

"INTERPOLYMER RESIN PARTICLES"

5 BACKGROUND OF THE INVENTION

1. Field of the invention

 The present invention relates to polyolefin
resin particles that contain a vinyl aromatic
10 monomer that is polymerized in the polyolefin matrix
to form an interpenetrating network of polyolefin-
polymerized vinyl aromatic monomer, e.g.
polystyrene. More particularly, the present
invention relates to interpolymer resin particles
15 that have little or no gel content; to a process for
producing the interpolymer resin particles; to a
polymer composition comprising the interpolymer
resin particles and a second polymer; and to
articles made from the interpolymer resin particles
20 and/or from the aforesaid polymer composition.
These articles may be formed via processing
techniques, e.g. sheet extrusion, rotomolding,
thermoforming, compression molding, injection
molding, blown film extrusion, and direct-injection
25 foamed sheet extrusion.

2. Background Art

 It is known to polymerize vinyl aromatic
monomers, such as styrene, in polyethylene. For
30 example, U.S. Patent No. 3,959,189 issued to
Kitamori and assigned to Sekisui Kaseihin Kabushiki
Kaisha, discloses a process for producing
polyethylene resin particles. The polyethylene
resin particles have a melt index (MI) value of 0.3
35 to 10 (Condition I, 190°C, 2.16 kg), a density of
less than 0.93 g/cm³ and a VICAT softening point
below 85°C. After the polyethylene resin particles

5 are added to an aqueous suspension, 30% to 100% by
weight based on the weight of the particles of a
styrene monomer and a catalyst for polymerizing the
monomer are added to the suspension, and the styrene
monomer is polymerized therein. Embodiments include
10 adding a cross-linking agent to the polyethylene
prior to the polymerization and cross-linking the
polyethylene before impregnating a blowing agent
into the polyethylene resin particles to form
foamable polyethylene resin particles. In view of
15 the cross-linking agent, the polyethylene-
polystyrene resin particles generally have high gel
content, i.e. from about 10% to 45% by weight. The
gel content of the final foamed shaped article can
be as high as 60% to 80% by weight. Even though
20 these cross-linked polyethylene-polystyrene resin
foams may have superior thermal stability and
toughness, these same characteristics make these
resin particles unsuitable for use in processes such
as compounding, extrusion processing, and injection
25 molding since the cross-linking effect tends to
reduce the melt flow of these particles which
affects the processability of these particles in
that the amperage needed to operate the processing
machinery, e.g. extruder is increased. The melt
30 fracture is therefore increased resulting in surface
irregularities, such as holes and ridges.

A further example of polystyrene resin
polymerized in polyethylene resin particles is
described in Japanese Patent No. 32623/70. Cross-
35 linking of the polyethylene resin, polymerization of
styrene, and impregnation of the blowing agent are

5 carried out at the same time. Since the polyethylene resin particles are cross-linked, the polyethylene resin particles generally contain a high gel content, i.e. at least about 24 % by weight, making these polyethylene resin particles generally
10 unsuitable for polymer processing, such as extrusion, injection molding, blown film, and direct injection foamed sheet extrusion. If the gel content is too high, hard spots form on the surface resulting in poor surface quality. The processing of
15 the polymer proves to be difficult due to the high amperage needed for the machinery used to process the polymer, for example, extruders or injection molding machines.

U.S. Patent No. 4,782,098 assigned to General
20 Electric Co. discloses expandable interpolymer beads comprising polyphenylene ether resin and a polymerized vinyl aromatic monomer such as styrene. The vinyl aromatic monomer is polymerized in the presence of a polymerization catalyst to polymerize
25 the vinyl aromatic monomer with the polyphenylene ether to form the interpolymer beads. Optionally, a cross-linking agent is added. A blowing agent is introduced under pressure into the thermoplastic resin beads. The cross-linking agent may be di-t-
30 butyl peroxide, t-butyl cumyl peroxide, dicumyl peroxide, alpha, alpha-bis(t-butyl peroxy)p-di-isopropylbenzene, 2,5-dimethyl-2,5-di(t-butyl peroxy)hexyne-3, 2,5-dimethyl-2,5-di(benzoyl peroxy)hexane, and t-butyl peroxy isopropyl
35 carbonate. Here again, when a cross-linking agent is used, the polyphenylene ether resin has a gel

5 content of at least about 24% by weight. Since the morphology of the polyphenylene ether resin is amorphous, the resin generally may have poor environmental stress crack resistance (ESCR) properties.

10 U.S. Patent Nos. 4,303,756 and 4,303,757 to Kajimura, et al. and assigned to Sekisui Kaseihin Kogyo Kabushiki Kaisha disclose a process for producing expandable thermoplastic resin beads. The process comprises suspending in an aqueous medium
15 20% to 70% by weight of a random copolymer of propylene and ethylene (U.S. Patent No. 4,303,756) or polypropylene resin particles (U.S. Patent No. 4,303,757), and 30% to 80% by weight of a vinyl aromatic monomer such as styrene; polymerizing the
20 vinyl aromatic monomer in the presence of a polymerization catalyst to graft the vinyl aromatic monomer onto the backbone of polypropylene; and, optionally, adding a cross-linking agent to form graft-copolymerized thermoplastic resin beads; and
25 introducing a blowing agent into the thermoplastic resin beads.

 In general, the interpolymer resin particles of the above prior art generally are expandable thermoplastic resin particles having a high gel
30 content of about 10% to 45% by weight in at least one instance and in the other instances about 24% by weight which limits the processability of the beads or particles when converted into articles such as solid sheet, film, etc. through processing
35 techniques such as sheet extrusion, rotomolding, thermoforming, compression molding, injection

5 molding, blown film extrusion, and direct-injection
foamed sheet extrusion. Also, in general the
interpolymer resin particles of the prior art are
impregnated with a blowing agent in a further
suspension process to form foamable or expandable
10 particles that are used for foam moldings.

Expandable interpolymers of polypropylene and
polyvinyl aromatic monomers are also disclosed in
Kent D. Fudge U.S. Patent Nos. 4,622,347; 4,677,134
and 4,692,471; and in Bartosiak et al U.S. Patent
15 No. 4,647,593, all of which are assigned to Atlantic
Richfield Company. These interpolymers may be
prepared according to the teachings of the aforesaid
U.S. Patent No. 4,303,756. The interpolymers are
made expandable by impregnating them with a blowing
20 agent. The particles are expanded under normal
conditions for polystyrene particles to low density,
fine cell structure foams by viscbreaking the
interpolymers to a melt flow (Condition L) of at
least double their original value and adding a
25 lubricant and cell control agent while maintaining
orientation stress in the final product at a
minimum. Since the interpolymer particles of these
aforesaid patents may be prepared according to U.S.
Patent No. 4,303,756, the interpolymer particles
30 have the same limitations outlined in the above
discussion of the '756 patent in that the
interpolymer particles generally have a high gel
content, i.e. about 24% by weight. These viscbroken
interpolymer particles are impregnated with a
35 blowing agent in a further suspension process to

5 produce foamable or expandable particles for foam moldings.

SUMMARY OF THE INVENTION

10 The invention overcomes the above-described disadvantages of the prior art.

The present invention provides non-expandable interpolymer resin particles with a crystalline morphology and having little or no gel content, whereby the processing characteristics of the particles in fabrication equipment for forming articles, such as sheet or film or foam, is improved. The gel content ranges from about 0 to about 1.5% by weight, preferably, from about 0 to about 0.8% by weight, and more preferably, from about 0 to about 0.5% by weight based on the weight of the interpolymer particles. The VICAT softening temperature of the interpolymer resin particles ranges from about 90°C to about 115°C, and the melt index values range from about 0.2 to about 35.0 g/10 minutes (Condition G).

