-acrylonitrile copolymer, and at least 5% olefinic polymer, all by weight of the total polymer. Additives that function as compatibilizers or permeability modifiers (e.g., hydrogenated styrene-conjugated diene rubber, hydrogenated and acid-modified vinylarene-diene block copolymers, ethylene-styrene interpolymers, styrene-isoprene-styrene triblock copolymer, glycerol monostearate, and monostearic acid glyceride, monostearin) generally are preferably present in an amount less than about 5% of the total polymer weight, more preferably less than about 2%, more preferably less than about 1%, even more preferably less than about 1% and most preferably absent entirely from the foamed article. By way of example, it is contemplated that such compatibilizers or permeability modifiers as listed above may be present, but preferably only in limited quantities (as a weight percentage of the total polymer) such as: less than 5% hydrogenated styrene-conjugated diene rubber; less than 5% hydrogenated and acid-modified vinylarene-diene block copolymers; less than 5% ethylene-styrene interpolymers; less than 3% styrene-isoprene-styrene triblock copolymer; less than 2% glycerol monostearate; less than 1% monostearic acid glyceride, less than 0.5% monostearin. For terms of the present application, a compatibilizer may be defined as a substance that prevents macroscopic phase separation of the polymer blend, and the polymer blend is melt processable to form a foam. For terms of the present application, a permeability modifier is a substance that can be used to modify the rate at which the blowing agent escapes from the cells of the foam after the foam is cooled. Other fillers or additives (e.g., a fire retardant, a colorant, a nucleator, a clay, an ultraviolet stabilizer, an infrared (IR) blocker, or the like) may be added to enhance article properties (e.g., article color, thermal transfer properties, flammability or the like).

[0024] As indicated, the present invention makes advantageous use of alkenyl aromatic compounds such as styrene. Accordingly, it should be appreciated that reference to a "styrenic" or "polystyrene" material, in the context of the extruded polystyrene foam article herein, includes polymeric materials containing greater than about 50, preferably about 75 or more, more preferably about 85 or more weight percent of a polymer derived from one or more alkenyl aromatic compounds such as styrene. The polymeric material may be entirely one or more alkenyl aromatic compound. Suitable amounts of copolymerizable compounds, such as C₁₋₄ methacrylates and acrylates, acrylic acid, methacrylic acid, maleic acid, acrylonitrile, maleic anhydride, and vinyl acetate may be incorporated into the polystyrene material. One preferred embodiment employs a copolymer of styrene with a vinyl, and more preferably with an ethylenically unsaturated nitrile, such as acrylonitrile, methacrylonitrile or combination thereof. For example, it is preferred to employ such ethylenically unsaturated nitrile as part of a co-polymer, and particularly a melt processable copolymer such as styrene-acrylonitrile ("SAN").

[0025] Examples of alternative styrenic polymers suitable for use herein may be found in paragraphs 28-38 of published U.S. Application No. USPub20020111389, incorporated by reference in its entirety.

[0026] It should be appreciated that reference to a styrene-acrylonitrile (SAN) copolymer (or, for simplicity, "SAN"), in the context of the extruded polystyrene foam article herein, can include a SAN copolymer that contains a graft copolymer content, a block copolymer content, a random copolymer content, or any combination thereof. The preferred SAN employed herein preferably include graft copolymer content. The SAN copolymer can also be linear or branched.

[0027] Typically, the weight-averaged molecular weight (Mw) of the SAN and, desirably, any additional polymer in the polymer foam is about 40,000 or more, preferably about 60,000 or more, more preferably, about 75,000 or more. The Mw of the SAN and, desirably, any additional polymer in the polymer foam is generally about 300,000 or less, preferably about 250,000 or less, and more preferably about 150,000 or less. By way of example the Mw ranges from about 100,000 to about 145,000, and more preferably about 120,000 to about 135,000.

[0028] Additionally, it is desirable for about 90% or more, and preferably all of the polymeric ingredients in the foam to have a Mw of less than about 1,000,000. The

weight average molecular weight of the SAN or any other polymeric ingredient is preferably selected to balance competing considerations. For example, the weight average molecular weight is desirably high enough that resulting foams will exhibit sufficient physical characteristics for the intended application. However, it is not so high that it will unduly increase gel viscosity during processing, so that reliable and consistent foaming will be compromised.

