

CASTELLANOS & CO LTDA
ABOGADOS
Calle 94 A No. 7 A-09
Bogotá, D.C. - COLOMBIA

SUPERINTENDENCIA DE INDUSTRIA Y COMERCIO



No. 06-126880- -00003-0000

Fecha: 2008-05-22 14:42:24 Dep. 2020 NUECREACIONE
Tra. 11 PATENTEIYII Eve: 379 FASENACIONALI
Act. 538 PETICION Folios: 2

Señor

Jefe de la División de Nuevas Creaciones

Superintendencia de Industria y Comercio

Ministerio de Comercio, Desarrollo, Industria y Turismo

E. S. D.

Re.: Expediente No. 06.126.880

Solicitud del examen de patentabilidad correspondiente a la Patente PCT
Entrada en Fase Nacional, para:

**"FIBRAS DE POLIMERO CARGADO, METODO PARA LA PRODUCCION DE
LAS MISMAS, USO DE LAS MISMAS Y COMPOSICION QUE CONTIENE
FIBRAS DE ESTE TIPO"**

Solicitante: SAINT-GOBAIN MATERIAUX DE CONSTRUCTION S.A.S. - SG

SOLICITUD EXAMEN DE PATENTABILIDAD

(CAPITULO II)

Yo, **MARGARITA CASTELLANOS ABONDANO**, mayor de edad, identificada y domiciliada como aparece al pié de mi firma, obrando en mi calidad de apoderada de la sociedad solicitante, atentamente me permito solicitar a Ud. que se realice el examen de patentabilidad de la invención arriba citada, para lo cual anexo el comprobante de pago No. 08- 40.865 de Mayo 21 de 2008, correspondiente a la tasa establecida para dicha solicitud.

En consecuencia, atentamente solicito a Ud. se anexe este memorial junto con el comprobante de pago al expediente correspondiente a esta solicitud de Patente PCT Entrada en Fase Nacional, y se continúe con su tramitación.

Señor Jefe,

Margarita Castellanos Abondano
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Bogotá,
MCA/gcb/P1437

22 MAYO 2008

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SUPERINTENDENCIA DE INDUSTRIA Y COMERCIO

NIT : 800176089-2

RECIBO OFICIAL DE CAJA : 08 - 40.865
FECHA : MAYO 21 DE 2008

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***** CONSIGNACION *****

DEPOSITANTE	TIPO PAGO	BANCO	CUENTA	No. PAGO	FECHA PGO	Vr. PAGO
ALVARO CASTELLANOS	CONSIGNACION	BANCO DE BOGOTA	062754387	328366	21/05/2008	14,538,100.00

***** CONCEPTO *****

CANT.	RENTISTICO	CONCEPTO	TOTAL CONCEPTO
1	50005-01-01	SOLICITUDES	
		114 PTC CAP-II EXAMEN DE PATENTIBILID	210,000.00
		TOTAL :	\$ 210,000.00

SON: DOSCIENTOS DIEZ MIL PESOS

RESPONSABLE :

RECIBO DE CAJA APLICADO AL EXPEDIENTE No.

DIVISIÓN DE NUEVAS CREACIONES

RADICACIÓN EXPEDIENTE No. 06-126880

EL EXTRACTO DE LA PRESENTE SOLICITUD DE **PATENTE DE INVENCION**

FUE PUBLICADO EN LA GACETA DE PROPIEDAD INDUSTRIAL No **582** DE

FECHA **30 DE NOVIEMBRE DE 2007** EL TÉRMINO HÁBIL SEÑALADO POR LA

LEY EXPIRA EL **27 DE FEBRERO DE 2008**

NOTA: Dentro del plazo de los seis (6) meses siguientes a partir de la fecha de publicación en la gaceta de Propiedad Industrial, deberá solicitar y cancelar el examen de patentabilidad so pena de que la solicitud caiga en abandono, de acuerdo con lo establecido en el Artículo 44 de la Decisión 486/2000.

SE PRESENTARON OPOSICIONES POR PARTE DE TERCEROS.

SI _____

NO X

FIBER REINFORCED CEMENT MOLDED BODY

Publication number: JP2180771 (A)

Also published as:

Publication date: 1990-07-13

 JP2604220 (B2)

Inventor(s): HORIGOME TSUGUO +

Applicant(s): TESAC KK +

Classification:

- International: **C04B38/00; C04B16/06; C04B38/00; C04B16/00;** (IPC1-7): C04B38/00

- European:

Application number: JP19880334637 19881228

Priority number(s): JP19880334637 19881228

Abstract of JP 2180771 (A)

PURPOSE: To efficiently obtain a molded body having superior strength characteristics even after curing in an autoclave under severe conditions by using specified PP film fibers as the reinforcing fibers of a fiber reinforced cement molded body to be cured in an autoclave after molding.

CONSTITUTION: Chopped filament type PP film fibers produced from a PP film contg. 15-30wt.% inorg. material and having a high degree of orientation rendered by stretching with a high stretching factor of ≥ 12 in one direction are used as the reinforcing fibers of a fiber reinforced cement molded body to be cured in an autoclave after molding by sheeting, extrusion or casting. The average grain size (μm) of the inorg. material is $\leq 2,000/(c \times s < 2 >)$ [c is the content (wt.%) of the inorg. material and s is the stretching factor].

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705
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POLYOLEFIN FIBER HAVING HIGH PRECIPITATION RATE IN WATER

Publication number: JP5044163 (A)

Publication date: 1993-02-23

Inventor(s): TAKAI YOSUKE; YAMASHITA KENJI; TAKEDA HIDEKAZU; USUI YOSHIHARU +

Applicant(s): DAIWABO CREATE KK +

Classification:

- International: D01F8/46; D01F8/06; D03D15/00; D06M13/02; D06M13/244; D06M13/282; D06M13/292; D06M101/00; D06M101/16; D06M101/18; D06M101/20; D01F8/48; D01F8/08; D03D15/00; D06M13/00; (IPC1-7): D01F8/46; D01F8/06; D03D15/00; D06M13/292; D06M101/20

- European:

Application number: JP19910216358 19910801

Priority number(s): JP19910216358 19910801

Abstract of JP 5044163 (A)

PURPOSE: To obtain the subject fiber having high precipitation rate in water and free from flotation phenomenon of fiber by attaching a specific alkylphosphate metal salt to a specific simple or conjugate fiber composed of a polyolefin component containing calcium carbonate fine powder.

CONSTITUTION: For example, a core-sheath conjugate fiber composed of a sheath component consisting of polypropylene or poly-4-methylpentene-1 containing fine powder obtained by the surface-treatment of calcium carbonate fine powder having an average diameter of <1 μm with a metallic soap and a core component consisting of polypropylene, etc., is subjected to spinning and drawing and the drawn fiber is impregnated with a surfactant liquid consisting of a 8-18C n-alkyl phosphate metal salt. The obtained fiber can easily be mixed and dispersed in fresh water and cement slurry, is resistant to the flotation phenomenon of the fiber, exerts effective reinforcing action and has excellent flexural strength, precipitation rate, etc. It is suitable as a fiber for cement reinforcement or rope for fishery.

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US 2002/0110686A1

(19) United States

(12) Patent Application Publication (30) Pub. No.: US 2002/0110686 A1
Dugan (43) Pub. Date: Aug. 15, 2002

(54) FIBERS INCLUDING A NANOCOMPOSITE MATERIAL

Related U.S. Application Data

(60) Provisional application No. 60/267,528, filed on Feb. 9, 2001.

Publication Classification

(51) Int. Cl.⁷ D02G 3/00
(52) U.S. Cl. 428/344; 428/373

(57) ABSTRACT

Fibers are disclosed which include a polymeric nanocomposite material. The nanocomposite material includes a polymeric resin matrix and a plurality of nanosized platelet particles dispersed therein.

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(21) Appl. No.: 10/968,740

(22) Filed: Feb. 5, 2002

US 2002/0110686 A1

Aug. 15, 2002

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FIBERS INCLUDING A NANOCOMPOSITE MATERIAL

CROSS REFERENCE TO RELATED APPLICATION

[0001] This application is related to commonly owned copending Provisional Application Serial No. 60/267,528, filed Feb. 9, 2001, incorporated herein by reference in its entirety, and claims the benefit of its earlier filing date under 35 U.S.C. 119(e).