25 The invention provides a process for producing interpolymer resin particles comprising: a) suspending in an aqueous medium from about 20% to 60% by weight of polyolefin resin particles having a VICAT softening temperature greater than 85°C and a melt flow of about 2.1 g/10 minutes (Condition I, 190°C, 2.16 kg); b) minimizing or eliminating the cross-linking of the polyolefin resin particles; c) adding to the aqueous suspension from about 40 to 80% by weight of a vinyl aromatic monomer and a polymerization initiator for polymerizing the vinyl

5 aromatic monomer within the polyolefin resin particles; and d) polymerizing said vinyl aromatic monomer in said polyolefin resin particles to form said interpolymer resin particles having a gel content ranging from about 0 to about 1.5% by weight, based on the weight of the interpolymer resin particles.

10 The invention provides a process for producing articles with improved processability and/or ESCR characteristics by using the process in the above paragraph to produce interpolymer particles and using these particles in polymer processing techniques such as those described herein.

15 According to a further aspect of the invention, there are provided interpolymer resin particles comprising from about 20% to 60% by weight of polyolefin particles and from about 40 to 80% by weight of polymerized vinyl aromatic monomer and which particles have a gel content ranging from about 0 to about 1.5% by weight, based on the weight of the interpolymer resin particles.

20 In the invention, the degree of cross-linking of the polyolefin in the interpolymer resin particles is minimal or eliminated. This can be accomplished by eliminating the use of the high temperature cross-linking agent, e.g. dicumyl peroxide for the polyolefin, e.g. polyethylene. The result is an interpolymer resin having a gel content ranging from 0 to 1.5% by weight based on the weight of the interpolymer resin particles. This feature of the invention in conjunction with the interpolymer particles having a VICAT softening temperature

5 ranging from about 90°C to about 115°C and a
resultant melt index ranging from about 0.2 to about
35.0 g/10 minutes (Condition G) improves the
processing characteristics or the processability of
the interpolymer resins.

10 The interpolymer resin particles preferably do
not contain a blowing agent that is impregnated in
the resin particles through a further suspension
process. Therefore, the resultant interpolymer
resin particles are not expandable or foamable
15 particles as those of the above prior art for use in
foam moldings.

The interpolymer resin particles are generally
intended to be used to produce articles through
polymer processing techniques, such as sheet
20 extrusion, injection molding, thermoforming,
compression molding, blown film extrusion,
rotomolding, and direct-injection foamed sheet
extrusion at low power consumption and no melt
fracture. A sheet or layer formed from the
25 interpolymer particles of the invention may be used
as a tie layer in multi-layer structures.

Unlike the interpolymer particles of the prior
art, the interpolymer particles of the invention are
easily extruded into solid sheets, films, etc. and
30 injection molded into articles with improved solvent
resistance (ESCR) compared to articles made solely
from polystyrene or polyethylene. The flexural and
tensile properties of the articles formed from the
interpolymer resin particles of the invention have
35 values that range between those values for articles
made solely from polystyrene and those values for

5 articles made solely from low-density polyethylene,
while the thermal and impact properties approach
that of pure polystyrene.

The interpolymer resin particles of the
invention are particularly advantageous in end-use
10 applications such as solid sheets, foamed sheets,
foamed boards, injection molded articles, barrier
films, and as a tie layer in multi-layer structures.
In a multi-layer structure, the adjacent layer or
layers are generally polyethylene, polystyrene or
15 high impact polystyrene. These formed articles have
improved properties such as those discussed in the
preceding paragraph.

The interpolymer particles may be easily
adapted to foam applications where a blowing agent
20 is mixed into the molten interpolymer resin
particles in conventional extrusion foaming
equipment to produce foamed sheet or foamed board
with improved cushioning and improved ESCR
properties compared to polystyrene foamed sheet or
25 foamed board applications. U.S. Patent No. 6,166,099
issuing to Steven M. Krupinski (NOVA Chemicals Inc.,
assignee) on December 26, 2000 teaches in columns 7
and 8, a conventional extrusion process and related
equipment, which may be used for foam applications
30 of the interpolymer resin particles, the teachings
of which are incorporated herein by reference.

The interpolymer resin particles generally are
pellets formed through a suspension process, which
pellets are then converted into film, sheet etc.
35 through an extrusion process, or injection molding
or thermoforming. The pellets formed in the

5 suspension process generally weight between about 8 milligrams to about 20 milligrams.

According to a further aspect of the invention, the resin particles may be used for the fabrication of articles via (extrusion or injection) blow
10 molding, injection molding, rotomolding, profile extrusion, solid sheet extrusion, thermoforming, and direct-injection foamed sheet extrusion.

A still further aspect of the invention is to provide a polymer composition comprising the
15 interpolymer particles and a second polymer such as polyethylene and polystyrene.

It is therefore, an object of the present invention to provide interpolymer resin particles or a polymer composition comprising the interpolymer
20 particles that result in improved processability to form articles with improved ESCR characteristics, improved vapor barrier characteristics, and/or improved physical properties.

It is a further object of the present invention to provide interpolymer resin particles that have
25 little or no gel content, i.e. ranging from about 0 to about 1.5% by weight based on the weight of the interpolymer resin particles.

These and other objects of the present invention will be better appreciated and understood
30 by those skilled in the art from single Figure and the following description and appended claims.

BRIEF DESCRIPTION OF THE DRAWING

35 The Figure is a graph showing the results when Composition A and Composition B are plotted against

5 Total Energy (ft. lbs.) (DYNATUP) versus the weight percentages of polystyrene, which is a component of Composition A and Composition B.

DETAILED DESCRIPTION OF THE INVENTION

10 The term "polyolefin" as used herein can be polyethylene, polypropylene, thermoplastic olefins (TPO'S), or thermoplastic elastomers (TPE'S) resins. Preferably, in the invention the polyolefin is a polyethylene resin or polypropylene resin.

15 The term "polyethylene resin" as used in the present specification and the appended claims, is meant to include not only a homopolymer of ethylene, but also an ethylene copolymer composed of at least 50 mole %, preferably at least 70 mole %, of an
20 ethylene unit and a minor proportion of a monomer copolymerizable with ethylene, and a blend of at least 50% by weight, preferably at least 60% by weight, of the ethylene homopolymer or copolymer with another polymer.

25 Examples of monomers copolymerizable with ethylene are vinyl acetate, vinyl chloride, propylene, butene, hexene, acrylic acid and its esters, methacrylic acid and its esters. The other polymer that may be blended with the ethylene
30 homopolymer or copolymer may be any polymer compatible with it. Examples are polypropylene, polybutadiene, polyisoprene, polychloroprene, chlorinated polyethylene, polyvinyl chloride, a styrene/butadiene copolymer, a vinyl
35 acetate/ethylene copolymer, an acrylonitrile/butadiene copolymer, a vinyl

5 chloride/vinyl acetate copolymer, etc. Especially preferred species are polypropylene, polybutadiene and styrene/butadiene copolymer.

Examples of polyethylene that can be advantageously employed in the present invention are
10 low-, medium-, and high-density polyethylene, an ethylene vinyl acetate copolymer, an ethylene/propylene copolymer, a blend of polyethylene and polypropylene, a blend of polyethylene and an ethylene/vinyl acetate
15 copolymer, and a blend of polyethylene and an ethylene/propylene copolymer.

The polyethylene resin particles used to form the interpolymer resin particles of the invention have a melt index (MI) of about 2.1g/10 minutes
20 under Condition I, 190°C, 2.16kg (equivalent to 11.9 g/10 minutes under Condition G, 230°C 5.0 kg); a number average molecular weight of 20,000 to 60,000; an intrinsic viscosity, at 75°C in xylene, of 0.8 to 1.1; a density of 0.910 to 0.940 g/cm³, and a VICAT
25 softening temperature greater than 85°C. In the embodiments herein, the polyethylene resin particles have a VICAT softening temperature of about 94°C, a melt index (MI) of 2.1 g/10 minutes (Condition I, 190°C, 2.16 kg which is equivalent to 11.9 g/10
30 minutes under Condition G, 230°C, 5.0kg), a density of 0.919 g/cm³, and a weight of about 20 milligrams. A suitable low-density polyethylene (LDPE) is that obtained from NOVA Chemicals Inc. under the trade name LA-0218-AF.

