Part I

Introduction

Chapter 1

Overview of Polyolefin Composites

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1.1 INTRODUCTION

Polyolefins are synthetic polymers of olefinic monomers. They are the largest polymer family by volume of production and consumption. Several million metric tons of polyolefins are produced and consumed worldwide each year, and as such they are regarded as commodity polymers. Polyolefins have enjoyed great success due to many application opportunities, relatively low cost, and wide range of properties. Polyolefins are recyclable and significant improvement in properties is available via blending and composite technologies.

Polyolefins may be classified based on their monomeric unit and chain structures as ethylene-based polyolefins (contain mostly ethylene units), propylene-based polyolefins (contain mostly propylene units), higher polyolefins (contain mostly higher olefin units), and polyolefin elastomers (1). Ethylene-based polyolefins are normally produced either under low pressure conditions using transition metal catalysts resulting in predominantly linear chain structure or under high pressure conditions using oxygen or peroxide initiators resulting in predominantly branched chain structures of various densities and crystallinity levels.

Propylene-based polyolefins are normally produced with transition metal catalysts resulting in linear chain structures with stereospecific arrangement of the propylene units or special stereoblock structures from a single-site catalyst. Higher polyolefins are normally produced using transition metal catalysts resulting in linear and stereospecific chain structures. Polyolefin elastomers based mainly on a combination of ethylene and propylene may be produced using metal or single-site catalysts with or without the inclusion of dienes (for cross-linking) and are mostly amorphous

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with high molecular weights and heterogeneous in phase structures. One may conclude that a given polyolefin may be a homopolymer, copolymer, or terpolymer depending on the number of monomers used in making the polyolefin, crystalline or amorphous depending on their chain conformation, configuration, and processing conditions.

Today polyolefins and polyolefin-based materials are used in many applications. These applications include transportation (automotive, aerospace), packaging, medical, consumer products (toys, appliances, etc.), electronics, cable and wire coating, thermal and acoustic insulation, and building and construction. Polyolefins can be extruded as filaments (fibers), films (cast and blown), and pipes/profiles. They can be molded into parts of various shapes. They can be foamed with physical and chemical foaming/blowing or/and can be coated onto other materials.

1.2 OLEFINIC MONOMERS

The alkenes having one or more unsaturated double bonds in their structures are the monomers used to synthesize polyolefins. They have the general formula C_nH_{2n}, $n \geq 2$. Table 1.1 shows the first 10 members of the olefinic monomers with one double bond, which are often called α-olefins.

The monomers in Table 1.1 form a homologous series of hydrocarbon compounds. Thus, apart from having the same general formula, all compounds in the series have the same functional groups. Each member of the group differs from the next in the series by the CH_2 group equivalent to 14 relative molecular mass units.

All members of the series have similar chemical properties. The physical properties of the compounds in the series show a progressive change with increasing relative molecular mass. The first three members of the alkenes homologous series are gases at room temperature. Those containing between 5 and 15 carbon atoms are colorless liquids and the higher compounds are waxy solids at room temperature. These α-olefinic monomers may be obtained as products of the cracking of gas–oil and naphtha fractions of petroleum distillations. They can also be obtained from synthetic organic chemistry methods.

Table 1.1 Alkene Monomers Having One Double Bond Used in the Synthesis of Polyolefins.

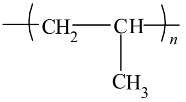
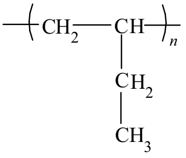
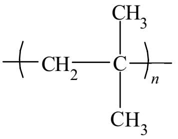
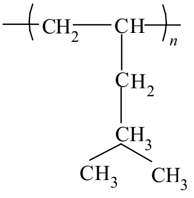
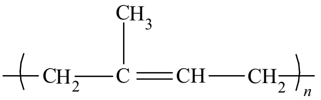
No. of carbon atoms (n)	Formula (C_n>H_{2n}, $n \geq 2$)	Name (other name)
2	C_2H_4	Ethene (ethylene)
3	C_3H_6	Propene (propylene)
4	C_4H_8	Butene-1 (butylene)
5	C_5H_{10}	Pentene-1
6	C_6H_{12}	Hexene-1
7	C_7H_{14}	Heptene-1
8	C_8H_{16}	Octene-1
9	C_9H_{18}	Nonene-1
10	$C_{10}H_{20}$	Decene-1

1.3 POLYOLEFIN HOMOPOLYMERS, COPOLYMERS, AND TERPOLYMERS

Polyolefin homopolymers, copolymers, and terpolymers are foundation materials for polyolefin blends. They may be obtained via radical or ionic chain growth polymerization of alkenes using conventional free radicals (e.g., from peroxides) and organometallic complexing (Ziegler–Natta and metallocenes) catalyst systems. Polyolefin polymerization technologies and novel catalyst systems have enabled the rapid development of polyolefins with a wide range of molecular chain structures, morphologies, properties, and particle size and shape.