FIELD OF THE INVENTION

[0002] The present invention is related to polymeric fibers, and more particularly to polymeric fibers formed of a nanocomposite material.

BACKGROUND OF THE INVENTION

[0003] Synthetic polymers have been widely used in the production of fibers, films, molded articles, and the like. Common thermoplastic polymers used in the production of these and other products include polyethylenes, polyesters, and polyamides, among others.

[0004] Typically one or more additives can be added to the polymer melt prior to extrusion to improve specific properties. For example, an anti-oxidant is typically added to polypropylene prior to extrusion to minimize degradation. As another example, fire retardant fillers can be added to control polymer flammability. Other fillers used in the production of polymeric articles include titanium oxide (TiO₂), kaolin, pigments, carbon black, and the like.

[0005] While the addition of these and other fillers can improve a particular property of the resultant product, there can be problems associated with the use of such fillers. For example, the presence of a filler may improve one property of the end product while concurrently diminishing or adversely impacting another property. As an example, certain fillers can improve the flame retardant property of a polymeric article. However, high levels of loading are typically required to impart the desired degree of flame resistance. This in turn can alter the mechanical properties of the product, such as mechanical robustness.

[0006] Still further, use of fillers in the production of polymeric articles can cause processing problems. For example, fillers present in an extruded polymer can pose filter and capillary plugging hazards. In addition, the surface properties of the end product can be compromised, creating a rough, abrasive surface, instead of a smooth surface.

SUMMARY OF THE INVENTION

[0007] The present invention provides fibers formed of a polymeric nanocomposite material. Nanocomposite materials generally include a polymer having nanosized inorganic platelet particles dispersed therein, e.g., platelet particles having a nanometer size range in at least one dimension thereof.

[0008] The size of the particles can vary depending upon the material used to make the same. Generally the platelet particles can be described as modified clay materials with very high aspect ratios (the ratio of the object's width to its thickness). Thus the particles may also be described as having a very minimal thickness relative to their length.

Generally such particles have an average diameter between about 10,000 Å and about 50 Å, and an aspect ratio (length/thickness) ranging from about 1000 to about 1. The particles typically have an average thickness of less than or about 20 Å, and preferably an average thickness of less than or about 10 Å. This is in contrast to conventional filler materials, such as kaolin, titanium dioxide, carbon and the like, which have much larger sizes and typically are roughly or substantially spherical in shape.

[0009] The resultant fibers can be nanocomposite fibers. Alternatively the fibers of the invention can be multicomponent fibers, which include at least two structured polymeric components. For multicomponent fibers of the invention, generally at least one polymer segment includes a nanocomposite material, while at least one other polymeric segment includes a different nanocomposite material and/or other fiber forming polymeric material, which may or may not include nanoparticles.

[0010] The inventors have found that the fibers of the invention can exhibit several advantageous properties, as compared to conventional fibers formed without a nanocomposite material and/or formed with conventional fillers. Further, these benefits can be achieved at relatively low loading levels of the nanosized platelet particles. For example, the resultant fibers can exhibit enhanced strength, e.g., tensile yield strength and flexural modulus, as compared with fibers which are free of the nanocomposite material, or which contain conventional powdered fillers such as unmodified clay, silica or carbon black. Indeed, in contrast to the present invention, the use of such conventional fillers in fiber production typically degrades fiber properties such as tensile strength, tensile modulus and the like. The fibers of the invention can also exhibit improved stiffness and heat resistance, and decreased moisture absorption, flammability, and permeability, as compared to fibers prepared without filler and/or with conventional fillers.

[0011] Still further, the present invention can provide processing advantages. The nanosized nature of the platelet particles can reduce processing problems associated with the extrusion and spinning of conventional filled polymers into fibers, such as filter and capillary plugging.

DETAILED DESCRIPTION OF THE INVENTION

[0012] The present invention will be described more fully hereinafter in connection with illustrative embodiments of the invention which are given so that the present disclosure will be thorough and complete and will fully convey the scope of the invention to those skilled in the art. However, it is to be understood that this invention may be embodied in many different forms and should not be construed as being limited to the specific embodiments described and illustrated herein. Although specific terms are used in the following description, these terms are merely for purposes of illustration and are not intended to define or limit the scope of the invention. The fibers of the invention include at least one polymeric segment or component which includes a nanocomposite material. Nanocomposite materials are known in the art and are commercially available, for example, under the trade designation KIP 299A, available from KIP Company.

[0013] Such materials may generally be defined as a polymer matrix having nanosized inorganic particles dis-

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posed therein, e.g., platelet particles having a nanometer size range in at least one dimension thereof. The size of the particles can vary depending upon the material used to make the same. Generally the platelet particles can be described as modified clay materials with very high aspect ratios (the ratio of the object's width to its thickness). Thus the particles may also be described as having a very minimal thickness relative to their length.

[0014] Although not intending to be limited to such dimensions, generally such particles have an average diameter between about 10,000 Å and about 50 Å, such that the aspect ratio length/thickness ranges from about 1000 to about 1. For the purposes of the present invention, the average diameter is defined as the diameter of a circle having an area equal to the surface area of one broad surface face of the platelet shaped particle. The average thickness of the particles can be less than or about 20 Å, and more preferably less than or about 10 Å.

[0015] The platelet particles include an exfoliated layered inorganic material. Useful exfoliated inorganic materials include those derived from swellable layered materials including natural or synthetic phyllosilicates clays. Illustrative of such materials are smectic clay minerals such as montmorillonite, muscovite, beidellite, bentonite, vermiculite, hectorite, saponite, sepiolite, magadiite, kornite, sverdrupite and the like, as well as vermiculite, halloysite, aluminosilicates, hydrotalcite and the like. These layered clays generally comprise particles containing a plurality of silicate platelets having a thickness of about 8-12 Å tightly bound together at interlayer spacings of 4 Å or less, and contain exchangeable cations such as Na^+ , Ca^{2+} , K^+ or Mg^{2+} present at the interlayer surface.

[0016] The platelet particles may be present in the polymer matrix in varying amounts, depending upon the desired end use and properties of the resultant fibers. The amount of platelet particles present in the nanocomposites in accordance with this invention can be sufficient, for example, to improve various desired properties of the resultant fibers, such as mechanical properties of flexural modulus and tensile strength. The nanocomposite material can also impart other advantageous properties to the fibers, such as improved cut resistance and flame retardance. The nanocomposite polymer matrix can include the platelet particles in amounts ranging from about 0.01 to about 20 percent by weight, based on the total weight of the nanocomposite material present in the fiber. One advantage of nanocomposite materials over traditional fiber polymers is that nanocomposites can achieve significant improvements in properties at loadings that are much lower than those typically required using conventional additives. Thus in one embodiment of the invention the particle loading of the nanocomposite material is at the lower end of this range, for example from about 0.1 to about 5 percent by weight.

[0017] As used herein, "platelet particles" are particles having two relatively flat opposite faces, the thickness of which platelets is the distance between the faces, which is relatively small compared to the size of the faces. The platelet particles dispersed in matrix polymers have the thicknesses of the individual layers or small multiples less than about 10, preferably less than about 5 and more preferably less than about 3 of the layers, and still more preferably 1 or 2 layers. Preferably, intercalation of every interlayer space is

complete so that all or substantially all individual layers delaminate one from the other to form separate platelet particles.

[0018] There are many methods to produce specific polymeric materials containing nanocomposites, which have been previously disclosed and are known by those skilled in the pertinent art. These methods to produce nanocomposites can be grouped into three general categories: (1) in situ polymerization; (2) solution intercalation; and (3) melt exfoliation. In the first two methods, such materials can be prepared by processes in which swelling agents, such as long-chain organic cations, and water-soluble oligomers or polymers, are intercalated or absorbed between adjacent layers of a layered silicate, such as smectite clay, to thereby increase the interlayer spacing between the adjacent silicate layers, so that polymer chains can be included between the silicate layers when mixing the layered silicate with a polymer melt. See for example, U.S. Pat. No. 5,552,469, WO 93/04117, Japanese Laid Open Patent Application Nos. 8-151,449, 7-228,134, 7-228,762, 7-331,092, 6-259,806, and 6-259,846.

[0019] An example of in situ polymerization of a nanocomposite is disclosed in Kawasumi et al., U.S. Pat. No. 4,810,734. Generally this process includes the steps of dispersing a predetermined amount of a clay mineral into a dispersion medium and then adding a swelling agent to this mixture. The mixture is also mixed with a monomer of a polymer. An example of solution intercalation of a nanocomposite is disclosed in Deguchi et al., U.S. Pat. No. 5,102,948. This process is similar to in situ polymerization, except the mixture is mixed with a polymer resin instead of a monomer of a polymer.