[0029] The proportion of weight average molecular weight (M_w) to number average molecular weight (M_n) for the SAN copolymer, expressed as M_w/M_n is preferably in the range of about 1.5 to about 5.0, and more preferably it is about 2.3.

[0030] The styrenic polymers preferably have a glass transition temperature of at least about 80°C, and more preferably at least about 100°C. The styrenic polymers preferably have a glass transition temperature of less than about 130°C, and more preferably less than about 120°C.

[0031] For the embodiments in which SAN is employed as the styrenic material or in combination with another styrenic material, preferably, the concentration of polymerized acrylonitrile (AN) in the SAN copolymer is sufficient that it allows foaming with commercially attractive blowing agents while maintaining attractive thermal stability characteristics while in the melt phase in an extruder. For example, it is preferable that the AN be present in an amount of at least about one (1) % by weight of the total styrenic material composition, more preferably at least about five (5) wt% or more, more desirably about ten (10) wt% or more and desirably about 50 wt% or less, typically about 35 wt% or less and more typically about 20 wt% or less based on total styrenic material composition weight. The total styrenic material represents at least 50 wt% of the total polymeric material of the foamed article and preferably greater than 50 wt% and can be 75 wt% or more, 90 wt% or more, 95 wt% or more and even 98 wt% based on total polymer weight in the foam.

[0032] As indicated, among the polymeric constituents of the foams of the present inventions, there is preferably included at least one polyolefinic material, and preferably an α -olefin. The amount of polyolefinic material to styrenic material is from about 2:98 to about 20:80 or greater, preferably about 5:95 and more preferably about 10:90, although always in an amount less than that of the styrenic material. The α -olefin polymer is a polymeric material that contains repeated units derived by polymerizing an α -olefin. The

α -olefin polymer may optionally be free of any polymerized vinyl aromatic monomers, sterically hindered aliphatic or cycloaliphatic vinyl or vinylidene monomers, or both. Particularly suitable α -olefins have from 2 to about 20 carbon atoms, preferably from 2 to about 8 carbon atoms, and include ethylene, propylene, 1-butene, 4-methyl-1-pentene, 1-hexene, 1-octene and the like. A particularly preferred polyolefinic material includes and more preferably consists essentially of ethane-1-octene copolymer. Preferred α -olefin polymers are homopolymers or copolymers of ethylene or propylene. It is preferred that the olefinic polymer includes polyethylene, polypropylene, ethylenic copolymer, or any combination thereof. The preferred olefinic polymer includes a polyethylene, such as low density polyethylene (LDPE), linear low density polyethylene (LLDPE), high density polyethylene (HDPE), Ultra high molecular weight polyethylene (UHMWPE), or any combination thereof. A particularly preferred olefinic polymer includes LLDPE, which may include an ethylene homopolymer, and/or an ethylene copolymerized with a longer chain olefin, such as butene, hexene, octene, or any combination thereof. The polyolefinic material may be the reaction product of a low pressure process employing conventional Ziegler-Natta type catalysts, as described in U.S. Pat. No. 4,076,698. It may have a generally homogeneous distribution of comonomer, as are described, for example, in U.S. Pat. No. 3,645,992 and U. S. Pat. Nos. 5,026,798 and 5,055,438, all incorporated by reference. The polyolefinic material may be a reaction product of polymerization using a metallocene catalyst.

[0033] Particularly suitable α -olefinic polymers have a melt index (ASTM D1238-05, 190°C /2.16 kg) of from about 0.01 to about 1000 grams per 10 minutes (g/10 min), and a density of from about 0.85 to about 0.97 grams per cubic centimeter (g/cc), and more preferably from about 0.85 to about 0.95 g/cc. The olefinic polymers preferably have a crystallization temperature of at least about 50°C, and more preferably at least about 70°C. The olefinic polymers preferably have a crystallization temperature of less than about 130°C, and more preferably less than about 120°C. The olefinic polymers preferably have a Tensile Yield ranging from about 8 to 13 mega-Pascal (MPa) and a Ultimate Elongation Percentage ranging from about 200% to 900%, all as measured by ASTM 882.