Polyolefin homopolymers include polyethylene (PE), polypropylene (PP), polybutene-1 (PB), polymethylpentene-1 (PMP), and higher polyolefins. Table 1.2 shows the structures of commercial polyolefin homopolymers.

Table 1.2 Structures of Commercial Polyolefin Homopolymers.

Name (other name)	Chemical structure (repeat unit)
Polyethylene (polyethene, polymethylene)	$-\!\left(CH_2-CH_2\right)_n\!-$
Polypropylene (polypropene)	 $-\!\left(CH_2-\underset{\displaystyle CH_3}{\underset{\vert}{CH}}\right)_n\!-$
Polybutylene (polybutene-1)	 $-\!\left(CH_2-\underset{\displaystyle \underset{\displaystyle CH_3}{\underset{\vert}{CH_2}}}{\underset{\vert}{CH}}\right)_n\!-$
Polyisobutylene (polysobutene-1)	 $-\!\left(CH_2-\overset{\displaystyle CH_3}{\overset{\vert}{\underset{\displaystyle CH_3}{\underset{\vert}{C}}}}\right)_n\!-$
Polybutadiene	$-\!\left(CH-CH=CH_2-CH_2\right)_n\!-$
Poly-4-methylpentene-1	 $-\!\left(CH_2-\underset{\displaystyle \underset{\displaystyle \underset{\displaystyle CH_3\ \ CH_3}{\underset{\diagup\ \diagdown}{CH_3}}}{\underset{\vert}{CH_2}}}{\underset{\vert}{CH}}\right)_n\!-$
Polyisoprene	 $-\!\left(CH_2-\overset{\displaystyle CH_3}{\overset{\vert}{C}}=CH-CH_2\right)_n\!-$

Of these, PE and PP are the largest by amount produced yearly by the global polyolefin companies (1). PE comes in various forms differing in chain structures, crystallinity, and density levels. These are high density polyethylene (HDPE), low density polyethylene (LDPE), linear low density polyethylene (LLDPE), ultralow density polyethylene (ULDPE), and ultrahigh molecular weight polyethylene (UHMWPE). PP and higher polyolefins come in three stereo specific forms of varying densities: isotactic, syndiotactic, and atactic forms.

Polyolefin copolymers involve two olefinic monomers. The process of copolymerization is normally used to control the properties of the polyolefins. Some of the consequences of copolymerization are reduced crystallinity, melting point, modulus, strength, hardness, and low temperature impact. Polyolefin copolymers are either random or block copolymers of same or different monomers and may be a single phase or heterophasic depending on the amount of comonomer, the polymerization catalyst, and the process. For polyolefin copolymer of same monomers, this can be achieved by having different segments of the copolymer with different tacticities. One can have polyolefin block copolymers of same block or of varying block lengths. One can also have polyolefin copolymers consisting of both block and random segments together in the same macromolecule. Polyolefin copolymers are usually not homogeneous in composition but are actually mixtures of copolymers of varying compositions. It is also possible with polyolefins to have block copolymers with only one monomer. These are called stereoblock copolymers and can be achieved by having sections of the polyolefin copolymer possess different tacticities.

Polyolefin copolymers started with LLDPE and ethylene–propylene rubber (EPR). Today there are polyolefin copolymers of ethylene with butene-1, hexene-1, octene, cyclopentene, and norbornene and copolymers of propylene with butene-1, pentene-1, and octene-1 in addition to ethylene. There are copolymers of butene-1 with pentene-1, 3-methylbutene-1, 4-methylpentene-1, and octane in addition to its copolymers with ethylene and propylene. There are copolymers of 4-methyl-pentene-1 with pentene-1 and hexane-1 in addition to its copolymers with butene-1 and propylene. The function of the comonomers is to reduce crystallinity, as compared to the homopolymers, resulting in copolymers that are highly elastomeric with very low glass transition temperatures, high impact strength, low modulus, low density, and often optical transparency. The most widely used multiphase polyolefin copolymer is polypropylene impact copolymer. These copolymers are typically composed of isotactic polypropylene (iPP) and EPR. Impact polypropylene copolymers are produced by various processes, but they are generally characterized by the synthesis of iPP in the first reactor and EPR in the second reactor. Therefore, these systems are typically reactor blends. Postreactor blending can be done, but the starting material is most often the reactor blend polypropylene copolymer. Polyolefin copolymers are often used for film applications or as impact modifiers.