[0020] Melt exfoliation or melt intercalation is described, for example, in Polymer Layered Silicate Nanocomposites, Giannelis, Adv. Mater. 1996, 8, No. 1, 29. Unlike in situ polymerization or solution intercalation, melt exfoliation only involves the clay mineral and the polymer without the need for a swelling agent or a dispersion medium. However, a swelling agent may be used to enhance the exfoliation of the clay mineral by particular polymers. In melt exfoliation, the clay mineral and the polymer are mixed together, then heated to a temperature above the softening point of the polymer.

[0021] In the nanocomposite material, two types of structures are possible. In one the structure is intercalated, in which a single extended polymer chain is inserted between layers resulting in a well ordered multilayer with alternating polymer/inorganic layers. In another, referred to as disordered or delaminated, the inorganic layers are substantially uniformly dispersed in the polymer with random orientation throughout the polymer matrix.

[0022] The polymer resin forming the nanocomposite matrix can be any of the types of polymer resins known in the art capable of being formed into a fiber construction. Examples of suitable polymers useful in the practice of the present invention include without limitation polyolefins, including polypropylene, polyethylene, polybutene, and polymethyl pentene (PMP), polyamides, including nylon 6, polyester, including polyethylene terephthalate, polyethylene naphthalene, polyvinylidene fluoride, poly(1,4-cyclohexylene dimethylene terephthalate) (PCT), and aliphatic polyesters such as polylactic acid (PLA),

polyphenylene sulfide, thermoplastic elastomers, polycarbonate, acetals, fluoropolymers, *ortho*- and *para*-polymers thereof and mixtures thereof. As noted above, the fibers of the invention can also include other conventional polymers, such as those listed above, but without the exfoliated platelet particles.

[0023] The fibers can optionally include other components not adversely affecting the desired properties thereof. Examples include, without limitation, antioxidants, stabilizers, particulates, pigments, and the like. These and other additives can be used in conventional amounts.

[0024] The fibers of the invention can be monocomponent fibers. In this regard, the fibers can be formed of 100% nanocomposite material. Alternatively, the fibers can be formed of a conventional polymer blended or randomly mixed with the nanocomposite material to form multicomponent fibers. In this regard, multicomponent fibers can be formed with intimate mixing, but can also be formed without intimate mixing.

[0025] Alternatively, the fibers of the invention can be multicomponent fibers, which include at least two structured polymeric components. In general, multicomponent fibers are formed of two or more polymeric materials which have been extruded together to provide continuous contiguous polymer segments which extend down the length of the fiber. For the multicomponent fibers of the invention, generally at least one polymer segment includes a nanocomposite material, while at least one other polymer segment or component includes a different nanocomposite material and/or other fiber forming polymeric material, which may or may not contain nanoparticles.

[0026] In this embodiment of the invention, the other fiber forming material can be the same or different from the polymer of the nanocomposite material. For example, the multicomponent fibers of the present invention can include at least one component formed of a nanocomposite material of a first polymer, and at least one other component formed of a different polymer and without filler. This can provide the advantage of imparting the respective properties of each of the polymers to the composite fiber. Alternatively, the fibers can include at least one component formed of a nanocomposite material of a first polymer, and at least one other component formed of the same polymer but without filler. As an example, the fibers could include at least one unfilled nylon component and at least one nanocomposite nylon component, such as a fiber having a nanocomposite nylon core and an unfilled nylon sheath. Nanocomposites can have higher heat deflection temperatures than similar unfilled polymers, so this combination can provide a bicomponent binder fiber that can be entirely recycled as a single polymer.

[0027] The term "fiber" as used herein means both fibers of finite length, such as conventional staple fiber, as well as substantially continuous structures, such as continuous filaments, unless otherwise indicated. The fibers of the invention can be hollow or non-hollow fibers, and further can have a substantially round or circular cross section or non-circular cross sections (for example, oval, rectangular, multi-lobed, and the like).

[0028] The multicomponent fibers can have a variety of fiber configurations as known in the art. The fiber components can be arranged so as to form distinct macroscopic

cross-sectional segments along the length of the fiber. For example, multicomponent fibers of the invention include those having cross-sectional configurations such as a pie/wedge, side-by-side, segmented round, segmented oval, segmented rectangular, segmented multi-lobed, and the like. However, other configurations are also included within the scope of the present invention, including those in which at least a portion of a one fiber segment is partially or fully occluded by an adjacent segment. Examples of other fiber shapes include, without limitation, islands in the sea, sheaf/core, and the like. The multicomponent fibers of the invention can be splittable, i.e., capable of separating into microfilaments upon appropriate chemical and/or mechanical action. Alternatively the multicomponent fibers of the invention can be substantially nondissectable.

[0029] The nanocomposite material (and other polymeric resin, if present, for example in a multicomponent fiber) can be either melt-spun into fibers, which may be formed into a web for instance by casting, airspinning, or wetlaying, or melt-spun directly into fibrous webs by a spunbonding or meltblowing process. The web can then be bonded to form a nonwoven fabric. Webs of the fibers of the invention can be made according to any of the known commercial processes for making nonwoven fabrics, including processes that use mechanical, electrical, pneumatic, or hydrodynamic means for assembling fibers into a web, for example casting, wetlaying, casting/hydroentanglement, wetlaying/hydroentanglement, and spunbonding.

[0030] The webs can be bonded using techniques as known in the art, such as but not limited to mechanical bonding, such as hydroentanglement and needle punching, adhesive bonding, thermal bonding, and the like, to form a coherent fabric structure. An example of thermal bonding is through air bonding, although other thermal bonding techniques, such as calendaring, microwave or other RF treatments, can be used.

[0031] The fibers of the invention can also be used to make other textile structures such as but not limited to woven and knit fabrics. These prepared for use in forming such woven and knit fabrics are similarly included within the scope of the present invention. Such yarns may be prepared from the continuous filament or spun yarns comprising staple fibers of the present invention by methods known in the art, such as twisting or air entanglement.

[0032] Many modifications and other embodiments of the invention will come to mind to one skilled in the art to which this invention pertains having the benefit of the teachings presented in the foregoing descriptions and the associated drawings. Therefore, it is to be understood that the invention is not to be limited to the specific embodiments disclosed and that modifications and other embodiments are intended to be included within the scope of the appended claims. Although specific terms are employed herein, they are used in a generic and descriptive sense only and not for purposes of limitation.

That which is claimed:

1. A fiber comprising a polymeric nanocomposite material.
2. The fiber of claim 1, wherein said polymeric nanocomposite material comprises a polymer having nanosized inorganic platelet particles dispersed therein.

3. The fiber of claim 2, wherein said fiber is a monocomponent fiber.

4. The fiber of claim 2, wherein said fiber comprises 100% by weight of said polymeric nanocomposite material.

5. The fiber of claim 2, wherein said fiber comprises a blend of said polymeric nanocomposite material and another polymer.

6. The fiber of claim 2, wherein said fiber is a multicomponent fiber comprising two or more polymeric segments, at least one of said segments comprising said polymeric nanocomposite material.

7. The fiber of claim 6, comprising at least one segment comprising a first nanocomposite material and at least one additional segment comprising a second nanocomposite material which is different from said first nanocomposite material.

8. The fiber of claim 6, comprising at least one segment comprising a first nanocomposite material and at least one additional segment comprising a polymer which is not a nanocomposite material.

9. The fiber of claim 6, wherein said at least one additional segment comprises a polymer which is different from the polymer of the nanocomposite material.

10. The fiber of claim 6, wherein said at least one additional segment comprises a polymer which is the same as the polymer of the nanocomposite material.

11. The fiber of claim 10, wherein said fiber comprises a nanocomposite nylon core and an unfilled nylon sheath.

12. The fiber of claim 2, wherein said platelet particles have an average diameter between about 10,000 Å and about 50 Å, and an aspect ratio length/width ranging from about 1000 to about 1.