35 The term "polypropylene resin" as used herein is meant to denote not only a homopolymer of

5 propylene, but also a block copolymer containing
polypropylene in an amount of at least 50% by weight
and another polyolefin, and a mixture of at least
50% by weight of polypropylene and another
polyolefin. In the present invention, the other
10 polyolefin includes, for example, polyethylene, an
ethylene/vinyl acetate copolymer, an ethylene/vinyl
chloride copolymer, an ethylene/propylene rubber,
polyisobutylene, butyl rubber, styrene/butadiene
rubber, polybutene, and polybutadiene. Similar to
15 the teachings of Kajimura, et al. U.S. Patent No.
4,303,757, the polypropylene resin can be used in
particulate form, preferably, in the form of
spheres, flattened particles or pellets having a
diameter of about 0.5 to 10mm in order to cause
20 rapid absorption of the vinyl aromatic monomer.

The amount of polyolefin used in the invention
ranges from about 20% to about 60% by weight based
on the weight of the interpolymer resin particles.

A vinyl aromatic monomer is used in the
25 invention. Examples of vinyl aromatic monomers are
styrene, alpha-methylstyrene, ethylstyrene,
chlorostyrene, bromostyrene, vinyltoluene,
vinylbenzene, and isopropylxylene. These monomers
may be used either alone or in admixture. A mixture
30 of at least 0.1% of the vinyl aromatic monomer and a
monomer copolymerizable with it, such as
acrylonitrile, methyl methacrylate, butyl acrylate,
or methyl acrylate can also be used. As used herein,
the term "vinyl aromatic monomer" means a vinyl
35 aromatic monomer used alone or in admixture.

5 In an embodiment, the vinyl aromatic monomer preferably is styrene polymerized within the polyolefin resin particles.

 The amount of vinyl aromatic monomer ranges from about 40% to about 80% by weight based on the weight of the interpolymer resin particles.

10 In general, the interpolymer resin particles are formed as follows: in a reactor, the polyolefin resin particles are dispersed in an aqueous medium prepared by adding 0.01 to 5%, preferably 2 to 3%,
15 by weight based on the weight of the water of a suspending or dispersing agent such as water soluble high molecular materials, e.g., polyvinyl alcohol, methyl cellulose, and slightly water soluble
inorganic materials, e.g., calcium phosphate or
20 magnesium pyrophosphate, and then the vinyl aromatic monomers are added to the suspension and polymerized inside the polyolefin resin particles to form an interpenetrating network of polyolefin and vinyl aromatic monomers.

25 Basically any of the conventionally known and commonly used suspending agents for polymerization may be employed. These agents are well known in the art and may be freely selected by one skilled in the art. Water is used in an amount generally from 0.7
30 to 5, preferably 3 to 5 times that of the starting polyolefin particles added to the aqueous suspension, on a weight basis.

 When the polymerization of the vinyl aromatic monomer is completed, the polymerized vinyl aromatic resin is uniformly dispersed inside the polyolefin
35 particles.

5 The resulting interpolymer resin particles may
be used as raw materials in producing articles such
as sheets, rods, pipes, and film using an extruder,
or in producing articles via injection molding, or
thermoforming processes. A blowing agent may be
10 introduced into the interpolymer resin particles to
form foamed sheet via an extruder.

 It has been found by the inventor that
unexpected results are derived when the interpolymer
particles of the invention are produced without a
15 cross-linking agent. That is, in the suspension
process no cross-linking agent such as a high
temperature initiator, e.g. dicumyl peroxide is
added to the polyolefin, e.g. polyethylene or
polypropylene. Since the polyolefin particles are
20 not cross-linked, the interpolymer particles have
very little or no gel content, i.e. a gel content
ranging from about 0 to about 1.5%, preferably from
about 0 to about 0.8% by weight, and more preferably
from about 0 to about 0.5% by weight, based on the
25 weight of the particles. The VICAT softening
temperature for the interpolymer resin particles
ranges from about 90°C to about 115°C, and
preferably from about 90°C and about 105°C.

 The viscosity of the interpolymer particles of
30 the invention is reduced compared to the
interpolymer particles of the prior art having
relatively high gel content. The reduced viscosity
results in improved processing characteristics or
processability of the particles so that better
35 quality shaped articles can be manufactured by

5 extrusion and/or by injection processes as discussed
herein above.

10 In a direct injection foaming process, a
blowing agent may be introduced into a molten mass
of the interpolymer particles of the invention to
produce a foamed article, such as foam sheet, foam
board, etc. Suitable blowing agents include
aliphatic hydrocarbons such as n-propane, n-butane,
iso-butane, n-pentane, iso-pentane, n-hexane, and
neopentane, cycloaliphatic hydrocarbons such as
15 cyclobutane and cyclopentane, and halogenated
hydrocarbons such trichlorofluoromethane,
dichlorofluoromethane, dichlorodifluormethane,
chlorodifluoromethane and
dichloroetetrafluoroethane, etc. HFC's such as
20 tetrafluoroethane, difluoroethane or HCFC's such as
chlorodifluoroethane may be used. These blowing
agents may be used alone or as mixtures.

25 A preferred amount of the blowing agent is in
a range of about 2 to about 15 by weight based on
the weight of the formed interpolymer particles.
The specific types of blowing agents are taught in
U.S. Patent No. 3,959,189, the teachings of which
are incorporated by reference. Preferably, the
interpolymer resin particles are not impregnated
30 with the blowing agent in a suspension process;
instead the blowing agent is added during the
formation of foam products in a conventional manner
and in a conventional extrusion process, more about
which is discussed herein after.

35 A process for producing the interpolymer
particles of the invention is conveniently carried

5 out as follows. The polyolefin particles are
suspended in an aqueous medium containing a
dispersing agent. The dispersing agent may be, for
example, polyvinyl alcohol, methylcellulose, calcium
phosphate, magnesium pyrophosphate, calcium
10 carbonate, tricalcium phosphate, etc. The amount of
dispersing agent employed is 0.01 to 5% by weight
based on the amount of water.

A surfactant may be added to the aqueous
medium. Generally, the surfactant is used to lower
15 the surface tension of the suspension and to help
emulsify the water/vinyl aromatic monomer mixture in
the initiator and wax mixes, if used. The aqueous
medium is generally heated to a temperature at which
the vinyl aromatic monomer can be polymerized, i.e.
20 from about 60°C to about 120°C over a period of time,
for example, 12 to 20 hours. Over this 12 to 20
hour period, the vinyl aromatic monomer and the low
temperature initiators for polymerizing the vinyl
aromatic monomer are added to the resulting
25 suspension containing the polyolefin particles
dispersed therein. These materials may be added all
at one time, or gradually in small portions. The
suspension is cooled to room temperature. The
interpolymer particles are acidified to remove
30 surface suspending agent, dewatered, screened, and
dried in a fluidized bed dryer.

Polymerization of the vinyl aromatic monomer
occurs in the polyolefin particles. Examples of
suitable initiators include organic peroxy
35 compounds, such as peroxides, peroxy carbonates and
peresters. Typical examples of these peroxy

5 compounds are C₆₋₂₀ acyl peroxides, such as decanoyl
peroxide, benzoyl peroxide, lauroyl peroxide,
octanoyl peroxide, stearyl peroxide, peresters, such
as t-butyl perbenzoate, t-butyl peracetate, t-butyl
perisobutyrate, t-butylperoxy 2-ethylhexyl
10 carbonate, or combinations thereof. Other
initiators, different from peroxy compounds, are
also possible, as for example α , α' -
azobisisobutyronitrile and azobis-
dimethyvaleronitrile.

15 The initiators are generally used in an amount
of about 0.05 to 2 percent by weight, preferably 0.1
to 1 percent by weight, based on the weight of the
vinyl aromatic monomer.