[0034] An example of one preferred olefinic polymer has a density of about 0.92 kg/m³ (ASTM D792-00), a melt mass flow rate of about 2.3 (190°C/2.16 kg, ASTM D1238-05) and a T_c of about 105°C.

[0035] An example of one preferred olefinic polymer is LLDPE Dowlex™ 2247, available from The Dow Chemical Company.

[0036] Other suitable olefinic polymers may be selected from those disclosed in paragraphs 38-45 of published U.S. Application No. USPub20020111389, incorporated by reference. Thus suitable α -olefin polymers may include homopolymers or copolymers of propylene. In addition, olefinic polymers that have been subjected to coupling or light crosslinking treatments are useful herein, provided that they remain melt processable.

[0037] The foams herein are made with the use of a suitable blowing agent, *e.g.*, a physical blowing agent, a chemical blowing agent, or both. Physical blowing agents include gasses and liquids that volatilize under the conditions of the foaming process, whereas chemical blowing agents produce a gas under the condition of the foaming process through some chemical means, usually decomposition. Particularly suitable physical blowing agents include halocarbons containing 1 or 5 carbon atoms such as methyl chloride, ethyl chloride, hydrofluorocarbons such as difluoromethane (HFC-32), perfluoromethane, ethyl fluoride (HFC-161), 1,1-difluoroethane (HFC-152a), 1,1,1-trifluoroethane (HFC-143a), 1,1,2,2-tetrafluoroethane (HFC-134), 1,1,1,2-tetrafluoroethane (HFC-134a), pentafluoroethane (HFC-125), perfluoroethane, 2,2-difluoropropane (HFC-272fb), 1,1,1-trifluoropropane (HFC-263fb), 1,1,1,2,3,3,3 – heptafluoropropane (HFC-227ea), 1,1,1,3,3-pentafluoropropane (HFC-245fa), and 1,1,1,3,3-pentafluorobutane (HFC-365mfc); organic blowing agents such as saturated or unsaturated and cyclic hydrocarbons having from two to nine carbons (C2-C9) including ethane, propane, n-butane, isobutane, n-pentane, isopentane, neopentane, cyclobutane, and cyclopentane; and aliphatic alcohols having from one to five carbons (C1-C5) such as methanol, ethanol, n-propanol, and isopropanol; carbonyl containing compounds such as acetone, 2-butanone, and acetaldehyde; ether containing compounds such as dimethyl ether, diethyl ether, methyl ethyl ether; carboxylate compounds such as methyl formate, methyl acetate, ethyl acetate; carboxylic acid compounds. Carbon dioxide, nitrogen, argon, water and the like are also useful. Mixtures of these physical expanding

agents can be used. Suitable chemical blowing agents include, for example, azodicarbonamide, dinitrosopentamethylene tetramine, benzenesulfonyl hydrazide, azodiisobutyronitrile, 4,4-oxybenzene sulfonyl semi-carbazide, p-toluene sulfonyl semi-carbazide, barium azodicarboxylate, N,N'-dimethyl-N,N'-dinitrosoterephthalamide, trihydrazino triazine and sodium bicarbonate. A particularly preferred blowing agent for use herein is selected from water, carbon dioxide, isobutane (iC4) or any combination thereof

[0038] By way of example, in one preferred embodiment, the blowing agent may be incorporated into the melt in a weight proportion of between about 0.5 to about 50 parts of blowing agent per hundred parts of the total polymer weight (e.g., LLDPE and SAN copolymer) to be expanded, more preferably about 1 to about 30 parts per hundred (pph) parts of the polymer to be expanded; and still more preferably from about 3 to 15 parts per hundred parts of the polymer to be expanded.