13. The fiber of claim 12, wherein said platelet particles have an average thickness of less than or about 20 Å.

14. The fiber of claim 13, wherein said platelet particles have an average thickness of less than or about 10 Å.

15. The fiber of claim 2, wherein said platelet particles comprise an exfoliated layered inorganic material.

16. The fiber of claim 15, wherein said exfoliated layered inorganic material is a phyllosilicate clay.

17. The fiber of claim 16, wherein said phyllosilicate is a smectic clay material.

18. The fiber of claim 17, wherein said smectic clay material is selected from the group consisting of montmorillonite, nanomonte, bentonite, hexamonte, vermiculite, lepidine, hectorite, saponite, sepiolite, magadite, harysine, stevensite, and mixtures thereof.

19. The fiber of claim 2, wherein the polymer of said nanocomposite material is a thermoplastic polymer.

20. The fiber of claim 19, wherein said thermoplastic polymer is selected from the group consisting of polyolefins, polyesters, polyamides, and mixtures thereof.

21. The fiber of claim 20, wherein said polymer is a polyamide.

22. The fiber of claim 2, wherein said platelet particles are present in an amount ranging from about 0.01 to about 20 percent by weight, based on the total weight of nanocomposite material present in the fiber.

23. The fiber of claim 22, wherein said platelet particles are present in an amount ranging from about 0.1 to about 5 percent by weight.

24. The fiber of claim 6, wherein said fiber is selected from the group consisting of pile/ridge fibers, side-by-side fibers, segmented round fibers, segmented oval fibers, segmented rectangular fibers, segmented multilobal fibers, strand in the sea fibers, and sheath/core fibers.

25. The fiber of claim 1, wherein said fiber is selected from the group consisting of continuous filaments, staple fibers, and melt-spun fibers.

26. A fabric comprising a plurality of fibers comprising a polymeric nanocomposite material.

27. The fabric of claim 26, wherein said fabric is selected from the group consisting of nonwoven fabrics, woven fabrics, and knit fabrics.

28. A yarn comprising a fiber formed of a polymeric nanocomposite material.

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Studies on Composite Filaments from Nanoclay Reinforced Polypropylene

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(Received May 3, 2003; Revised December 30, 2003; Accepted January 6, 2004)

Abstract: The development of high tenacity, high modulus monofilaments from Polypropylene/Clay nanocomposites has been investigated. Pure sodium montmorillonite nanoclay was modified using benzoyl trimethyl ammonium bromide (HTAB) via an ion exchange reaction. Pure and modified clay were characterized through X-ray diffraction, FTIR and TGA. The modified clay was well blended with polypropylene (PP) in presence of a swelling agent. Composite filaments from PP/Clay nanocomposites were prepared at different weight percentages of nanoclay and the spinning and drawing conditions were optimized. The filaments were characterized for their mechanical, morphological and thermal properties. The composite PP filaments with modified clay showed improved tensile strength, modulus and reduced elongation at break. The composite filaments with unmodified clay did not show any improvement in tensile strength but the modulus improved. The sharp and narrow X-ray diffraction peaks of PP/clay composite filaments indicate increase in crystallinity in presence of modified clay at small loadings (0.5 %). The improved thermal stability was observed in filaments with modified as well as unmodified clays.

Keywords: Polypropylene, Modified sodium montmorillonite nanoclay, Composite filaments, Nanocomposites

Introduction

In recent years, there has been a lot of interest in research on hybrid organic-inorganic nanocomposites [1,2]. These composites exhibit superior strength, modulus, thermal resistance and heat distortion temperature. Layered silicates (nanoclays) are one of the important nanofiller materials. The basic approach of the polymer layered silicate nanocomposites is to separate and disperse the clay platelets in polymer matrix, with dispersions ranging from simple intercalation to complete exfoliation of the silicate layers.

Polypropylene/Clay nanocomposites have been prepared by various ways. Functionalized polypropylene has been used to make PP more compatible with montmorillonite (MMT) [3-5]. Use of swelling agents (like ethylene glycol, heptane, naphtha) facilitates the intercalation of polymer chains into the lamellae [6]. In situ polymerization of propylene in presence of activated sodium montmorillonite with Ziegler-Natta catalyst leads to the formation of fully exfoliated nanocomposites [7]. Modification of MMT with compatibilizers like benzoyl trimethyl ammonium bromide (HTAB) and octadecyl ammonium bromide to make it compatible with PP has also been reported [8]. PP/Clay nanocomposites generally show improvement in tensile strength, tensile modulus and storage modulus. They also exhibit improved thermal stability as compared to neat PP [7,9].

PP has found extensive application as a technical textile material in various engineering and high technology applications such as machine tapes, spacers, sport goods, composites etc. [10]. High tenacity, high modulus polypropylene monofilaments are generally prepared by drawing polypropylene filaments at very high draw ratios, high temperatures and at very slow

strain rates [11,12]. Practically there is no literature on the preparation of monofilaments from PP/MMT nanocomposites. In this study, we attempt to develop high tenacity, high modulus polypropylene filaments from PP/MMT nanocomposites.

Experimental

Materials

The isotactic PP used was Korylene ADL (AM L20 N), a homopolymer with an MFI of 12, obtained from IPCL (India). Purified sodium montmorillonite was supplied by Kummine Industries Co. Ltd., Japan. HTAB (Mack, AR grade) was used to modify MMT and ethylene glycol (Mack, AR grade) was used as the swelling agent. Modification of the MMT to make it more organophilic was carried out using ion exchange reaction with HTAB as per method used by earlier workers [7].

Preparation of Composite Filaments

Preparation of Master Batch

Master batches containing 10 wt% modified MMT and 10 wt% unmodified MMT in PP were prepared using Laboratory Mixing Extruder (manufactured by SDI, UK, Model No. S255). Before mixing, the MMT was swollen in ethylene glycol to help it disperse better in PP. The temperature of the mixer and die zone was kept at 180 and 210 °C respectively. The rotor speed was 45 rpm. The extruded strands were then cut using a granulator into chips.

Extrusion of Filament

The dispersion of nanoclay in PP matrix for preparation of PP/clay nanocomposite monofilaments was done by melt mixing. The mixing and extrusion of filaments was carried

Table 1. Sample composition and coding

Sample code	Composition
PP	Neat Polypropylene
M5	PP + 0.5 % Modified Clay
M10	PP + 1.0 % Modified Clay
M15	PP + 1.5 % Modified Clay
U5	PP + 0.5 % Unmodified Clay
U10	PP + 1.0 % Unmodified Clay
UC	Unmodified Clay
MC	Modified Clay

out in a laboratory model Single Screw Extruder, Barel, U.K. The melt spinning parameters i.e. screw speed, temperature at different zones in the extruder and take up speed were optimized to get a uniform filament. The temperature of I, II, III and die zones were kept at 170, 180, 200, and 225 °C respectively. The screw speed was 5 rpm and the as spun PP filaments were quenched in a water bath maintained at a temperature of 12-14 °C and collected at a take-up speed of 4 m/min. PP filaments with modified clay loading of 0.5, 1.0, 1.5, and 2.0 wt% and unmodified clay loading of 0.5 and 1.0 wt% were prepared. The filaments containing 1.5 wt% and more of unmodified clay could not be produced as the spinline was found to be unstable on account of frequent breakages of the as spun filaments. The composition of different sample batches is given in Table 1.

Drawing of Filament

Each as spun filament sample was drawn using laboratory model two zone drawing machine (fabricated) and the optimum conditions for drawing of the composite filaments were investigated. The filaments were drawn in three stages. The drawing temperatures were kept at 80, 110, and 160 °C and the draw ratios of 6.0, 1.2, and 1.8 were used in I, II, and III stages respectively. The total draw ratio was ~13.

Characterization of Unmodified and Modified MMT

Thermogravimetric Analysis (TGA)

The TGA of unmodified, modified MMT and HTAB was performed on Perkin-Elmer TGA-7 Thermogravimetric Analyzer. The temperature range was kept between 25 and 750 °C with a heating rate of 10 °C/min in nitrogen atmosphere.

FTIR Analysis

The IR spectra of the samples were recorded in KBr pellet form on Perkin-Elmer 883 Infrared Spectrometer. Separate KBr pellets were prepared with 1 % by weight of the samples (unmodified clay, modified clay and HTAB). The frequency range varied from 400 to 4000 cm⁻¹ at a nominal resolution of 4 cm⁻¹. The number of scans for each spectrum was fixed at 36.

Small Angle X-ray Diffraction

The SAXS studies on modified and unmodified MMT were carried out on Bruker Axis X-ray diffractometer (D8 Advance) using CuK α radiation operated at 40 kV and 30 mA. The data were collected in 2 θ angle range 2-12 ° at a scanning rate of 0.5 °/min.