20 These initiators may be used alone or in
combination of two or more initiators. These
initiators can be dissolved in the vinyl aromatic
monomers, which are to be absorbed in the polyolefin
particles. These teachings appear in the aforesaid
U.S. Patent Nos. 3,959,189. The initiators can be
25 dissolved in a solvent. Such solvents include
toluene, benzene and 1,2-dichloropropane, etc.

30 The suspension polymerization is carried out in
the presence of suspension stabilizers. Suitable
suspension stabilizers are well known in the art and
comprise organic stabilizers, such as poly (vinyl
alcohol), gelatine, agar, polyvinyl pyrrolidine,
polyacrylamide; inorganic stabilizers, such as
alumina, bentonite, magnesium silicate; surfactants,
such as sodium dodecyl benzene sulfonate; or
35 phosphates, like tricalciumphosphate, disodium-

5 hydrogen phosphate, optionally in combination with any of the stabilizing compounds mentioned earlier.

The amount of stabilizer may suitably vary from about 0.01% to about 5.0% by weight, based on the weight of the aqueous phase.

10 The polyolefin particles, and/or the interpolymer resin particles of the invention may contain an anti-static additive; a flame retardant; a colorant or dye; a filler material; stabilizers; and plasticizers, such as white oil.

15 The interpolymer resin particles of the invention may suitably be coated with compositions comprising silicones, metal or glycerol carboxylates, suitable carboxylates are glycerol mono-, di- and tri-stearate, zinc stearate, calcium
20 stearate, and magnesium stearate; and mixtures thereof. Examples of such compositions may be those disclosed in GB Patent No. 1,409,285 and in Stickley U. S. Patent No. 4,781,983. The coating composition can be applied to the interpolymer resin particles
25 via dry coating or via a slurry or solution in a readily vaporizing liquid in various types of batch and continuous mixing devices. The coating aids in transferring the interpolymer resin particles easily through the processing equipment.

30 The interpolymer resin particles may contain other additives such as chain transfer agents, suitable examples including C₂₋₁₅ alkyl mercaptans, such as n-dodecyl mercaptan, t-dodecyl mercaptan, t-butyl mercaptan and n-butyl mercaptan, and other
35 agents such as pentaphenyl ethane and the dimer of α -methyl styrene, as well as nucleating agents, such

5 as polyolefin waxes. The polyolefin waxes, i.e.
polyethylene waxes, have a weight average molecular
weight of 250 to 5,000, which are typically finely
divided through the polymer matrix in a quantity of
0.01 to 2.0% by weight, based on the amount of resin
10 composition. The interpolymer resin particles may
also contain from 0.1 to 0.5% by weight, talc,
organic bromide-containing compounds, and polar
agents as described in e.g. WO 98/01489 which
comprise isalkylsulphosuccinates, sorbital-C₈ - C₂₀ -
15 carboxylates, and C₈ - C₂₀- alkylxylene sulphonates.

The interpolymer resin particles may be used in
extrusion processing. For example, the particles
may be fed into an extruder, and then extruded as a
single layer or co-extruded into multi-layer
20 structures, e.g. sheet or film. Optionally, a
blowing agent may be forced into the particles
passing through the extruder and a foamed sheet,
board, or rod may be formed. In further embodiments,
the interpolymer resin particles may be used in
25 injection molding or may be thermoformed into
desired shapes in a manner well known to those
skilled in the art.

Alternatively, an extruded sheet or film
produced from the interpolymer resin particles may
30 be used as a tie layer in a multi-layer structure.
A co-extrusion process may be employed wherein the
interpolymer resin particles are extruded between
sheets of incompatible polymers, e.g. a sheet made
of polystyrene or polystyrene and rubber, e.g. high
35 impact polystyrene (HIPS), and a sheet made of
polyethylene, thereby giving improved adhesion to

5 the multi-layer structure. It is hypothesized that
the polyethylene resin in the interpolymer resin
particles creates a chemical bond with the
polyethylene in the polyethylene layer and that the
polystyrene in the interpolymer resin particles
10 creates a chemical bond with the polystyrene in the
polystyrene layer.

This improved adhesion becomes important when a
cap layer of polyethylene is extruded onto
polystyrene sheet to improve ESCR, i.e.
15 environmental stress crack resistance. A further
example may pertain to the food industry where in
view of FDA requirements food cannot come into
contact with the substrate layer of the food
container.

20 When polypropylene is used as the base
polyolefin resin in the interpolymer particles of
the invention, it may be preferable to viscbreak the
polypropylene prior to, during or after interpolymer
formation.

25 Viscbreaking is the intentional chain scission
of polypropylene to produce lower molecular weight,
a narrower molecular weight distribution, a slow
crystallization rate, and faster molecular
relaxation time in the molten state. Viscbreaking
30 can be accomplished by extrusion under high shear to
mechanically degrade the higher molecular weight
chains as taught in the above Fudge U.S. Patent Nos.
4,622,347 and 4,677,134 or can be accomplished by
the use of peroxides as taught in the aforesaid
35 Fudge U.S. Patent No. 4,692,471 during the formation
of the interpolymer resin particles. The peroxide

5 blended with the polymers may be any of the
compounds having a 10-hour half-life temperature of
between 100°C and 130°C such as dicumyl peroxide
(117°C) or 1,3-bis (α -tert-butylperoxyisopropyl)
10 benzene (116°C). Examples 7 and 8 use dicumyl
peroxide to viscbreak the polypropylene of the
interpolymer resin particles of the invention.

15 The interpolymer resin particles of the
invention may be combined with a second polymer to
form a polymer composition which may then be used to
form articles via processing, for example, sheet
15 extrusion, rotomolding, thermoforming, compression
molding, injection molding, and blown film
extrusion. The second polymer may be selected from
the group consisting of polyethylene and
20 polystyrene. In this composition, the interpolymer
resin particles may be present in an amount ranging
from about 0.1% by weight to about 99.9% by weight
and the second polymer may be present in an amount
ranging from about 99.9% by weight to about 0.1% by
25 weight. In an embodiment of the invention, the
interpolymer resin particles are present in an
amount ranging from about 10% by weight to about 90%
by weight and the second polymer is present in an
amount ranging from about 90% to about 10% by weight
30 based on the weight of the interpolymer resin.

35 The following examples are intended to aid in
understanding the present invention, however, in no
way, should these examples be interpreted as
limiting the scope thereof.

ExamplesExample 1

This Example 1 relates to styrene-polyethylene interpolymer resin particles comprised of 60% by weight polystyrene and 40% by weight of low-density polyethylene, based on the weight of the interpolymer resin particles. No dicumyl peroxide cross-linking agent was added to the formulation.

A mixture of 520 pounds of de-ionized water, 9.6 pounds of tri-calcium phosphate as a suspending agent, and 27 grams of a strong anionic surfactant were charged to a polymerization reactor with the agitator running at 88 rpm to prepare an aqueous medium. The surfactant was Nacconol® 90 (Stephan Chemical Co.), which is sodium n-dodecyl benzene sulfonate. The aqueous medium was heated to about 91°C and held for about 10 minutes. Then 112 pounds of low density polyethylene (LDPE) pellets (LA-0218-AF from NOVA Chemicals Inc.), each weighing about 20 milligrams, having a melt index at condition I (190°C, 2.16kg) of 2.1 g/10 minutes, and a VICAT softening point of about 93°C were added to the aqueous medium. This suspension of beads and water continued to be stirred at 88 rpm. The low temperature polystyrene initiators, i.e. 373 grams of benzyl peroxide (BPO) (75% active) and 70 grams of tertiary butyl perbenzoate (TBP) were dissolved in 84 pounds of styrene monomer to prepare a monomer solution, and this mixture was pumped into the reactor over 200 minutes. A second batch of 84 pounds of pure styrene was then added to the reactor over 100 minutes at a temperature of 91°C. The

5 reactor contents were held at 91°C for an additional
90 minutes to allow the styrene to soak into and
react within the polyethylene. Then the reactor
contents were heated to 140°C over 2 hours and held
for an additional 4 hours to polymerize the
10 remaining styrene into polystyrene within the
polyethylene matrix.