Polyolefin terpolymers contain three olefinic monomers in their structures. A well-known example is ethylene propylene diene monomer (EPDM). The diene (double bond) monomer is usually ethylidene norbornene or 1,4-hexadiene. EPDM was introduced because of the difficulty in cross-linking saturated polyolefin homopolymers and copolymers.

There are also functionalized polyolefins. These are usually copolymer or terpolymer containing functional groups like epoxide, anhydride, hydroxyl, acrylate, and carboxylic acid. These functional groups are either grafted onto the polyolefin after polymerization or added directly *in situ* during polymerization reactions involving olefins and functional groups bearing polar monomers such as vinyl acetate, methyl acrylate, butyl acrylate, glycidyl methacrylate, and acrylic acid. Functionalized polyolefins are useful compatibilizers and impact modifiers in blends and composites containing polyolefins and nonpolyolefins. In this sense, functionalized polyolefins may be considered as additives rather than matrix materials in the formulation of polyolefin composites.

Commercial polyolefins often contain additives such as colorants, flame retardants, antioxidants, light stabilizers, nucleating agents, antistatic agents, and lubricants (microcrystalline waxes, hydrocarbon waxes, stearic acid, and metal stearates). These additives aid the processing and fabrication of products from polyolefins. Detailed treatments about specific polyolefins, polymerization systems/mechanism/ processes, structures, properties, processing, and applications may be found in References (2–9).

1.4 POLYOLEFIN COMPOSITES

Polyolefin composites are a subset of polymer composites (10–14). They emerged as a result of the need to meet the increasing application demands not satisfied by synthesized neat polyolefins. Polyolefin composites may be defined as polyolefin-based materials containing at least one functional nonpolymeric additive of organic or inorganic origin. Polyolefin composites may be consolidated, void containing, with random or oriented additives. Additives of interest in the formulation of polyolefin composite may be of natural or synthetic origin, different sizes, continuous (long) or discontinuous (short) in length, fibrous, flaky, disc-like or spherical.

Additives in polyolefin composites may be classified according to their functions as modifiers (e.g., fillers, plasticizers, blowing agents, coupling agents, impact modifier, and nucleating/clarifying agents), property extenders (e.g., heat stabilizer, antioxidants, flame retardants, light stabilizers, antistatic agents, and biocides), and processing aids (e.g., lubricants, slip agents, and antiblocking agent). In terms of specific chemical names, additives used in polyolefin composites include, but not limited to, the following: glass fibers, hollow glass bubbles, clay minerals, carbon black, carbon nanotubes, carbon fibers, graphite, wollastonite, magnesium hydroxide, aluminum trihydroxide, attapulgite, titanium dioxide, hydroxyapatite, calcium carbonate, silica, and natural fibers.

In addition to the polyolefin and the additives, polyolefin composites may contain other thermoplastics or thermosetting polymers. The characteristics of polyolefin composites are determined by the properties of their components, compositions, structures, and interactions, as is case with any multicomponent material. Table 1.3 lists the typical polyolefin composites studied in the literature (15–396). These composites involve the following polyolefins: PP, PE (LLDPE, LDPE, HDPE, and UHMWPE), EPR, EPDM, and PB.

Polyolefin composites may be prepared by processes that involve mixing and/or melting the components of the composites in a batch or in continuous mixers (single and twin screw extruders), followed by fabrication (molding, thermoforming) into the desired shape. The mixing process may be physical or accompanied by chemical reactions in situations where chemical or reactive modifiers are used.

There are plenty of publications (journal articles and patents) on polyolefin composites in the literature. To illustrate the abundant publications in the field, Tables 1.4 shows the total number of journal articles and patents published each year during the 6-year period from 2000 to 2005.