[0039] In one preferred embodiment, the foam contains less than about 5%, preferably less than about 2%, or most preferably is free of any random interpolmer. Such a compatibilizer is discussed in published U.S. Application No. USPub20020111389, incorporated herein by reference. Additionally, is preferably free of any α -olefin/vinyl aromatic interpolmer. This includes copolymers, terpolymers and tetrapolymers. "Monomeric unit" refers to a polymer backbone portion that is derived from a single monomer. "Hydrocarbyl" means any aliphatic, cycloaliphatic, aromatic, aryl substituted aliphatic, aryl-substituted cycloaliphatic, aliphatic substituted aromatic, or aliphatic substituted cycloaliphatic groups. "Hydrocarbyloxy" means a hydrocarbyl group having an oxygen linkage between it and the carbon atom to which it is attached. "Aliphatic" means a compound having a straight- or branched-chain arrangement of its carbon atoms.

[0040] An interpolmer is defined as a polymer blend that contains a polymeric compatibilizer for the alkenyl aromatic polymer and the α -olefin polymer. The polymeric compatibilizer prevents macroscopic phase separation of the polymer blend, and the polymer blend is melt processable to form a foam. The compatibilizer enhances the mixing between the polymeric components. Suitable compatibilizers include certain aliphatic α -olefin/vinyl aromatic interpolymers.

[0041] Examples of α -olefins include for example, α -olefins containing from 3 to about 20, preferably from 3 to about 12, more preferably from 3 to about 8 carbon atoms. Particularly suitable are ethylene, propylene, butene-1, 4-methyl-1-pentene, hexene-1 or octene-1 or ethylene in combination with one or more of propylene, butene-1, 4-methyl-1-pentene, hexene-1 or octene-1. These α -olefins do not contain an aromatic, hindered aliphatic or cycloaliphatic moieties. Other optional polymerizable ethylenically unsaturated monomer(s) include norbornene and C 1-10 alkyl or C 6-10 aryl substituted norbornenes, with an exemplary interpolymer being ethylene/styrene/norbornene.

[0042] Additionally, in a preferred embodiment, the foam may be a binary blend, free of cross linking styrene and olefin. It may also be free of an interpenetrating network derived by solution or emulsion polymerization of styrene and olefin, and free of grafted ethylene propylene diene monomer (EPDM).

[0043] In another preferred embodiment, it is desirable for the foam article to have a substantial thickness such that it is useful in applications such as Thorax impacts (e.g., impacts that effect vehicle passengers in a side impact) and Safety barrier impacts (e.g., vehicle impacts to roadway barriers) applications. The foam article is preferably at least about 10 mm thick, more preferably about 15 mm thick and most preferably about 20 mm thick or more.

[0044] Though the compositions herein may have application for making expanded bead foams, preferably, the present foam structures are formed by an extrusion process, such as that described in paragraphs 74-81 of published U.S. Application No. USPub20020111389, incorporated by reference.

[0045] The process thus will typically include melting and mixing the polymer or polymers as previously described or with other polymers, additive ingredients (such as one or more of slip agents, dyes, pigments, fillers (optionally, the material may be free of any filler, e.g., it is free of any talc), antioxidants, extrusion aids, nucleating agents, stabilizing agents, antistatic agents, fire retardants, acid scavengers, and infrared attenuators including carbon black and graphite), or any combination thereof, to form a plastic melt.

[0046] A blowing agent is incorporated into the plastic melt for forming a foamable gel, and the foamable gel is extruded through a die for forming a resulting foamed structure. During melting and mixing, the polymers are heated to a temperature at or above the

softening temperature of the styrenic polymer and at or above the T_c of the olefinic polymer. The polymer blend can be prepared by simple melt blending. If desired, the individual polymers can be separately charged into an extruder together with blowing agent and other additives to form the polymer blend as part of the foam-making process. Alternatively, the polymer blend can be made separately prior to the foaming process. The dispersion of the polymer components preferably is substantially uniform (e.g., components dispersed in proportional amounts throughout the polymer blend).

[0047] Melting and mixing of ingredients can be accomplished by any means known in the art such as with an extruder, mixer, or blender. Mixing can be accomplished as a separate step, or it can be integrated into the extrusion step by selecting an extruder embodiment suitable for mixing (e.g., a single screw extruder using a mixing screw or a twin screw extruder). The blowing agent is typically blended with the plastic melt at an elevated pressure sufficient to prevent substantial expansion of the resulting gel or loss of generally homogeneous dispersion of the blowing agent within the gel (e.g., typically pressures ranging from about 100 to about 300 bar, where 1 bar is essentially equivalent to 1 atmospheric pressure at sea level).