After polymerization, the reactive mixture was
cooled and hydrochloric acid was added to dissolve
the suspending agents. The resin particles were
15 then washed and dried.

The average gel content for two samples of the
resin particles was 0.65 weight % based on the
weight of the formed interpolymer resin particles.
The melt index was 1.046 g/10 minutes at condition G
20 (230°C and 5.0 kg).

Example 2

This Example 2 relates to interpolymer styrene-
polyethylene interpolymer resin particles comprised
of 70% by weight polystyrene and 30% by weight low-
25 density polyethylene, based on the weight of the
interpolymer resin particles. No dicumyl peroxide
cross-linking agent was added to the formulation.

A mixture of 520 pounds of deionized water, 9.6
pounds of tri-calcium phosphate as a suspending
30 agent, and 27 grams of a strong anionic surfactant
(Nacconol® 90) were charged to a polymerization
reactor with the agitator running at about 88 rpm to
prepare an aqueous medium. The aqueous medium was
heated to about 91°C and held for about 10 minutes.
35 Then 84 pounds of low-density polyethylene pellets
(LA-0218-AF) were suspended in the aqueous medium.

5 The suspension continued to be stirred at 88 rpm.
The low temperature polystyrene initiators, i.e. 356
grams of benzyl peroxide (BPO) and 66.8 grams of
tertiary butyl perbenzoate (TBP) were dissolved in
98 pounds of styrene monomer to prepare a monomer
10 solution, and this mixture was pumped into the
reactor over 200 minutes. A second batch of 98
pounds of pure styrene was then added to the reactor
over 100 minutes at a temperature of 91°C. The
reactor contents were held at 91°C for an additional
15 90 minutes to allow the styrene to soak into and
react within the polyethylene. Then the reactor
contents were heated to 140°C over 2 hours and held
at this temperature for an additional 4 hours to
polymerize the remaining styrene into polystyrene
20 within the polyethylene matrix.

After polymerization, the reactive mixture was
cooled and hydrochloric acid was added to dissolve
the suspending agents. The resin particles were
then washed and dried.

25 The average gel content for two samples of
resin particles was 0.45% by weight based on the
weight of the particles. The melt index was .501g/10
minutes at condition G (230°C and 5.0kg).

Example 3

30 This Example 3 relates to styrene-polyethylene
interpolymer resin particles comprised of 50% by
weight polystyrene and 50% by weight low-density
polyethylene, based on the weight of the
interpolymer resin particles. No dicumyl peroxide
35 cross-linking agent was added to the formulation.

5 A mixture of 520 pounds of de-ionized water,
9.6 pounds of tri-calcium phosphate as a suspending
agent, and 27 grams of a strong anionic surfactant
(Nacconol® 90) were charged to a polymerization
reactor with the agitator running at about 88 rpm to
10 prepare an aqueous medium. The aqueous medium was
heated to about 91°C and held for about 10 minutes.
Then 140 pounds of low-density polyethylene pellets
(LA-0218-AF) were suspended in the aqueous medium.
The suspension continued to be stirred at 88 rpm.
15 The low temperature polystyrene initiators, i.e. 350
grams of benzyl peroxide (BPO) and 65.63 grams of
tertiary butyl perbenzoate (TBP), were dissolved in
70 pounds of styrene monomer to prepare a monomer
solution, and this mixture was pumped into the
20 reactor over 200 minutes. A second batch of 70
pounds of pure styrene was then added to the reactor
over 100 minutes at a temperature of 91°C. The
reactor contents were held at 91°C for an additional
90 minutes to allow the styrene to soak into and
25 react within the polyethylene. Then the reactor
contents were heated to 140°C over 2 hours and held
for an additional 4 hours to polymerize the
remaining styrene into polystyrene within the
polyethylene matrix. After polymerization, the
30 reactive mixture was cooled and hydrochloric acid
was added to dissolve the suspending agents. The
resin particles were then washed and dried.

35 The average gel content for two samples of
resin particles was 0.69% by weight based on the
weight of the formed interpolymer resin particles.

5 The melt index was 1.022g/10 minutes at condition G
(230°C and 5.0kg).

Example 4

10 This Example 4 is similar to Example 1 in that
a styrene-polyethylene interpolymers with 60% by
weight polystyrene and 40% by weight low density
polyethylene based on the weight of the interpolymers
particles was produced. In this Example 4, however,
a chain transfer agent was used in an attempt to
increase the melt flow rate of the interpolymers
resin.

15 Alpha methyl styrene dimer (a chain transfer
agent) in an amount of 163 grams, i.e. about 0.20
parts per hundred of styrene was added to the
suspension with the benzyl peroxide (BPO) and the
tertiary butyl perbenzoate (TBP).

20 The average gel content for two samples of the
resin particles was 1.01% by weight based on the
weight of the formed interpolymers resin particles.
The melt index was 2.688g/10 minutes (Condition G).
25 These results demonstrate that when using a chain
transfer agent without a cross-linking agent the
melt index was increased compared to Example 1.

Example 5

30 In this Example 5, interpolymers resin particles
were produced comprising 60% by weight polystyrene
and 40% by weight ethylene vinyl acetate copolymer
(EVA), based on the weight of the resin particles.
No high temperature cross-linking agent, i.e.
dicumyl peroxide initiator was added.

35 A mixture of 380 pounds of de-ionized water, 13
pounds of tri-calcium phosphate as a suspending

5 agent, and 8.6 grams of Nacconol® 90 anionic
surfactant were charged to a polymerization reactor
with the agitator running at about 102 rpm to
prepare an aqueous medium.

10 The aqueous medium was heated to about 60°C and
held for about 30 minutes. Then 125 pounds of a
low-density polyethylene vinyl acetate (EVA) pellets
containing 4.5% by weight vinyl acetate and 95.5% by
weight ethylene (NA 480 from Equistar Chemicals, LP,
Houston, Texas) and having a density of about 0.923
15 g/cc and a melt index of 0.25 g/10 minutes
(Condition I, 190°C, 2.16kg) were suspended in the
aqueous medium. The reactor temperature was
increased to 85°C. The low temperature polystyrene
initiators, i.e. 246 grams of benzoyl peroxide (BPO)
20 and 30 grams of tertiary butyl perbenzoate (TBP),
were dissolved in 22.6 pounds of styrene monomer to
prepare a monomer solution, and this mixture was
pumped into the reactor over 96 minutes. A second
batch of 146 pounds of pure styrene and 5.0 lbs of
25 butyl acrylate was then added to the reactor over
215 minutes. Then the reactor contents were heated
and held at 140°C for over 8 hours to finish the
polymerization of styrene within the polyethylene
matrix.

30 After polymerization was completed, the
reactive mixture was cooled and removed to a wash
kettle where muriatic acid (HCl) was added to
dissolve the suspending agents from the pellet
surfaces. The pellets were then washed and dried.

35 The average gel content for two samples of the
resin pellets was 0.46 weight % based on the weight

5 of the formed interpolymer resin particles. The
melt index of the pellets was 0.21g/10 minutes
(Condition G).

Example 6

10 This Example 6 relates to interpolymer resin
particles comprising 70% by weight polystyrene based
on the weight of the interpolymer resin particles,
and 30% by weight of ethylene vinyl acetate
copolymer (EVA). No high temperature cross-linking
agent, i.e. no dicumyl peroxide initiator was added.
15 The process for making the particles was similar to
that for Example 5. The low-density polyethylene
vinyl acetate (EVA) used in Example 5 was the same
as used in Example 6.