The date range is arbitrarily chosen. The number of journal articles for each year was electronically searched and obtained from polymer and polymer-related journals using the keywords polyolefin and composite. Within the polyolefin keyword, the subkeywords used in the search are polyethylene (PE, LLDPE, LDPE, HDPE, UHMWPE, PE, etc.), polypropylene (PP, iPP, sPP, aPP, etc.), polybutene-1 (PB), poly-4-methyl pentene-1 (PMP), ethylene–diene monomer (EPDM), ethylene–propylene–diene terpolymer(EPDM), ethylene–propylene rubber (EPR), thermoplastic olefins (TPO), natural rubber, polybutadiene, polyisobutylene, polyisoprene, and polyolefin elastomer (POE). Within the keyword composite, subkeywords used are clay (kaolin, talc, mica, smectite, (bentonite (montmorillonite) and hectorite)), alumina, hydroxyapatite, carbon black, carbon nanotube, carbon fiber, natural fiber (flax, jute, silk, wool, cotton, linen, kenaf, cashmere, sisal, bamboo, hemp, coconut, etc.), wood flakes, flour, fiber, ammonium polyphosphate, aluminum trihydroxide (aluminum trihydrate), magnesium hydroxide, graphite, attapulgite, cellulosic flour, wollasonite, silica, titanium dioxide, mica, calcium carbonate, glass beads, glass fiber, glass bubbles, and so on.

Regarding the patent search, polymer indexing codes and manual codes were used to search for the patents in Derwent World Patent Index based on the above keywords. Table 1.4 shows a growing trend in the number of publications in the polyolefin composites. It should be noted that in the preparation of Table 1.4, it is possible that some publications were missed in the reference search period (2000–2005).

Polyolefin composites research involves many issues such as the following:

i. Effect of additives (e.g., coupling agent, fillers, and impact modifier).

ii. Batch and continuous mixing/compounding and molding.

iii. Morphological characterization using techniques such as scanning electron microscopy (SEM), transmission electron microscopy (TEM), atomic force microscopy (AFM), and polarized light microscopy (PLM).

Table 1.3 Polyolefin Composites Studied in the Literature.

Polyolefin composites		
PE/calcium carbonate (35, 200, 228, 235, 238, 242, 243, 247, 250, 258)	PP/PS/montmorillonite (177)	PP/PET/glass beads (55, 109, 346)
PE/wood flour (42, 48, 56, 101, 138, 192, 259, 395)	PP/polyamide 6/glass fiber (107, 249, 335, 382)	PP/lignocellulosic flour (145)
PE/PA6/montmorillonite (175)	PP/PA6/talc (110)	PP/PMMA/calcium carbonate (118)
PE/PA6/glass fiber (128)	PP/PA6/calcium carbonate (110)	PP/SBR/calcium carbonate (143)
PE/alumina (148)	PP/PA6/ammonium polyphosphate (110)	PP/EPDM/wollasonite (45)
PE/Ethylene-co-ethyl acrylate/carbon black (164)	PP/glass beads (42, 70, 203, 224, 312, 383)	PP/EPDM/clay (33, 351)
PE/silica (171)	PP/PA6/montmorillonite (68, 82, 147, 360)	PP/EPDM/glass beads (157)
PE/EVA/montmorillonite (26)	PP/sisal fiber (285, 318, 338, 379)	PP/EPDM/glass fiber (333)
PE/carbon nanotube (91)	PP/carbon black (16, 209, 276, 295, 339)	PP/EPDM/calcium carbonate (187)
PE/titanium dioxide (137)	PP/HDPE/wood flour (287)	PP/EPDM/antimoniumntrioxide (257)
PE/glass fiber (309, 310, 317)	PP/magnesium–aluminum silicate (60)	PP/EPDM/flax fiber (41, 172)
PE/LCP fiber/glass fiber (326)	PP/SBS/glass fiber (324, 325)	PP/flax fiber (321)
PE/hydroxyapatite (161)	PP/alumina (132)	PP/copper wire/glass fiber fabric (327)
PE/magnesium hydroxide (254, 278, 394)	PP/magnesium hydroxide (47, 86, 103, 134, 262)	PP/EPDM/talc (158)
PE/graphite (72, 286, 329, 350, 366)	PP/basalt fiber (112)	PP/EPDM/white rice husk (220, 241, 244)
PE/natural fiber(252)	PP/PET/montmorillonite (114)	Polybutadiene/montmorillonite (263)
PE/jute fiber (229)	PP/natural rubber/rice husk ash (245)	Polybutadiene/carbon black (377)
PE/glass fiber/sisal fiber (359)	PP/talc (51, 65, 160, 169, 170, 214, 362, 380, 389)	Poly(butene-1)/montmorillonite (74)
PE/sisal fiber (199, 305)	PP/aluminum trihydroxide (42, 43, 186)	Poly(4-methyl-1-pentene)/ montmorillonite (364)
PE/montmorillonite (18, 22, 30, 37, 46, 67, 73, 75, 85, 100, 119, 135, 136, 151, 154, 156, 173, 175, 183, 191, 197, 198, 225, 261, 266, 272, 274, 337, 341, 342, 365)	PP/glass fiber (102, 211, 221, 237, 281, 296, 298, 301, 304, 306, 328, 330, 332)	EPR/silica (106, 293)