[0048] The resulting foamable gel is preferably passed through a cooling apparatus (e.g., a single screw extruder, a heat exchanger, or some combination thereof) to lower the gel temperature to an optimum foaming temperature while still maintaining the temperature at or above the T_g of the styrenic polymer and at or above the T_c of the olefinic polymer. Typical foaming temperatures will range from about 110°C to about 150°C. The preferred foaming temperature for the present invention is selected to take into account the T_g of the styrenic polymer and the T_c of the olefinic polymer. The cooled foamable gel then arrives at the die. The pressure at the die entrance is of about 30 to about 150 bar, and more preferably about 50 to 120 bar. The gel is then extruded through a die into atmospheric pressure to form a foam structure of the present invention. A preferable foaming temperature is within a range from about 110°C to about 150°C, and more preferably from about 120°C to about 145°C based upon the particular styrenic polymer blend and olefinic polymer contemplated for use therein.

[0049] The foams of the present invention may be used in any application where hard, ridged foams are used. Such applications include, without limit, packaging (e.g., corner blocks, braces, saddles, pouches, bags, envelopes, overwraps, interleaving, and

encapsulation); material handling (e.g., trays, tote boxes, box liners, tote box inserts and dividers, shunts, stuffing, boards, parts spacers and parts separators); automotive (e.g., headliners, impact absorption in bumpers or doors, carpet underlayment, sound insulation, and helmet liners); roadway barriers (exit ramp barriers); flotation (e.g., life jackets, vests, and belts); sports and leisure (e.g., gym mats and bodyboards); thermal insulation such as that used in building and construction. The foregoing list merely illustrates a number of suitable applications. Skilled artisans can readily envision additional applications without departing from the scope or spirit of the present invention.

[0050] The following examples are provided to illustrate the invention but are not intended to limit the scope thereof. All parts and percentages (except percent open cell) are by weight unless otherwise indicated. The dimension of the foams produced in table 2 were not large enough for measuring representative compressive strengths.

Example 1

[0051] A blend of 95% SAN copolymer ($M_w = 131,000$ with $M_w/M_n = 2.3$, $T_g = 105^\circ\text{C}$) and 5% of LLDPE (Dowlex™ 2247, $T_c = 105^\circ\text{C}$) is fed into an extruder where the temperature of the melting zone is set at 200°C . The melt is extruded into a mixer, where a mixture of blowing agents consisting of CO_2 , H_2O and isobutane (iC_4) (measured as part per hundred parts of total polymer weight ("pph")) is injected into and mixed with the polymer melt. The total blowing agent concentration is 0.183 mol per hundred grams (mph) of total polymer. The mixture is then cooled down to about 130°C (e.g., the foaming temperature) and extruded through a slit die into atmospheric pressure. Foaming occurs as the mixture exits the die. The expected foam properties are presented in Table 1. Foam density is determined per ASTM D1622-03, the cell size per ASTM D3576, open cell content per ASTM D6226, and compressive strength per ASTM D1621-04.

[0052] A density of 25.3 kg/m^3 is achieved with a vertical compressive strength of 230 kPa and a compressive strength ratio (R_c) of about 0.55. The open cell content is less than about 0.8% and the foam is achieved with an essentially crack free surface finish (e.g., visual inspection of surface).

Example 2

[0053] The Example 1 is repeated except the LLDPE loading is increased to 10%. A density of 25.5 kg/m³ is produced, with a vertical compressive strength of 187 kPa and a compressive strength ratio of 0.55. An open cell content of about less than 1.4% is achieved with an essentially crack free surface finish.

Example 3

[0054] The Example 2 is repeated except that the foaming temperature is increased to 133°C. A density of 23.7 kg/m³ is achieved, with a vertical compressive strength of 204 kPa and a compressive strength ratio of 0.58. The open cell content is less than about 15.0% and the foam has an essentially crack free surface finish.