Composite Filament Structure and Morphological Characterization

The nanocomposite filaments were characterized for their morphological features using XRD, optical birefringence, optical microscopy and SEM.

Sonic Modulus

Sonic modulus of the filaments was measured on the dynamic modulus tester PPM-R, made by RDM Morgan Co. It is a measure of the overall molecular orientation. Sonic modulus is calculated using the formula

$$E = \rho v^2 C^2 \quad (1)$$

Where E = modulus in GPa, ρ = density in g/cm³ ($\rho = 0.92$ for PP), C = sonic velocity in km/sec.

Wide Angle X-ray Diffraction

WAXD studies of the samples were carried out on Philips X-ray diffractometer with Nickel-filtered CuK α as radiation source. The diffractometer was operated at 40 kV and 30 mA in reflection mode with angle 2 θ range 10-35 ° at a scanning rate of 2 °/min.

Crystalline Orientation Function

WAXD studies were used to determine the crystalline orientation of melt-spun filaments. The Herman's orientation function [13] is defined as:

$$f_x = \frac{3(\cos^2 \theta_{hkl}) - 1}{2} \quad (2)$$

where θ is angle between reference direction (filament axis) and the x-crystallographic axis ($x = a, b$ or c). Assuming rotational symmetry about the filament axis:

$$\langle \cos^2 \theta_{hkl} \rangle = \frac{\int_0^\pi I(\theta) \cos^2 \theta \sin \theta d\theta}{\int_0^\pi I(\theta) \sin \theta d\theta} \quad (3)$$

where $I(\theta)$ is the intensity of diffraction from the (hkl) planes which are normal to the x-crystallographic direction. The integrals in equation are evaluated immediately from the intensity distribution of (110) and (040) reflection planes.

For monoclinic PP,

$$f_x = \frac{3(\cos^2 \theta_{hkl}) - 1}{2} \quad (4)$$

$$\text{where } \cos^2 \theta_{hkl} = 1 - 1.09 \cos^2 \theta_{110} - 0.90 \cos^2 \theta_{040} \quad (5)$$

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$\cos^2 \theta_{110}$ and $\cos^2 \theta_{040}$ are obtained from azimuthal intensity distribution measurements of (110) and (040) reflection using the equation (3).

X-ray Crystallinity

The weight fraction crystallinity of the nanocomposite filaments was determined by superimposing the amorphous curve of PP [14] on the $I(\theta)$ vs 2θ diffraction scans obtained from the x-ray diffractometer and using the following formula after segregating the crystalline contribution:

$$X_c = \frac{\int_{\theta_1}^{\theta_2} I_c(\theta) d\theta}{\int_{\theta_1}^{\theta_2} I_t(\theta) d\theta} \quad (6)$$

Where X_c = Crystalline mass fraction, $\theta = 2\sin\theta/\lambda$, I_c = Crystalline Diffraction Intensity, I_t = Total Diffraction Intensity, $\lambda = 1.54 \text{ \AA}$, θ = Bragg's angle, α = maximum scan angle.

Optical Birefringence

The optical retardation of the filaments was measured on optical polarizing microscope manufactured by Vickers Instrument, England with Leitz Wetzlar tilting plate type quartz compensator to get the birefringence values. The diameter of the filaments was measured using Leica DM-LP Optical Microscope. An average of ten readings was taken for each sample. The optical birefringence of the filaments was calculated using formula:

$$\Delta n = \frac{6.18 \times \text{phase difference (nm)}}{1000 \times \text{diameter (um)}} \quad (7)$$

Scanning Electron Microscopy

The surface and cross sectional views were observed under scanning electron microscope, Cambridge Stereoscan S-60. The samples were fractured in liquid nitrogen for cross sectional view.

Mechanical Property Evaluation

The tensile strength, tensile modulus, elongation at break and work of rupture were measured on tensile testing equipment, Sautimat M6 from Teutchno, Germany. A gauge length of 100 mm was used and the strain rate was selected in such way so as to keep the time to break at approximately 20 ± 2 seconds. All tensile testing results reported are in terms of average of 10 readings.

Creep Studies

Creep, the time dependent property of the nanocomposite filaments was studied using a simple instrument, which consisted of a vertical wooden scale and different weights for application of load. The filaments were stressed at a stress level of 260 MPa and a gauge length of 24 cm was used. The elongation of the filaments was recorded for three hours with measurements being taken at an interval of every two minutes for the first half an hour and 30 minutes for the

later part. The creep compliance was calculated and plotted as a function of time.

Results and Discussion

Characterization of Unmodified and Modified MMT

The nanoclay in its unmodified form is not dispersible in PP due to its polar nature. Therefore the clay was modified with long chain alkyl quaternary ammonium salt - HTAB by an ion exchange reaction. Modification of the clay makes it organophilic so that it can easily disperse in the PP matrix. The unmodified and modified clays were characterized using following techniques:

FTIR Spectra

The FTIR spectra of modified clay, unmodified clay and HTAB are shown in the Figure 1 and the characteristic peaks for the same are summarized in Table 2. The peaks at 3628.13 and 1633.99 cm^{-1} represent O-H groups in the silicate structure [15]. These are present in both unmodified and modified clays. The C-H stretching bands at 2922.61 and 2850.25 cm^{-1} , the N-H bending vibration at 1490.01 cm^{-1} and the peak at 1470.01 cm^{-1} representing C-H₂ bending is seen only in the spectra of modified clay [16]. The presence of peaks due to C-H and N-H bonds which are characteristic

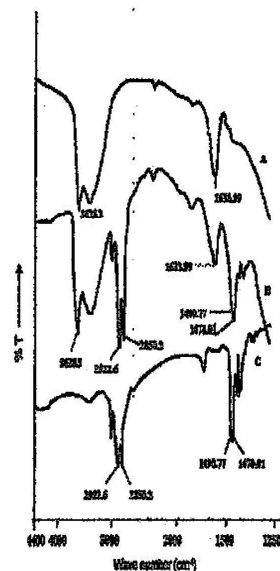


Figure 1. FTIR Spectra: A-Unmodified Clay, B-Modified Clay, C-HTAB.

Table 2. Summary of FTIR study of modified and unmodified clay

Wave number (cm ⁻¹)	Bond	Modified clay	Unmodified clay
3628.13	O-H	✓	✓
1633.99	O-H	✓	✓
2922.61	C-H Stretching	✓	×
2850.25	C-H Stretching	✓	×
1490.01	C-H ₂ Bending	✓	×
1470	N-H Bending	✓	×

✓, ✓ present and ✗, ✗ absent

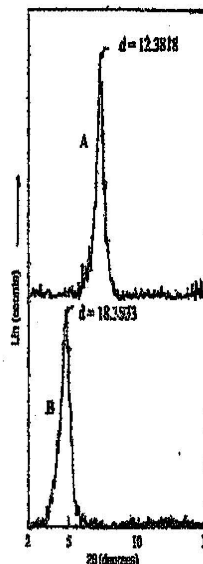


Figure 2. XRD diffractograms: A-Unmodified Clay, B-Modified Clay.

of the HTAB, in the FTIR of the modified clay confirms that the clay has been chemically modified by an ion exchange reaction with HTAB.

X-Ray Diffraction

The XRD of both unmodified and modified clay was carried out in powder form and the profiles are shown in Figure 2. The unmodified and modified clay show their characteristic diffraction peaks at 2θ angles 7.06° and 4.68° respectively. The shifting of diffraction maxima from 7.04° to 4.68° indicates that the d spacing of ordered clay domains has increased from 1.23 nm to 1.83 nm after modification.

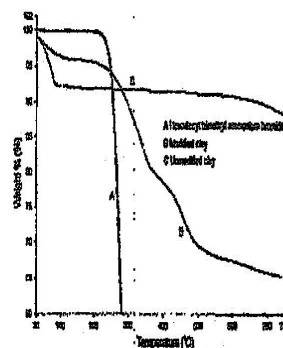


Figure 3. TGA Thermograms of HTAB, modified and unmodified clay.

The inter layer d spacing increases as the alkyl chains are tethered in the clay galleries. The increase in the basal distance ' d spacing' facilitates the intercalation of polymeric molecular chains into the inter layer galleries of the clay.