Table 1.3 (*Continued*)

Polyolefin composites		
PE/wood fiber (267, 386)	PP/carbon nanotube (21, 34, 134, 144)	EPR/wollasonite (182)
PE/wood flake (292)	PP/EVA/calcium carbonate (57)	EPR/carbon fiber (96)
PE/PP/glass fiber (230)	PP/natural fiber/magnesium hydroxide (265)	EPDM/silica (375)
PE/red phosphorus (278)	PP/HDPE/EPR/calcium carbonate (284)	EPDM/white rice husk (375)
PE/PP/calcium carbonate (64, 284)	PP/PE/magnesium hydroxide (277)	EPDM/melamine fiber (275, 315)
PE/carbon black (23, 72, 79, 104, 126, 129, 279, 282, 289, 290, 349, 361)	PP/carbon fiber (202, 295, 322, 331)	EPDM/clay (52, 210, 255, 357, 373)
PE/calcite/zeolite (387)	PP/calcium carbonate (32, 80, 84, 120, 152, 184, 201, 204, 207, 246, 302, 311, 334, 336, 371)	EPDM/butyl rubber/carbon black (193)
PE/magnesium–aluminum double hydroxide (194)	PP/wood fiber (58, 14, 208, 222, 299–300)	EPDM/aluminum trihydroxide (291)
PP/talc/calcium carbonate (264)	PP/silica (77, 78, 117, 353, 363, 390)	EPDM/carbon black (49, 196, 268, 291)
PP/kenaf fiber (58)	PP/ethylene-co-octene/clay (99)	Natural rubber/oil palm wood flour
PP/cotton fibers/wood flakes (111)	PP/polyhedral oligomeric silsesquioxanes (89)	Natural rubber/aluminium powder (280)
PP/wood flour (59, 138, 162, 181, 248, 294)	PP/graphite (176)	Natural rubber/ montmorillonite (205, 358)
PP/kaolin (240)	PP/SBS/glass fiber (323, 324)	Natural rubber/silica (206, 212, 227, 233, 283, 354, 368)
PP/clay/calcium carbonate (142)	PP/montmorillonite (19, 15, 17, 25, 27, 29, 31, 38–40, 44, 54, 61, 63, 66, 69, 71, 75, 76, 81, 83, 87, 88, 92–95, 98, 108, 113, 115, 116, 121, 124, 130, 131, 133, 153, 155, 159, 165–167, 178, 179, 190, 194, 213, 226, 231, 232, 256, 260, 266, 269–271, 273, 297, 308, 340, 342, 343, 345, 347, 352, 355, 367, 385, 393, 395)	Natural rubber/bamboo fiber (216)

PP/natural fiber (234, 303)	PP/ethylene-co-octene/magnesium hydroxide (123)	Natural rubber/rubber wood fiber (384)
PP/rice husk powder (214, 353)	PP/EPDM/paper sludge (253)	Natural rubber/kenaf (236)
PP/silver (370)	PP/natural rubber/white husk powder (245)	Natural rubber/carbon black (215, 218, 223, 239, 283, 368, 374, 376, 388)
PP/HDPE/sisal fiber (379)	PP/EPR/talc (24)	Natural rubber/calcium carbonate (239)
PP/HDPE/EPR/sisal fiber (379)	PP/EPR/silica (90)	Natural rubber/lead (316)
PP/PC/attapulgite (150, 391)	PP/EPR/montmorillonite (50, 174)	Natural rubber/carbon fiber (374, 390)
PP/calcite/zeolite (387)	PP/epoxy/glass fiber/carbon black (127)	Natural rubber/lignocellulosic (251)

Table 1.4 Summary of Number of Electronic Articles on Polyolefin Composites Published between 2000 and 2005 in English Language-Based Polymer and Polymer-Related Journals in Comparison to the Number of Patents.