Example 4

[0055] The Example 3 is repeated except that the blowing agent mixture consists of CO₂ and H₂O only. A density of 24.9 kg/m³ is produced, with a vertical compressive strength of 216 kPa and a compressive strength ratio of 0.57. An open cell content of about 31.4% is produced (classifying this as an open cell foam) with an essentially crack free surface finish.

Comparative Example 1

[0056] The comparative-1 foam is produced with SAN copolymer (Mw = 131,000 with Mw/Mn = 2.3) and with only 0.4% of LLDPE Dowlex-2247. The blowing agent mixture is like the one in Example 1 (CO₂, H₂O, and iC₄). The foaming temperature is at 130°C. Foam density is higher than the previous examples and measured to be about 28.8 kg/m³. The foam has a vertical compressive strength of about 307 kPa and is not suitable for use in energy absorption applications, such as Thorax and Safety barriers.

Example 5

[0057] A blend of SAN copolymer (Mw = 131,000 with Mw/Mn = 2.3) with a T_g of 105°C and 10% of LDPE (Dow LDPE 620I) with a T_c around 100°C is fed into an extruder at a temperature around 210°C. The melt is extruded into a mixer, where a blowing agent mixture consisting of 4 pph of CO₂ and 1.2 pph of H₂O is injected into and mixed with the polymer melt. The mixture is then cooled down to about 145°C and extruded through a slit die into atmospheric pressure. Foaming occurs as the polymer-blowing agent mixture exits the die. The foam has a density of 35.8 kg/m³ with zero open cell and is achieved with an essentially crack free surface finish.

Example 6

[0058] Example 5 is repeated except that the polyolefin is a HDPE (HDPE DMDA-6230) with a T_c of about 117°C. The foam has a density of 33.3 kg/m³ with only 5% open cell and is achieved with an essentially crack free surface.

Example 7

[0059] Example 5 is repeated except that the polyolefin is a HPDE (HDPE DGDH-1059) with a T_c of about 120°C. The foam has a density of 32.4 kg/m³ with zero open cell and is achieved with an essentially crack free surface.

Example 8

[0060] Example 7 is repeated except that concentration of HDPE DGDH-1059 is increased to 20%. The foam has a density of 32.5 kg/m³ with only 5% open cell and is achieved with an essentially crack free surface finish.

Comparative Example 2

[0061] The comparative foam for examples 5 through 8 is produced with SAN copolymer ($M_w = 131,000$ with $M_w/M_n = 2.3$) and without any polyolefin resin. The blowing agent mixture and the foaming conditions are like the ones in Examples 5 through 8. Foam density for the comparative example is again higher, about 40.4 kg/m³.

Comparative Example 3

Comparative Example 3 is produced with SAN copolymer ($M_w = 131,000$ with $M_w/M_n = 2.3$) and with 20% of an elastomeric resin (ENR-6386) with a T_c of about 47°C. The blowing agent mixture and the foaming conditions are like the ones in Examples 5 through 8. Foam density for Comparative Example 3 is 39.4 kg/m³ indicating that this formulation containing a resin with a low T_c does not produce the low densities embodied in our invention. The open cell content is 50%

Table 1: Results for illustrative foams in examples 1 - 4 according to the invention

		Ex. 1	Ex. 2	Ex. 3	Ex. 4	Comparat ive Ex. 1
FORMULATIONS	unit	Very low density	Very low density	Very low density	Very low density	Low density
	pph (part per hundred)					
CO ₂		3.00	3.00	3.00	4.00	3.00
iC4	pph	1.50	1.50	1.50	0.00	1.50

H ₂ O	pph	1.60	1.60	1.60	1.60	1.60
BA moles	mph	0.183	0.183	0.183	0.180	0.183
LLDPE	pph	5.00	10.00	10.00	10.00	0.40
Talc	pph	0.00	0.00	0.00	0.00	0.40
Foaming Temperature	°C	133	130	133	133	130
Blowing Agent Pressure	bar	134	138	132	134	139
Die Pressure	bar	82	79	80	76	87
Foam Thickness	mm	22	28	28	27	23
Foam Density	kg/m ³	25.3	25.5	23.7	24.9	28.8
Cell Size	mm	0.22	0.46	0.41	0.35	0.31
Open Cell	%	0.8	1.4	15.0	31.4	1.1
Compressive Strength Vertical	kPa	230	187	204	216	307
Compressive Strength Extrusion	kPa	100	69	56	74	180
Compressive Strength Horizontal	kPa	102	87	92	86	125
Compressive strength ratio	Rc	0.53	0.55	0.58	0.57	0.50