Thermogravimetric Analysis

The TGA thermograms of HTAB, unmodified MMT and modified MMT are shown in Figure 3. In the case of unmodified MMT, the initial weight loss of 6.9 % in the temperature range $30-100^\circ\text{C}$ occurs due to loss of water, which hydrates Na⁺ ions. Further weight loss between 630 and 744°C is reported to occur due to dehydroxylation of MMT [15,16]. The dehydroxylation of O-H present in the silicate structure occurs when it is heated beyond 500°C . In case of modified MMT, the weight loss takes place in 3 steps. A very small loss of about 3 wt% takes place between 28 and 100°C on account of loss of water. The water loss value decreases from ~ 7 wt% for pure MMT to 3 wt% for modified MMT indicating reduced hydrophilic nature of modified MMT. The second weight loss (~ 25 %) takes place between 230 and 500°C and is due to decomposition and oxidation of intercalated quaternary salt in two subsequent steps. The melt mixing of polypropylene and MMT was carried out at 225°C , so no significant decomposition of the intercalated quaternary salt is expected at this temperature.

Spinning

PP/Clay nanocomposite filaments were prepared as per details given in the experimental section. While with modified MMT the filaments could be spun successfully up to 2 % by weight, in case of unmodified MMT they could not be spun beyond 1.0 wt% as spin-line was found to be unstable leading to frequent breakages. Although the presence of clay in PP adversely affects its spinning performance as presence of any foreign material in a fibre forming polymer changes

NO

its rheological properties, it is clear that modification of MMT clay with HDAI helps in dispersing MMT better in PP matrix (as compared to unmodified MMT) and thus improves the spinnability of PP/clay nanocomposites.

Drawing

All the PP/clay nanocomposites and neat PP filaments were drawn to a draw ratio of 13 as per details given in the experimental section. The drawability of synthetic fibres depends on the initial morphology of an spun filaments, which influences its structure and properties. PP filaments under applied tensile stress mostly form α crystalline structure when quenched in air and smectic crystalline structure when quenched in ice-cold water [11]. When the spun filaments come out of the spinner, the crystallization takes place by two mechanisms. One is stress-induced crystallization and the other is heterogeneous crystallization. At higher take up speeds the stress-induced crystallization dominates while the lower take up speeds favor heterogeneous crystallization. In case of quiescent crystallization, neat PP forms α crystallites at low cooling rates and mesomorphic smectic crystallites at higher cooling rates in presence of montmorillonite [17]. The as spun composite filaments would be containing both α and smectic crystalline forms, the composition of which is determined by the actual cooling rate. PP with less stable pseudo crystalline smectic structure can be drawn to a higher degree than one with more stable α -crystalline form. Hence the extruded filaments were quenched in ice-cold water to facilitate the formation of smectic structure. The drawability of this smectic structure is maximum at 60 °C [18], hence the filaments were initially drawn at a temperature of 60 °C in the first drawing zone but showed multiple necking and whitening (indication of void formation due to overdrawing). The temperature and strain rate are important parameters to control these two drawbacks. The strain rate was kept constant and temperature was gradually increased. Stable neck was formed when the temperature was raised to 80 °C. At low speed of extrusion as used here, heterogeneous crystallization along with stress induced crystallization influences the morphology of the filaments. The heterogeneous crystallization due to the presence of clay in as spun composite filaments leads to increase in crystallinity as compared to the neat PP filaments, making it more difficult to draw composite filaments as compared to the neat PP filaments and hence the need for higher drawing temperature (~80 °C). After the first stage drawing, the filaments develop crystallinity and their drawability reduces, hence higher drawing temperature (110 °C) and lower draw ratio were used in the second stage drawing. The filaments were drawn at 160 °C in the third stage. The higher drawing temperature of 160 °C in the third zone initiates partial melting and recrystallization of imperfect and smaller crystals leading to perfection of crystals.

Characterization of PP/clay Composite Filaments

Morphological Characterization

The structural and morphological characterization of PP/clay nanoclay reinforced composite filaments was done using wide angle X-ray, DSC, sonic modulus and birefringence.

Wide Angle X-ray Diffraction

The X-ray diffractograms of PP, M5, and U5 are shown in Figure 4. The diffraction peaks at (110), (040) and (111) planes are sharper in composite filaments as compared to corresponding peaks in neat PP. The narrowing down of the peaks is associated with perfection of crystals. The X-ray crystallinity values of drawn PP/clay composite filaments are given in the Table 3.

It is observed that the crystallinity has significantly increased in composite filaments with lower weight percentage of modified MMT i.e., 0.5 wt% compared to neat PP whereas at higher weight percentages, the crystallinity decreases. The

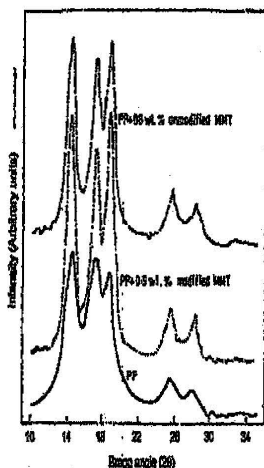


Figure 4. XRD Diffractograms of PP/clay nanocomposite filaments.

Table 3. X-Ray crystallinity of composite filaments

Sample code	XRD crystallinity (%)
PP	62
M5	69
M10	60
M15	50
U5	56
U10	58

Table 4. Crystalline orientation function, Birefringence and sonic modulus of composite filaments

Sample code	Crystalline orientation	Birefringence (C/P)	Sonic modulus (GPa)
PP	0.87	0.035	30.5
M5	0.89	0.028	25.5
U5	0.79	0.025	24.0

composite filaments from unmodified MMT at 0.5 and 1.0 wt% show lower values as compared to modified MMT composites. This may be due to modified MMT being better dispersed in PP matrix at lower concentration of 0.5 wt%, and hence not interfering in the crystallization process. The crystalline orientation function was calculated using equation (5) and the values are given in the Table 4. The crystalline orientation function, which depends on the spin line stress, is realized in case of nanocomposites having 0.5 wt% modified MMT (M5) and decreases slightly in case of nanocomposites with 0.5 wt% unmodified MMT (U5).

Birefringence

Birefringence measures the overall (amorphous and crystalline) molecular orientation of the polymer chains. The birefringence values for the various composite filaments are given in Table 4. The birefringence values for composite filaments with both modified and unmodified clays are lower than those for neat PP. This may be due to the presence of rigid clay particles in PP matrix, which restrict orientation of PP chains during drawing. This trend is also supported by the sonic modulus values for the composite filaments.

Sonic Modulus

The sonic modulus of all the filament samples was determined and is given in Table 4. Although the sonic modulus values of composite filaments are lower than those for neat PP filaments, there is not much difference in the sonic modulus values of different composite filaments. The presence of clay (modified and unmodified) in the polymer melt disturbs molecular orientation of the chains, which is also evident from the birefringence values of composite filaments, which are lower than those for neat PP filaments. The molecular orientation is disturbed more in case of unmodified clay, which is less homogeneously dispersed and forms micro size clay agglomerates dispersed in the matrix.

Mechanical Properties

The mechanical properties of the filaments such as tensile strength, tensile modulus and elongation are given in Table 5 and the stress-strain curves shown in Figure 5. It is observed that there is a significant improvement in the modulus and the breaking stress of the modified clay composite filaments even at very low weight fractions of clay (~0.5-1.0 wt%) as compared to neat PP filaments. The breaking stress of the

Table 5. Tensile properties of composite filaments

Sample code	Breaking stress (MPa)	Breaking elongation (%)	Modulus at 1% elongation (GPa)	Modulus at 5% elongation (GPa)
PP	532	19.7	7.6	5.13
M5	690	7.2	15.0	11.0
M10	642	6.6	15.5	10.8
M15	432	12.2	7.1	5.0
U5	525	7.6	11	7.9
U10	421	5.2	9.5	5.3

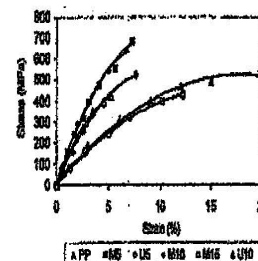


Figure 5. Stress-strain curves of PP/clay nanocomposite filaments.