20 A mixture of 411 pounds of de-ionized water,
9.8 pounds of tri-calcium phosphate as a suspending
agent, and 6.5 grams of anionic surfactant
(Nacconol® 90) were charged to a polymerization
reactor with the agitator running at about 102 rpm
to prepare an aqueous medium. The aqueous medium
25 was heated to about 60°C and held for about 30
minutes. Then 87 pounds of the low-density ethyl
vinyl acetate pellets were suspended in the aqueous
medium. The reactor temperature was increased to
85°C. The low temperature polystyrene initiators,
30 i.e. 246 grams of benzoyl peroxide (BPO) and 30
grams of tertiary butyl perbenzoate (TBP), were
dissolved in 22.6 pounds of styrene monomer to
prepare a monomer solution, and this mixture was
pumped into the reactor over 96 minutes. A second
35 batch of 146 pounds of pure styrene and 5.0 lbs of
butyl acrylate was then added to the reactor over a

5 period of 215 minutes. Then the reactor contents were heated and held at 140°C for over 8 hours to finish the polymerization of styrene within the polyethylene matrix.

10 After polymerization was completed, the reactive mixture was cooled and removed to a wash kettle where muriatic acid (HCl) was added to dissolve the suspending agents from the pellet surfaces. The pellets were then washed and dried.

15 The average gel content for two samples of the resin pellets was 0.32% by weight based on the weight of the formed interpolymer resin particles. The melt index of the pellets was 0.25g/10 minutes (Condition G).

20 Examples 7 and 8 below show that the use of dicumyl peroxide for viscbreaking purposes increases the melt index of the resin.

Example 7

25 This Example 7 relates to interpolymer resin particles comprising 60% by weight polystyrene based on the weight of the interpolymer resin particles, and 40% by weight of polypropylene. Dicumyl peroxide was added to viscbreak the polypropylene.

30 A mixture of 520 pounds of deionized water, 9.6 pounds of tri-calcium phosphate as a suspending agent, and 27 grams of Nacconol® 90 were charged to a polymerization reactor with the agitator running at about 88 rpm to prepare an aqueous medium. The aqueous medium was heated to about 91°C and held for about 10 minutes. Then 112 pounds of polypropylene pellets (Huntsman P5M4K-046), each weighing about 20
35 milligrams and having a MI of 25.5 g/10 minutes

5 (Condition G) were suspended in the aqueous medium.
The suspension continued to be stirred at 88 rpm.
The low temperature polystyrene initiators, i.e. 473
grams of benzyl peroxide (BPO) and 145 grams of
tertiary butyl perbenzoate (TBP), and 173 grams of
10 dicumyl peroxide (for viscbreaking the
polypropylene) were dissolved in 84 pounds of
styrene monomer to prepare a monomer solution, and
this mixture was pumped into the reactor over 200
minutes. A second batch of 84 pounds of pure
15 styrene was then added to the reactor over 100
minutes at a temperature of 91°C. The reactor
contents were held at 91°C for an additional 90
minutes to allow the styrene to soak into and react
with the polypropylene. Then the reactor contents
20 were heated to 140°C for over 2 hours and held for an
additional 4 hours to polymerize the styrene into
polystyrene within the matrix of the polyethylene.

After polymerization, the reactive mixture was
cooled and removed, and an acid was added to
25 dissolve the suspending agents.

The average gel content for two samples of the
resin particles was 0.47% by weight based on the
weight of the formed interpolymer resin particles.
The melt index was 32.61g/10 minutes (Condition G).

30 Example 8

This Example 8 relates to interpolymer resin
particles comprising 70% by weight polystyrene based
on the weight of the interpolymer resin particles,
and 30% by weight of polypropylene. Dicumyl peroxide
35 was added to the formulation to viscbreak the

5 polypropylene. The process for producing the
interpolymer resins is similar to Example 7.

A mixture of 520 pounds of de-ionized water,
9.6 pounds of tri-calcium phosphate as a suspending
agent, and 27 grams of an anionic surfactant
10 (Nacconol® 90) were charged to a polymerization
reactor with the agitator running at about 88 rpm to
prepare an aqueous medium. The aqueous medium was
heated to about 91°C and held for about 10 minutes.
Then 112 pounds of polypropylene pellets (Huntsman
15 P5M4K-046) each weighing about 20 milligrams and
having a MI of 25.5 g/10 minutes (Condition G) were
suspended in the aqueous medium. The suspension
continued to be stirred at 88 rpm. The low
temperature polystyrene initiators, i.e. 475 grams
20 of benzyl peroxide (BPO) (for improved grafting) and
145 grams of tertiary butyl perbenzoate (TBP) (for
reducing the styrene residuals), and 173 grams of
dicumyl peroxide for viscbreaking the polypropylene
were dissolved in 98 pounds of styrene monomer to
25 prepare a monomer solution, and this mixture was
pumped into the reactor over 200 minutes. A second
batch of 98 pounds of pure styrene was then added to
the reactor over 100 minutes at a temperature of
91°C. The reactor contents were held at 91°C for an
30 additional 90 minutes to allow the styrene to soak
into and react within the polypropylene. Then the
reactor contents were heated to 140°C for over 2
hours and held for an additional 4 hours to
polymerize the styrene into polystyrene within the
35 matrix of the polypropylene.

5 After polymerization was completed, the reactive mixture was cooled and removed, and an acid was added to dissolve the suspending agents.

The average gel content for two samples was 0.41% by weight based on the weight of the formed interpolymer resin particles. The melt index was 21.92g/10 minutes (Condition G).

10 The particles produced in Examples 1-8 were oven dried at 120°F and then molded into plaques using an Engel Model 80 injection-molding machine. The mechanical and physical properties were measured and tested according to the standards set up by ASTM. These properties appear in Table 1 below.

15 As stated herein above, the flexural and tensile properties of the articles formed from the interpolymer resin particles of the invention have values that range between those values for articles made solely from polystyrene and those values for articles made solely from low-density polyethylene, while the thermal and impact properties of the articles made from the interpolymer resin particles approach that of pure polystyrene.

Table 1

Property	Example 1	Example 2	Example 3	Example 4	Example 5	Example 6	Example 7	Example 8
Flex Modulus (KSI)	200.52	256.47	170.46	222.63	211.19	269.25	303.89	348.76
Flex Stress@5% (KSI)	6.67	8.34	5.63	7.55	6.78	NA	9.14	9.08
Strain at Break(Auto)%	2.14	4.43	3.00	5.48	3.17	3.30	2.51	2.07
Stress at Break(Auto) (KSI)	3.69	5.09	3.32	4.54	4.97	4.91	4.88	5.43
YOUNGS Modulus (Auto) (KSI)	NA	281.92	NA	242.02	279.95	281.52	325.65	366.07
IZOD Impact Mean	0.404	0.233	0.490	0.446	0.430	0.338	0.174	0.150
DYNATUP-Total Energy (ft-lbs)	0.43	0.47	0.55	0.50	NA	NA	0.53	0.42
MI (Condition G)	1.046	.501	1.022	2.688	0.21	0.25	32.61	21.92
VICAT-Mean (°C)	101.00	104.8	99.00	101.6	NA	NA	110.2	108.7
Gel wt%(Average)	0.65	0.45	0.69	1.01	0.46	0.32	0.47	0.41

5

Example 9

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15

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Two and three layer sheet structures using the compositions and type of extruders shown in Table 2 were formed in a WELEX® sheet co-extrusion line. Each stream at temperatures between 430°F and 450°F was passed into an extrusion die head (WELEX® standard 54" flip lip) to form a continuous multi-layer sheet structure, which in turn, was passed through a roll stack at roll pressures of about 70psig and roll temperatures ranging between 180° to 190°F for a curing process. The multi-layer sheet structures were then visually inspected and qualitatively evaluated for adhesion, the results of which appear in Table 2. "Good" indicates no peeling of the layers. "Poor" indicates peeling of the layers.