Table 2: Results for illustrative foams in examples 5 - 10 according to the invention

		Ex. 5	Ex. 6	Ex. 7	Ex. 8	Comparative Ex. 2	Comparative Ex. 3
		Low Density	Low Density	Low Density	Low Density	Medium Density	Medium Density
Polyolefin (PO) Type		LDPE 620I	HDPE 6230	HDPE 1059	HDPE 1059	No Polyolefin	Elastomer ENR-6386
MI of PO (2.16 kg, 190°C)	g/10 min	1.8	0.3	0.9	0.9	-	<0.1
PO Density	g/cm ³	0.92	0.95	0.96	0.96	-	0.88
T _c of PO	°C	100	117	120	120	-	47
PO Loading	%	10	10	10	20	0	20
BaSt Loading	pph	0	0	0	0	0.15	0
Talc Loading	pph	0.3	0.3	0.3	0.3	0	0.3
Blowing Agent Pressure	bar	183	228	207	190	169	207
Die Pressure	bar	83	95	90	83	41	86

Foam Density	kg/m ³	35.8	33.3	32.4	32.5	40.4	39.4
Open cell	%	0	5	0	5	0	50
Average 3D Cell Size	mm	0.38	0.34	0.47	0.26	0.34	0.27
Vertical Cell Size	mm	0.45	0.37	0.48	0.26	0.38	0.25
Horizontal Cell Size	mm	0.36	0.35	0.48	0.28	0.34	0.26
Extruded Cell Size	mm	0.34	0.31	0.46	0.24	0.32	0.30

[0062] The explanations and illustrations presented herein are intended to acquaint others skilled in the art with the invention, its principles, and its practical application. Those skilled in the art may adapt and apply the invention in its numerous forms, as may be best suited to the requirements of a particular use. Accordingly, the embodiments of the present invention as set forth are not intended as being exhaustive or limiting of the invention. The scope of the invention should, therefore, be determined not with reference to the above description, but should instead be determined with reference to the appended claims, along with the full scope of equivalents to which such claims are entitled.

CLAIMS**WHAT IS CLAIMED IS:**

1. An extruded polymeric foam comprising:
a styrenic polymer having a glass transition temperature (T_g); an olefinic polymer having a crystallization temperature (T_c), and being present in an amount by weight less than the amount of the styrenic polymer; wherein the styrenic polymer and the olefinic polymer are present as a blend including less than 5% interpolymer or less than 2% of any other compatibilizers by total polymer weight and wherein the foam has a density below about 36 kg/m³ (as measured by ASTM D1622-03), a vertical compressive strength greater than about 100 kPa as measured by ASTM D1621-04, and a compressive strength ratio (R_c) greater than about 0.35.
2. The extruded polymeric foam article according to claim 1, wherein the foam article is entirely free of any compatibilizers.
3. The extruded polymeric foam article according to claims 1 or 2, wherein the styrenic polymer consists essentially of a styrene-acrylonitrile copolymer.
4. The extruded polymeric foam article according to any of claims 1 through 3, wherein the T_g (in °C) of the styrenic polymer and the T_c (in °C) of the polyolefin, are within about 30°C of each other.
5. The extruded polymeric foam according to any of claims 1 through 4, wherein the olefinic polymer consists essentially of polyethylene, polypropylene, ethylenic copolymer, or any combination thereof.
6. The extruded polymeric foam according to any of claims 3 through 5, wherein the styrene-acrylonitrile copolymer has an acrylonitrile component of about 5% to 25% based on total weight of styrene-acrylonitrile copolymer.
7. The extruded polymeric foam according to any of claims 1 through 6, wherein the T_g is in the range of about 90 to 120°C, the T_c is in the range of about 70 to 130°C.