composite filaments with modified clay at 0.5 and 1.0 wt% (M5, M10) increases by up to 30 % as compared to neat PP filaments. As the loading of modified clay increases in the PP matrix, the increase in the breaking stress starts decreasing and at 1.5 wt% of clay it is actually lower than neat PP. The enhancement in mechanical properties can be explained on the basis of modified clay platelets reinforcing the PP matrix at nanoscale due to nanodispersion resulting in combination of intercalated and exfoliated structure. The formation of intercalated/exfoliated structure is favored at higher temperatures where the rate of crystallization is lower and the chain mobility is high. Under such conditions polymer chains can diffuse into the interlayers of the clays [19]. The processing of composite filaments (which were extruded at fairly high temperature i.e., 225 °C and drawn at 160 °C in final stage) may have favored the process of intercalation and exfoliation of clays in the PP matrix. The specific surface area of dispersed clay increases with decrease in the particle size. This results in increase in the volume fraction of the interfacial regions and helps in better stress transfer. It is reported that increase in the volume fraction of the interfacial regions in case of nanocomposites leads to increase in the tensile strength [20]. The modulus of composite filaments also shows significant increase (almost doubles with 0.5 and 1.0 wt% of modified MMT), whereas the percentage elongation at break shows a sharp decrease. This trend is in line with increase in stiffness as well as brittleness in composite materials on addition of reinforcing fillers [21]. However, with unmodified

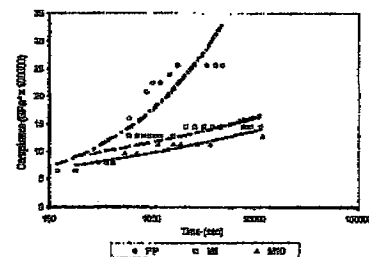


Figure 6. Creep compliance vs time plot of PP/clay nanocomposite filaments.

Table 6. Data on creep behavior of composite filaments at (stress 260 MPa)

Sample code	Extension % after 3 min	Extension % after 3 hrs
PP	1.67	8.3
M5	1.67	3.75
M10	1.67	3.33

clay the breaking strength and breaking elongation is reduced at similar clay loadings as compared to the neat PP filaments, indicating that unmodified clay was unable to disperse at nanolevel, forming micro-sized aggregates of clay particles dispersed in PP matrix and hence could not reinforce the polymer.

Creep Studies

The creep compliance of filaments is plotted with log time under loading stress of 260 MPa, which is equal to the 50 % of the breaking stress level of the neat and composite filaments. The variation of creep compliance with time is shown in Figure 6. It is observed that the creep compliance of PP filament increases with time at a much higher rate as compared to composite filaments indicating better resistance to creep for composite filaments. Table 6 shows the elongation percentage at different times of loading. While the extension just after loading (within 3 min) is same in all the cases, the extension after 3 hours is considerably lower in case of composite filaments as compared to neat PP filaments. The increased crystallinity and restricted mobility of the chains due to intercalation and reinforcement of the polypropylene chains by clay may be the reasons for improvement in the creep resistance of composite filaments.

Thermal Properties

The thermal resistance of the composite and neat PP

Table 7. Data from TGA analysis of filaments

Sample code	Onset temperature (°C)	Inflection peak (°C)
PP	338	446
M5	368	474
M10	370	478
US	374	387
U10	372	486

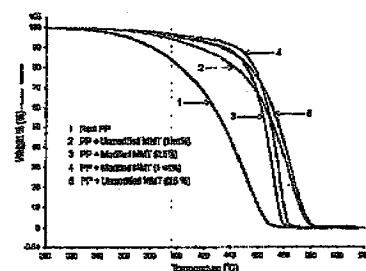


Figure 7. TGA thermograms of PP/clay nanocomposite filaments.

filaments was studied using Thermogravimetric analysis (TGA). The results are given in the Table 7 and thermograms in Figure 7. It is clear that the composite filaments with both modified and unmodified MMT have increased thermal resistance and the onset temperature of degradation increases by approximately 30 °C over neat PP at 0.5 and 1.0 wt% clay loadings. Since clay is itself a thermally resistant material, it imparts improved thermal stability to the composite filaments. In nanocomposites, high thermal resistance is also attributed to thin char formation, which does not fracture as the uniformly dispersed clay platelets reinforce the char structure [1,2].

SEM

The Scanning Electron microscopic (SEM) images of surface and cross-sections of composite filaments are shown in Figure 8. The cross sectional view of PP/modified MMT (M5) composite filament (Figure 8(a)) clearly shows that the clay is evenly dispersed in the PP matrix and the dispersed clay particles are in the range of 100-500 nm in diameter. Ideally, the particle size of the dispersed phase should be below 100 nm for a composite to be called a true nanocomposite. However, since the composite filaments with modified MMT show reinforcement, it can be postulated that intercalation of clay has taken place and smaller clay particles (<100 nm) are also present which may be too small to be resolved at the given magnification. The presence of few bigger size particles (500 nm)

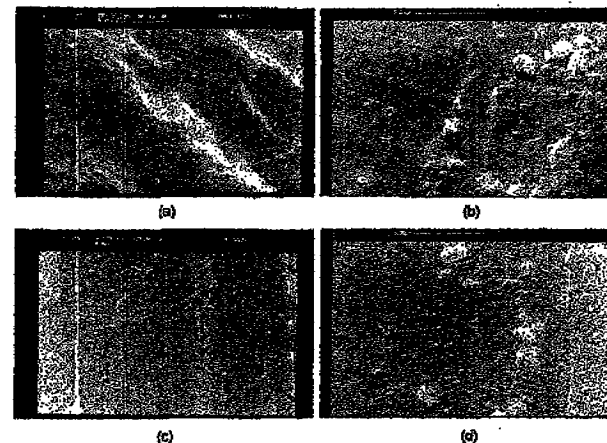


Figure 8. SEM micrographs (a) Cross sectional view of 0.5 wt% of modified clay/PP filament (M5), (b) Cross sectional view of 0.5 wt% of unmodified clay/PP filament (US), (c) Surface view of 0.5 wt% of modified clay/PP filament (M5), (d) Surface view of 0.5 wt% of unmodified clay/PP filament (US).

can be explained on the basis of lack of high shear during melt mixing in the single screw extruder. Typically, twin-screw extruders are used for providing high shear and intensive mixing conditions for proper dispersion of nanomaterials in polymeric matrices in melt intercalation techniques. The presence of clay leads to fibrillation in composite filaments as seen in the surface view micrographs (Figure 8(c) and 8(d)). From SEM image of cross section of PP/unmodified MMT (US) composite filament (Figure 8(b)), it is observed that unmodified clay particles are not uniformly dispersed in the PP matrix and tend to agglomerate as is evident from the micrographs. Also, the size of clay particles is much larger (1-2 microns) than those in PP/modified MMT composite filament. The polar nature of unmodified MMT is the primary reason for the incompatibility between hydrophilic unmodified clay and hydrophobic PP, which results in poor properties of PP/unmodified MMT composite filaments as compared to neat PP filaments.

Conclusion

Nanoclay reinforced PP nanocomposites could be spun and drawn successfully for 0.5, 1.0, and 1.5 wt% of the modified clay loading. Beyond 1.5 wt% the spinnability was

poor. The 0.5 and 1.0 wt% reinforced PP/modified nanoclay composite filaments show significant increase in tenacity (~30 %) as well as modulus (~100 %) as compared to neat PP filaments. The modified clay content in the range of 0.5-1.0 wt% gives optimum properties. The composite filaments also show improved creep resistance and thermal stability as compared to neat PP filaments. The XRD of the samples showed enhanced crystallinity in presence of modified clay. The SEM studies indicate good dispersion of modified clay in the PP. The modification of MMT using HTAB increases the interlayer d-spacing from 1.23 to 1.83 nm as is evident from the XRD data. This has facilitated the homogeneous dispersion of clay in PP matrix forming a combination of intercalated and exfoliated structure, responsible for enhanced mechanical and thermal properties. However, true molecular dispersion resulting in a completely intercalated/exfoliated composite structure is expected through the use of twin-screw extruder for intensive melt mixing and suitable compatibilizers, which is under investigation.