Table 2

Extruder	Layer Structure	Thickness	Adhesion
Berlyn Mark II Mark I	SCLAIR 31E 50%SCLAIR 31E/50%HIPS5410 HIPS 5410	2 mil 2 mil 20 mil	Poor
Berlyn Mark II Mark I	SCLAIR 31E Example 4 (40%PE/60%PS) HIPS 5410	2 mil 2 mil 20 mil	Good
Berlyn Mark II Mark I	SCLAIR 31E Example 6 (70%PS/30%EVA) HIPS 5410	2 mil 2 mil 20 mil	Good
Berlyn Mark II Mark I	SCLAIR 61C 50%SCLAIR 61C/50%HIPS5410 HIPS 5410	2 mil 2 mil 20 mil	Poor
Berlyn Mark II Mark I	SCLAIR 31E 40%SCLAIR31E/HIPS5410/20% Example 6 (70%PS/30%EVA) HIPS 5410	2 mil 2 mil 20 mil	Poor
Berlyn Mark I	Example 5 (60%PS/40%EVA) HIPS 5410	2 mil 20 mil	Good
Berlyn Mark I	Example 6 HIPS 5410	2 mil 20 mil	Good
Berlyn Mark I	SCLAIR 31E HIPS 5410	2 mil 20 mil	Poor
Berlyn Mark I	SCLAIR 61C HIPS 5410	2 mil 20 mil	Poor

5 HIPS 5410 (Product of NOVA Chemicals Inc.- high impact
polystyrene comprised of styrene and polybutadiene.
SCLAIR 31E (Polyethylene-Melt Index 11.5)
SCLAIR 61C (Polyethylene-Melt Index 5.3)

10 This Example 9 shows that "good" adhesion
occurred when one of the layers of the multi-layer
sheet structures consisted of 100% of the
interpolymer resin particles of the invention, i.e.
Examples 4, 5, and 6.

Example 10

15 This Example 10 relates to the single Figure
that illustrates instrumented impact testing values
(DYNATUP-Total Energy in ft. lbs.) for Composition A
and Composition B. Composition A is the composition
of the invention comprising the interpolymer
20 particles of Example 1 (60%PS/40%PE) containing
polystyrene (NOVA 1210-crystal) in the weight
percentages shown in the single Figure.

25 Composition B is a mixture of low-density
polyethylene (NOVA-LA-0218-AF pellets) and
polystyrene in the weight percentages shown in the
Figure. Composition A and Composition B were
produced by dry blending, compounding in a single-
screw extruder, and the cut pellets were injection
30 molded into 3 inch diameter circular plaques for
impact testing. The DYNATUP values for Composition
A of the invention remained relatively constant as
the weight percentage of polystyrene increased,
while those for Composition B dropped as the weight
percentage of polystyrene increased. This indicates
35 improved compatibility between polyethylene and
polystyrene for the interpolymer particles of the
invention compared to pure physical blends of
polyethylene and polystyrene.

Example 11

This Example 11 illustrates the environmental stress crack resistance properties (ESCR) of the 70%PS/30%EVA (Example 6) resin vs. 100% Polystyrene (PS) crystal (NOVA 1510 grade). Ten specimens were used as control samples. The chemical resistance test results for molded samples are shown in Table 3.

Table 3

<u>70%PS/30%EVA</u>	0.5% Strain No oil exposure	0.5% Strain With oil exposure
Specimen Thickness	0.12 inches	0.12 inches
Specimen Width	0.51 inches	0.51 inches
Tensile Stress @ Break	5.1 kpsi	5.07 kpsi
Standard Deviation	0.1	0.1
Tensile Strain @ Break	2.6%	2.6%
Standard Deviation	0.2	0.2
Young's Modulus	268 kpsi	271 kpsi
Standard Deviation	10.4	8.6
Percentage Decrease in Mechanical Properties		
Tensile Stress @ Break	----	0
Tensile Strain @ Break	----	0
Young's Modulus	----	0
<u>100% PS CRYSTAL</u> (1510 NOVA grade)	0.5% Strain No Oil exposure	0.5% Strain With oil exposure
Specimen Thickness	0.125 inches	0.125 inches
Specimen Width	0.51 inches	0.51 inches
Tensile Stress @ Break	6.25 kpsi	1.34 kpsi
Standard Deviation	+/- 0.3	+/- 0.4
Tensile Strain @ Break	1.5%	0.4%
Standard Deviation	0.1	0.1
Young's Modulus	507.3 ksi	558.5 ksi
Standard Deviation	10.2	93.9
Percentage Decrease in Mechanical Properties		

Tensile Stress @ Break	----	80
Tensile Strain @ Break	----	74
Young's Modulus	----	-10.1

Notes: All specimens were conditioned at 23°C and 50% relative humidity before chemical resistance exposure or testing. The food substance used was a solution of 50% cottonseed oil and 50% oleic acid (by weight). All tensile testing was performed using a 0.2 inch/minute crosshead speed using a 2" extensiometer.

For a 7-day period, the "Tensile Stress @ Break" fell off by 80% for PS Crystal specimens exposed to 0.5% strain and a mixture of oils, i.e. 6.25 kpsi was reduced to 1.34 kpsi, while the "Tensile Stress @ Break" for the interpolymer of Example 6 remained relatively the same, i.e. 5.07 kpsi vs. 5.10 kpsi.

While the present invention has been particularly set forth in terms of specific embodiments thereof, it will be understood in view of the instant disclosure that numerous variations upon the invention are now enabled yet reside within the scope of the invention. Accordingly, the invention is to be broadly construed and limited only by the scope and spirit of the claims now appended hereto.

WHAT IS CLAIMED IS:

1. A process for forming interpolymers comprising:

a) suspending in an aqueous medium from about 20% to 60% by weight of polyolefin resin particles having a VICAT softening temperature greater than 85°C and a melt index of about 2.1 g/10 minutes (Condition I, 190°C, 2.16 kg) , thereby forming a first aqueous suspension;

b) adding continuously to the first aqueous suspension a first vinyl aromatic monomer composition comprising vinyl aromatic monomer and a polymerization initiator for polymerizing the vinyl aromatic monomer within the polyolefin resin particles, thereby forming a second aqueous suspension, maintaining said second aqueous suspension at a first temperature such that a portion of said first vinyl aromatic monomer composition polymerizes within said polyolefin resin particles;

c) adding continuously to the second aqueous suspension a second vinyl aromatic monomer composition comprising vinyl aromatic monomer, thereby forming a third aqueous suspension, said second vinyl aromatic monomer composition being substantially free of polymerization initiator, said second vinyl aromatic monomer composition being added to said second aqueous suspension after addition of said first vinyl aromatic monomer composition,

said first vinyl aromatic monomer composition and said second vinyl aromatic monomer composition together providing from 40 to 80 percent by weight of vinyl aromatic monomer, based on total weight of polyolefin resin particles and vinyl aromatic monomer,

said third aqueous suspension being maintained substantially at said first temperature such that a portion of said second vinyl aromatic monomer composition polymerizes within said polyolefin resin particles; and

d) holding said third aqueous suspension at a second temperature so as to substantially completely polymerize said first vinyl aromatic monomer composition and said second vinyl aromatic monomer composition within said polyolefin resin particles, said second temperature being greater than said first temperature, thereby forming said interpolymer resin particles,

wherein polymerization of said first vinyl aromatic monomer composition and said second vinyl aromatic monomer composition being performed in the substantial absence of cross-linking agent such that said interpolymer resin particles have a gel content ranging from about 0 to about 1.5% by weight, based on the weight of the interpolymer resin particles, further wherein said interpolymer resin particles have a VICAT softening temperature ranging from about 90°C to about 115°C and a melt index ranging from about 0.2 to about 35g/10 minutes (Condition G).

2. A process of claim 1 wherein said interpolymer resin particles have a VICAT softening temperature from about 90°C to about 105°C, and a gel content ranging from about 0 to about 0.8% by weight, based on the weight of the interpolymer resin particles.

3. A process of claim 1 the steps further comprising:

e) using said interpolymers resin particles in an extrusion process, and introducing a blowing agent into said interpolymers resin particles to form a foamed article.