8. The extruded polymeric foam according to claims 6 or 7, consists essentially of at least 50% styrene-acrylonitrile copolymer and at least 5% linear low density polyethylene based on combined weight of styrene-acrylonitrile copolymer and linear low density polyethylene.
9. The extruded polymeric foam according to claim 8 wherein it has a compressive strength ratio of greater than about 0.40 (as measured per ASTM D1621-04).
10. The extruded polymeric foam according to claim 9 wherein the resultant density is below less than about 28 kg/m³ as measured by ASTM D1622-03.
11. The extruded polymeric foam according to claim 10, wherein the foam is a closed cell foam.
12. The extruded polymeric foam according to claim 10, wherein the foam has a vertical compressive strength that is less than about 300 kPa.
13. A process of forming an extruded polymeric foam article, comprising the steps of:
 - (a) blending a styrenic polymer with an olefinic polymer having a T_c greater than 70°C in an amount less than the amount of the styrenic polymer to form a polymeric blend that including less than 5% interpolymer or less than 2% of any other compatibilizers by total polymer weight;
 - (b) introducing a blowing agent including water (H₂O), carbon dioxide (CO₂), or both to the polymeric blend; and
 - (c) foaming the polymeric blend at a temperature above the T_c of the olefinic polymer to directly form a foam having a density below about 36 kg/m³(as measured by ASTM D1622-03) and a compressive strength ratio (R_c) greater than about 0.35.

14. The process according to claim 13, wherein the styrenic polymer consists essentially of polystyrene, styrene-acrylonitrile copolymer, or a combination thereof.
15. The process according to any of claims 13 through 14, wherein the polyolefin consists essentially of polyethylene, polypropylene, ethylenic copolymer, or any combination thereof.
16. The process according to claim 14, wherein the styrene-acrylonitrile copolymer has an acrylonitrile component of about 1% to 35% by total weight of styrene-acrylonitrile copolymer.
17. The process according to any of claims 13 through 16, wherein the polymeric blend consists essentially of at least 50% by weight of the polymeric blend styrene-acrylonitrile copolymer and at least 5% linear low density polyethylene by weight of the polymeric blend.
18. The process according to any of claims 13 through 17, wherein the foam is a closed cell foam.
19. The process according to any of claims 13 through 18, wherein the polymeric blend is free of any compatibilizer.

INTERNATIONAL SEARCH REPORT

International application No

PCT/US2008/086797

A. CLASSIFICATION OF SUBJECT MATTER

INV. C08J9/00 C08J9/12 C08L25/04 C08L25/12 C08L23/00

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)

C08J

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 practical, search terms used)

EPO-Internal

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 4 515 907 A (MCCULLOUGH THOMAS W [US] ET AL) 7 May 1985 (1985-05-07) cited in the application column 3, line 32 - line 50 column 4, line 33 - line 40; examples	1-19
X	EP 1 847 566 A (TOPOX FOAM S L [ES]) 24 October 2007 (2007-10-24) cited in the application examples 4,6,7,9-11	1-19
X	WO 00/78851 A (DOW CHEMICAL CO [US]; CHAUDHARY BHARAT I [US]) 28 December 2000 (2000-12-28) example 2	1-19
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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.

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Date of the actual completion of the international search

25 February 2009

Date of mailing of the international search report

04/03/2009

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INTERNATIONAL SEARCH REPORT

International application No

PCT/US2008/086797

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WONG C-M ET AL: "EFFECT OF LOW DENSITY POLYETHYLENE ON POLYSTYRENE FOAM" JOURNAL OF CELLULAR PLASTICS, SAGE PUBLICATIONS, LONDON, GB, vol. 42, no. 2, 1 March 2006 (2006-03-01), pages 153-163, XP001244963 ISSN: 0021-955X page 154 - page 155; table 1 -----	1-19
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X	WO 96/11970 A (DOW CHEMICAL CO [US]; SUH KYUNG [US]; DEMIROERS MEHMET [NL]; VO CHAU V) 25 April 1996 (1996-04-25) example 8 -----	1-19
X	US 5 426 125 A (VO CHAU V [US] ET AL) 20 June 1995 (1995-06-20) examples #19,#20,#21,#22,#23 -----	1-19

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