	Journal articles						
Year	2000	2001	2002	2003	2004	2005	Total
Number of articles	34	53	44	57	95	99	382
	Patents						
Year	2000	2001	2002	2003	2004	2005	Total
Number of patents	244	259	379	374	407	442	2105

iv. Structural characterization using radiation scattering and diffraction techniques such as X-ray scattering (XRS), X-ray diffraction (XRD), electron diffraction, and small-angle neutron scattering (SANS).

v. Structural characterization using spectroscopic techniques such as nuclear magnetic resonance (NMR) and Fourier transform infrared (FTIR).

vi. Rheological characterization.

vii. Thermal transitions and thermal stability.

viii. Isothermal and nonisothermal crystallization behavior under quiescent and nonquiescent conditions.

ix. Mechanical (static and dynamic) behavior under tensile, shear, compressive, or impact mode.

x. Plastic deformation.

xi. Thermomechanical behavior and thermal stability.

xii. Adhesion, interfacial, and interphase behavior.

xiii. Fire resistance/flammability behavior.

xiv. Barrier and transport properties.

xv. Surface/tribological properties.

xvi. Electrical/dielectric properties.

xvii. Ageing effects (time–temperature-dependent behavior).

xviii. Modeling and simulation via the use of phenomenological, atomistic, molecular dynamics, and Monte Carlo methods and comparisons with experimental results.

Of these issues, investigation of the effect of the use of coupling agents on interfacial adhesion between the hydrophobic polyolefin and the hydrophilic additive (particulate or fibrous) appears to be the most important. Poor polyolefin–additve interaction gives rise to poor properties, in particular mechanical property. The function of the coupling agent is to reduce the interfacial tension between a polymer and the nonviscoelastic particulate or fibrous additive and hence improve the desired composite properties. Table 1.5 shows a list of coupling agents used in the formulation of

Table 1.5 Polymeric and Nonpolymeric Coupling Agents Used in the Formulation of Polyolefin Composites.

Coupling agent	Polyolefin composite
PE-g-maleic anhydride	PE/montmorillonite (18, 30, 37, 46, 53, 67, 75, 100, 153, 173, 175, 183)
Ethylene-co-vinyl acetate	PE/montmorillonite (26)
Ethylene-co-glycidyl methacrylate	PE/montmorillonite (53)
Hydroxyl (OH) functionalized PE	PE/montmorillonite (53)
Ethylene-co-methacrylic acid	PE/montmorillonite (100)
PE-g-maleic anhydride	PE/PA6,6/montmorillonite (175)
PE-g-maleic anhydride	PE/wood flour (42)
Stearic anhydride	PE/wood flour (48)
PP-g-maleic anhydride	PE/wood flour (48)
Ethylene-co-methacrylic acid	PE/wood flour (101)
PE-g-acrylic acid	PE/wood flour (395)
PE-g-maleic anhydride	PE/wood fiber (267)
2-Hydroxy ethyl methacrylate	PE/jute fiber (229)
PE-g-maleic anhydride	PE/calcium carbonate
PP-g-maleic anhydride	PP/montmorillonite (19, 25, 27, 44, 62, 66, 76, 83, 92–94, 98, 116, 121, 125, 159, 165–167, 178, 179, 213, 269, 270, 308, 342, 395)
Styrene-co-maleic anhydride	PP/montmorillonite (83)
Hydroxyl (OH) functionalized PP	PP/montmorillonite (87)
Hydroxyl (OH) and amine (NH_2) Functionalized PP	PP/montmorillonite (115)
Hexamethylene modified PP-g-maleic anhydride	PP/montmorillonite (131)
PP-g-acrylic acid	PP/montmorillonite (159)
PP-g-maleic anhydride	PP/PA6/montmorillonite (147)
EPR-g-maleic anhydride	PP/PA6/montmorillonite (68, 147)
PP-g-maleic anhydride	PP/ethylene-co-octene/montmorillonite (99)
PP-g-maleic anhydride	PP/EPDM/montmorillonite (33, 63)
PP-g-maleic anhydride	PP/PET/montmorillonite (114)
PP-g-maleic anhydride	PP/EPDM/flax fiber (41)
PP-g-maleic anhydride	PP/PET/glass bead (55)
PP-g-maleic anhydride	PP/wood flour (59, 138, 162, 181, 248)
Styrene-co-ethylene-co-butadaiene-co-styrene-g-maleic anhydride	PP/wood flour (138)
PP-g-maleic anhydride	PP/wood flour (294)
PP-g-maleic anhydride	PP/wood fiber (222, 300)
PP-g-maleic anhydride	PP/natural fiber (234)
PP-g-maleic anhydride	PP/kenaf fiber (121, 323)
PP-g-monomethyl itaconate	PP/rice husk
PP-g-succinyl-fuoresceine	PP/talc (65)
PP-g-maleic anhydride	PP/silica (90)
Ethylene-co-butyl acrylate	PP/silica (90)
Ethylene-co-butyl acrylate-co-maleic anhydride	PP/silica (90)

Table 1.5 (*Continued*)