4. A process of claim 1 wherein said weight of said polyolefin particles is about 30% by weight based on the weight of said interpolymers particles, and said weight of said vinyl aromatic monomer is about 70% by weight based on the weight of said interpolymers particles.
5. A process of claim 1 wherein said weight of said polyolefin resin particles is about 40% by weight based on the weight of said interpolymers particles, and said weight of said vinyl aromatic monomer is about 60% by weight based on the weight of interpolymers particles.
6. A process of claim 1 wherein said weight of said polyolefin resin particles is about 50% by weight based on the weight of said interpolymers particles, and said weight of said vinyl aromatic monomer is about 50% by weight based on the weight of said interpolymers particles.
7. A process of claim 1 wherein said vinyl aromatic monomer is selected from the group consisting of styrene, alpha-methylstyrene, ethylstyrene, chlorostyrene, bromostyrene, vinyltoluene, vinylbenzene, isopropylxylene, and mixtures thereof.
8. A process of claim 7 wherein said polyolefin resin of said polyolefin resin particles is selected

from the group consisting of polyethylene, polypropylene, thermoplastic olefins, and thermoplastic elastomers resins.

9. A process of claim 8 wherein said vinyl aromatic monomer is styrene and wherein said polyolefin resin of said polyolefin resin particles is selected from the group consisting of polyethylene and polypropylene resins.

10. A process of claim 8 wherein said polyethylene resin is selected from the group consisting of low-density polyethylene and ethylene vinyl acetate copolymer.

11. A process of claim 1 wherein said interpolymer resin particles are pellets and said each pellet weighs from about 8 milligrams to 20 milligrams.

12. Interpolymer resin particles comprised of about from 20% to about 60% by weight based on the weight of the particles of uncross-linked polyolefin resin particles having a VICAT softening temperature greater than 85°C and a melt index of about 2.1 g/10 minutes (Condition I, 190°C, 2.16 kg); and from about 40% to about 80% by weight based on the weight of said interpolymer particles of polystyrene particles that have been polymerized in said uncross-linked polyolefin resin particles to form an interpenetrating network of polyolefin resin particles and polystyrene particles, and said interpolymer resin particles having a gel content ranging from about 0 to about 1.5% by weight based on the weight of said interpolymer resin particles, a VICAT softening temperature ranging from about 90°C to about 115°C, and a melt index value ranging

from about 0.2 to about 35.0 g/10 minutes (Condition G), wherein said interpolymer resin particles are prepared by a process comprising,

- a) suspending in an aqueous medium from about 20% to 60% by weight of polyolefin resin particles having a VICAT softening temperature greater than 85°C and a melt index of about 2.1 g/10 minutes (Condition 1.190°C. 2.16 kg), thereby forming a first aqueous suspension;
- b) adding continuously to the first aqueous suspension a first vinyl aromatic monomer composition comprising vinyl aromatic monomer and a polymerization initiator for polymerizing the vinyl aromatic monomer within the polyolefin resin particles, thereby forming a second aqueous suspension, maintaining said second aqueous suspension at a first temperature such that a portion of said first vinyl aromatic monomer composition polymerizes within said polyolefin resin particles;
- c) adding continuously to the second aqueous suspension a second vinyl aromatic monomer composition comprising vinyl aromatic monomer, thereby forming a third aqueous suspension, said second vinyl aromatic monomer composition being substantially free of polymerization initiator, said second vinyl aromatic monomer composition being added to said second aqueous suspension after addition of said first vinyl aromatic monomer composition.

said first vinyl aromatic monomer composition and said second vinyl aromatic monomer composition together providing from 40 to 80 percent by weight of vinyl aromatic monomer, based on total weight of polyolefin resin particles and vinyl aromatic monomer.

said third aqueous suspension being maintained substantially at said first temperature such that a portion of said second vinyl aromatic monomer composition polymerizes within said polyolefin resin particles: and

d) holding said third aqueous suspension at a second temperature so as to substantially completely polymerize said first vinyl aromatic monomer composition and said second vinyl aromatic monomer composition within said polyolefin resin particles, said second temperature being greater than said first temperature, thereby forming said interpolymer resin particles, wherein polymerization of said first vinyl aromatic monomer composition and said second vinyl aromatic monomer composition are performed in the substantial absence of cross-linking agent such that said interpolymer resin particles have a gel content ranging from about 0 to about 1.5% by weight, based on the weight of the interpolymer resin particles.

13. Interpolymer resin particles of claim 12 wherein said interpolymer resin particles have a VICAT temperature ranging from about 90°C to about 105°C and a gel content ranging from about 0 to about 0.8% by weight based on the weight of said interpolymer resin particles.

14. Interpolymer resin particles of claim 12 wherein said weight of said polyolefin resin particles is about 30% by weight and said weight of said polystyrene is about 70% based on the weight of said interpolymer resin particles.

15. Interpolymer resin particles of claim 12 wherein said weight of said polyolefin resin particles is about 40% and said weight of said polystyrene is about 60%, based on the weight of said interpolymer resin particles.

16. Interpolymer resin particles of claim 12 wherein said weight of said polyolefin resin particles is about 50% and said weight of said polystyrene is about 50%, based on the weight of said interpolymer resin particles.

17. Interpolymer resin particles of claim 12 wherein said vinyl aromatic monomer is selected from the group consisting of styrene, alpha-methylstyrene, ethylstyrene, chlorostyrene, bromostyrene, vinyltoluene, vinylbenzene, isopropylxylene, and mixtures thereof.

18. Interpolymer resin particles of claim 17 wherein said polyolefin resin of said polyolefin resin particles is selected from the group consisting of polyethylene, polypropylene, thermoplastic olefins (TPO'S), and thermoplastic elastomers (TPE'S) resins.

19. Interpolymer resin particles of claim 18 wherein said vinyl aromatic monomer is styrene and wherein said polyolefin resin of said polyolefin resin particles is selected from the group consisting of polyethylene resin and polypropylene resin.
20. Interpolymer resin particles of claim 19 wherein said polyethylene resin is selected from the group consisting of low-density polyethylene and ethylene vinyl acetate.
21. Interpolymer resin particles of claim 12 wherein said resin particles are pellets and said each pellet weighs from about 8 milligrams to about 20 milligrams.
22. An extruded article made from the interpolymer resin particles of claim 12.
23. An article of claim 22 wherein said article is an extruded solid sheet.
24. An article of claim 22 wherein said article is an extruded solid film.
25. An article of claim 22 wherein said article is an extruded foamed sheet.
26. An injection molded article made from the interpolymer resin particles of claim 12.

27. A thermoformed article made from the interpolymer resin particles of claim 12.

28. A multi-layer structure comprising a first layer formed from the interpolymer resin particles of claim 12 and positioned adjacent to at least a second layer wherein said first layer adheres to said second layer to form said multi-layer structure.

29. A polymer composition comprising said interpolymer resin particles of claim 12 and at least a second polymer.

30. A polymer composition of claim 29 wherein said second polymer is selected from the group consisting of polyethylene and polystyrene.

31. A polymer composition of claim 30 wherein the amount of said second polymer ranges from about 0.1% to about 99.9% by weight and the amount of said interpolymer resin particles ranges from about 99.9% to about 0.1% by weight based on the weight of said polymer composition.

32. A process for producing articles with improved ESCR properties by using the interpolymer resin particles of claim 12.

33. The process of Claim 1 wherein said polymerization initiator comprises benzoyl peroxide, said first temperature is substantially 91°C, and said second temperature is substantially 140°C.

34. The process of Claim 1 further comprising holding said third aqueous suspension at said first temperature for a period

of time, after addition of said second vinyl aromatic monomer composition and prior to holding at said second temperature, so as to allow residual of said first vinyl aromatic monomer composition and residual of said second vinyl aromatic monomer composition to further penetrate into said polyolefin resin particles.