Coupling agent	Polyolefin composite
PP-g-monomethyl itaconate	PP/silica (353)
Styrene-co-ethylene-co-butadaiene-co-styrene	PP/glass fiber (306)
Styrene-co-ethylene-co-butadaiene-co-styrene-g-maleic anhydride	PP/glass fiber (306)
PP-g-maleic anhydride	PP/PA6/glass fiber (107)
PP-g-acrylic acid	PP/PA6/glass fiber (382)
4,4′-Diphenylmethane carbodiimide	PP/PA6/glass fiber (249)
4,4′-Diphenylmethane bismaleimide	PP/PA6/glass fiber (249)
2,2′-(1,4-Phenylene) bisoxazoline	PP/PA6/glass fiber (249)
PP-g-maleic anhydride	PP/PET/glass beads (109)
PP-g-maleic anhydride	PP/silicon dioxide (117)
PP-g-maleic anhydride	PP/calcium carbonate (204)
PP-g-acrylic acid	PP/calcium carbonate (334)
PP-g-maleic anhydride	PP/graphite (176)
PP-g-maleic anhydride	PP/graphite oxide (176)
PP-g-maleic anhydride	PP/EPDM/paper sludge (253)
PP-g-acrylic acid	PP/magnesium hydroxide (47)
1, 3-Phenylene dimaleimide	PP/magnesium hydroxide (103)
PP-g-maleic anhydride	PP/aluminum trihydroxide (185)
Hydroxyl (OH) functionalized PP	PP/aluminum trihydroxide (185)
EPDM-g-maleic anhydride	EPDM/montmorillonite (255)
PP-g-maleic anhydride	EPR/carbon fiber (96)
Ethylene diamine dilaurate	Natural rubber/silica (229)
Propylene-co-ethylene-g-acrylic acid	Natural rubber/LLDPE/white rice husk ash (219)
PP-g-maleic anhydride	Natural rubber/kenaf fiber (236)

polyolefin composites. These coupling agents are either polymer based (high molecular weight) or low molecular weight.

1.5 TRENDS IN POLYOLEFIN COMPOSITES

The first trend and perhaps the most important one is the addition of nanosized additives to form polyolefin nanocomposites. This trend has begun to gain wider acceptance and utility. Polyolefin nanocomposites are expected to be used in applications such as automotive, packaging, electronics, and electrical industries. The nanosized additives include layered clay minerals such as smectite, (bentonite (montmorillonite), vermiculite, kaolinite, hectorite), carbon nanotubes/nanofibers, and silica nanoparticles. The layered nanoclays and nanofibers/nanotubes have large aspect ratio, whereas other nanoparticles have large surface areas. In polyolefin nanocomposites, properties such as strength and stiffness, flame retardation/char formation, barrier, heat distortation temperature, electrical and thermal conductivities can be improved significantly at low (typically < 5 wt%) nanosized additive loadings, compared with composites of macro- and microsized additives. Methods to make polyolefin nanocomposites include *in situ* polymerization, melt blending, solution blending, and ultrasonic mixing. In the nanocomposites containing layered clay, the clay may be exfoliated, intercalated, or in tactoid form. It is preferred to have the clay in the exfoliated form to obtain the desired property improvement. The main application of polyolefin nanocomposites is in the automotive industry with rising demand in packaging, building and construction, appliances, electrical and electronics, tools, sporting equipment, and so on.

The second trend is the growing use of certain functional additives. These additives improve either processing or performance of polyolefin composites. For example, there is a growing importance of the use of flame retardants, particularly nonhalogenated flame retardant such as nitrogen/phosporus-based compounds (ammonium polyphosphates, melamine, melamine cyanurate, melamine phosphates, etc.), metal oxides (e.g., aluminum oxide and antimonum oxides), hydroxides (e.g., alumnum trihydroxide and magnesium hydroxide), and metal hydrates (e.g., zinc borate). Flame retardant polyolefin composites find applications in electrical and electronics, transportation (motor vehicles, aircraft, and rail), building and construction, plastic-based consumer goods, and wire and cables. There is a growing use of block copolymers containing amine, epoxy, anhydride, and acid functionality as coupling agents in polyolefin composites formulation, in contrast to maleic anhydride-grafted polyolefins, which comparatively are not very efficient. This leads to polyolefin composites with improved impact. Reinforced/filled polyolefin composites with improved impact are increasingly replacing low end engineering polymers such as ABS and polyamides. Other functional additives that are increasingly used in the development polyolefin composites are stabilizers that improve long-term heat and light stability, high molecular weight antioxidants that address yellow discoloration caused by conventional phenolic antioxidants, surface modifiers that render polyolefin surface hydrophilic, processing aids (silica, flouropolymers, hydrocarbon waxes, and metal stearates) that improve flow properties, dispersants for better distribution of the additives within the polyolefin matrix, antimicrobials/biocides to inhibit the growth of bacteria, fungi, molds, mildews, and hollow glass bubbles/foaming agents for density reductions.

The third trend is particularly important from the standpoint of the development of polyolefin composites that are environmentally sustainanble, recyclable, renewable, and reusable. Example is the increasing modification of polyolefins (notably PE, PP, and EPDM) with biodegradable additives such as flax, jute, silk, wool, cotton, linen, kenaf, cashmere, sisal, bamboo, hemp, coconut, ramie, bagasse, abaca, corn starch, tapioca, and sago to form the so-called ecocomposites or green composites. The most widely used "ecocomposites" is polyolefin/wood flour or fiber composite. The challenge in polyolefin/biodegradable additive composites is the improvement of adhesion between the additives and the polyolefin and the dispersion of additives within the polyolefin matrix. These two challenges are being addressed by the use of coupling agents and chemical pretreatment of the biodegradable additives. There are other advanatages of "ecocomposites" apart from their recyclability and potential biodegradability. They are usually cheap since the natural additives are obtained from abundant plants (often from waste). From a processing standpoint, they are less abrasive to processing equipment and pose less inhalation hazards than glass fibers. They usually have good thermal and acoustic insulating properties. Finally, they are of low density compared to mineral-filled polyolefin composites, and with a proper and careful choice of coupling agents and processing aids, they can provide an aesthetic appeal and good mechanical properties.

The fourth trend is the increasing use of novel processing methods, particularly for polyolefin nanocomposites processing. For example, there is a growing use of supercritical fluids (e.g., supercritical carbon dioxide and nitrogen gases) such as dispersants to aid distribution and exfoliation of nanoclays or to reduce composites density in general. There is the use of ultrasound to exfoliate nanoclays during the preparation of polyolefin/layered clay nanocomposites. There is the use of solid-state shear processing to delaminate layered clay.

In conclusion, interest in polyolefin composites is growing and will continue to grow as new polyolefins are made and as new applications are sought for these materials.

NOMENCLATURE

AFM	Atomic force microscopy
BR	Butyl rubber
DSC	Differential scanning calorimetry
ED	Electron diffraction
EPDM	Ethylene propylene diene monomer
EPR	Ethylene propylene rubber
EVA	Ethylene-co-vinyl acetate
EVOH	Ethylene-co-vinyl alcohol
FTIR	Fourier transform infrared
HDPE	High density polyethylene
LCP	Liquid crystal polymer
LDPE	Low density polyethylene
LLDPE	Linear low density polyethylene
MLDPE	Medium density polyethylene
NBR	Nitrile butadiene rubber
NMR	Nuclear magnetic resonance
NR	Natural rubber
PA	Polyamide
PA12	Polyamide 12
PA6	Polyamide 6
PA66	Polyamide 66
PB	Polybutene-1
PBT	Polybutylene terephthalate

PC	Polycarbonate
PCL	Polycaprolactone
PE	Polyethylene
PEN	Polyethylene naphthalate
PET	Polyethylene terephthalate
PIB	Polyisobutylene
PLM	Polarized light microscopy
PMMA	Polymethylmethacrylate
PMP	Polymethylpentene-1
PP	Polypropylene
iPP	Isotactic polypropylene
sPP	Syndiotactic polypropylene
aPP	Atactic polypropylene
POE	Polyolefin elastomer
PPE	Poly(2,6 dimethyl-1,4-phenylene ether)
PPO	Polyphenylene oxide
PPS	Polyphenylene sulfide
PS	Polystyrene
PTT	Polytrimethylene terephthalate
PVC	Polyvinyl chloride
SAN	Styrene-co-acrylonitrile
SB	Styrene-co-butadiene
SBR	Styrene butadiene rubber
SBS	Styrene-co-butadiene-co-styrene
SEM	Scanning electron microcopy
SEBS	Styrene-co-ethylene-co-butadiene-co-styrene
SEP	Styrene-co-ethylene-co-propylene
SEPs	Styrene-co-ethylene-co-propylene-co-styrene
TEM	Transmission electron microscopy
TPE	Thermoplastic elastomer
TPO	Thermoplastic olefin
TPU	Thermoplastic polyurethane
TPV	Thermoplastic vulcanizate
TREF	Temperature rising elution fractionation
UHMWPE	Ultrahigh molecular weight polyethylene
ULDPE	Ultralow density polyethylene
VLDPE	Very low density polyethylene
XRD	X-ray diffraction
XRS	X-ray scattering

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