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**SUPERINTENDENCIA DE INDUSTRIA Y COMERCIO
DIRECCION DE NUEVAS CREACIONES**

2020

Abogada
MARGARITA CASTELLANOS ABONDANO
Apoderada

ASUNTO	Radicación	06-126880
	Trámite	011
	Evento	379
	Actuación	519
	Oficio	
	Folios	-018 -4355

Patente de Invención	☒	Patente de Modelo de Utilidad	☐
Vía Nacional	☐		
Vía PCT (fase nacional)	☒	Cap. I	☐
		Cap. II	☒

No. Sol. Internacional	<u>PCT/FR2005/0500364</u>
Fecha de Presentación Internal.	<u>25 de Mayo de 2005</u>

Como resultado del examen de fondo, realizado con fundamento en el artículo 45 de la Decisión 486 de la Comisión de la Comunidad Andina, se le comunica:

1. Documentos estudiados

- | | | |
|------|--------------------------|------------------------------------|
| 1.1. | <u>Descripción:</u> | Folios: 36 a 66 |
| 1.2. | <u>Reivindicaciones:</u> | Folios: 67 a 71 |
| 1.3. | <u>Prioridad:</u> | 25 de Mayo de 2004, FR No. 0451027 |

2. Objeto de la invención

- El Título de la solicitud es: **"FIBRAS DE POLIMERO CARGADO, METODO PARA LA PRODUCCION DE LAS MISMAS, USO DE LAS MISMAS Y COMPOSICION QUE CONTIENE FIBRAS DE ESTE TIPO"** (Folio 1).
- La presente invención se refiere al campo de las fibras, y se refiere más particularmente a fibras hechas de polímero cargado.

En la **Reivindicación Independiente 1** se plantea una fibra de polímero cargado que contiene aditivos dentro de ella, teniendo la fibra de polímero cargado un Módulo de Young superior al de una fibra de polímero sin cargar,

caracterizada porque los aditivos incluyen aditivos minerales que tienen por lo menos una dimensión submicrónica.

En la **Reivindicación Independiente 16** se plantea el uso de una fibra de polímero cargado de acuerdo con una cualquiera de las reivindicaciones precedentes, **caracterizado porque** se utiliza como una fibra de refuerzo en un producto.

En la **Reivindicación Independiente 17** se plantea un producto basado en fibras, **caracterizado porque** incluye fibras de polímero cargado de acuerdo con una cualquiera de las reivindicaciones 1 a 15.

En la **Reivindicación Independiente 23** se plantea un método para manufacturar una fibra de polímero cargado de acuerdo con cualquiera de las reivindicaciones 1 a 15 **caracterizado porque** incluye un paso de hilatura de una composición de polímero que contiene aditivos minerales que tienen por lo menos una dimensión submicrónica.

En la **Reivindicación Independiente 26** se plantea un método para manufacturar un producto sobre la base de fibras y de un material de fraguado hidráulico **caracterizado porque**

- Se prepara una mezcla inicial basada en ligante hidráulico, agua y fibras de polímero cargado definidas de acuerdo con cualquiera de las reivindicaciones 1 a 15;
- Se filtran las fibras sobre un soporte, móvil o estacionario, para formar una capa elemental húmeda; y
- Se superpone una pluralidad de capas elementales para formar un producto intermedio húmedo, y porque se somete el producto intermedio a un secado.

En la **Reivindicación Independiente 27** se plantea una composición para un material de fraguado hidráulico, **caracterizado porque** incluye un ligante hidráulico y fibras de polímero cargado de acuerdo con cualquiera de las reivindicaciones 1 a 15.

En la **Reivindicación Independiente 28** se plantea una composición **caracterizada porque** incluye una matriz de polímero y fibras de polímero cargado de acuerdo con cualquiera de las reivindicaciones 1 a 15.

- Se afirma que son conocidas las fibras de polipropileno cargado con nanotubos de carbono pero es difícil lograr una elevada pureza para los nanotubos pues los residuos de catalizador pueden formar impurezas con un tamaño en el orden del micrón que disminuyen las propiedades de las fibras terminadas. Además, es difícil producir nanotubos en grandes cantidades lo cual se refleja en el costo de las fibras. Estas fibras han sido preparadas bajo condiciones de laboratorio sin tomar en cuenta las restricciones industriales, especialmente en términos de fiabilidad y productividad. Por ello, el proceso de fabricación propuesto no es realista para la producción industrial.
- El objeto de la invención definido se catalogó como **D01F 6/06, D01F 8/06, E04C 2/06, E04C 2/22, E04C 5/07**, según la Clasificación Internacional de Patentes (IPC).

3. De la solicitud de Patente

3.1. Reivindicaciones (artículo 30 D 486)

- En la reivindicación independiente 1 se plantea una fibra de polímero cargado y solo se caracteriza por que "...los aditivos incluyen aditivos minerales que tienen por lo menos una dimensión submicrónica".

La forma de caracterizar la fibra es demasiado amplia pues "aditivos minerales" y "dimensión submicrónica" son expresiones indefinidas. Se recomienda definir claramente la naturaleza polimérica de las fibras, la naturaleza química de los minerales y cuantificar la dimensión submicrónica.

- En las reivindicaciones 18 a 22 se plantea "un producto basado en fibras" sin embargo, dicho preámbulo es indefinido pues no se comprende claramente cuál es el producto. Se recomienda modificar el preámbulo de modo que se plantee cuál es el producto que se espera reclamar. De manera similar ocurre con las reivindicaciones 27 y 28 donde se plantea "una composición".
- En la reivindicación independiente 23 se plantea un método para manufacturar una fibra de polímero incluyendo el paso de hilatura de la composición. Sin embargo, en la reivindicación dependiente 24 se menciona otra etapa diferente y en la reivindicación dependiente 25 otra más.

La manera como se está planteando el método no es apropiada ni clara ya que la reivindicación independiente 23 debería expresar de manera general pero completa, la secuencia de etapas del método pues del modo como lo plantea el señor solicitante no es comprensible dicha secuencia, es decir, el orden exacto de las etapas. Además, las reivindicaciones 24 y 25 que son dependientes no se están empleando para dar detalles específicos del invento revelado en la reivindicación independiente 23 como debería ser.

- En conclusión, las reivindicaciones 1, 18 a 25, 27 y 28 no cumplirían con el requisito de definición de la invención y claridad de acuerdo con el artículo 30 de la Decisión 486.

4. Unidad de invención (artículo 25 D 486)

- Analizar y argumentar la falta de unidad de invención haciendo énfasis en los diferentes conceptos inventivos que se presentan y las razones por las cuáles no existe una relación entre ellos.
- A continuación se listan las invenciones que contiene el capítulo reivindicatorio de la solicitud en estudio:

Reivindicaciones 1 a 15: La fibra de polímero cargado

Reivindicaciones 23 a 25: El método para manufacturar una fibra de polímero cargado

Reivindicación 26: Un método para manufacturar un producto sobre la base de fibras de polímero cargado y de un material de fraguado hidráulico

Reivindicación 27: Una composición para un material de fraguado hidráulico

Reivindicación 28: Una composición que incluye matriz de polímero

- Al analizar los grupos de invenciones anteriores se encuentra más de un concepto inventivo. Por ejemplo, la reivindicación 27 enuncia una composición para un material de fraguado hidráulico y la reivindicación 28 enuncia una composición polimérica; la reivindicación 26 enuncia un método para manufacturar un producto de fraguado hidráulico mientras que la reivindicación 28 enuncia una composición de matriz polimérica; la reivindicación 23 enuncia un método para manufacturar una fibra de polímero cargado mientras que la reivindicación 26 enuncia un método para manufacturar un producto sobre la base de fibras de polímero cargado y de un material de fraguado hidráulico.

Dado lo anterior, no hay unidad de invención entre las reivindicaciones 27 y 28; entre las reivindicaciones 26 y 28 ni entre las reivindicaciones 23 y 26. Por lo tanto, se recomienda realizar las modificaciones de claridad definidas en la presente comunicación y considerar la posibilidad de realizar divisionales de modo que cada solicitud contenga un grupo de reivindicaciones con un único concepto común inventivo.

- En conclusión, las reivindicaciones 27 y 28, 26 y 28, 23 y 26, no cumplirían con el requisito de unidad de invención como se debe según el artículo 25 de la Decisión 486.

5. Reivindicaciones relativas a un uso o a un segundo uso (artículos 14 y 21 D 486)

- En la reivindicación 16 se plantea el "uso de una fibra de polímero cargado de acuerdo con una cualquiera de las reivindicaciones precedentes caracterizado porque se utiliza como una fibra de refuerzo en un producto".

Mediante esta reivindicación el señor solicitante quiere proteger el uso de las fibras de polímero lo cual no se considera una invención y según la legislación colombiana no es posible aceptar ni tampoco puede otorgársele patente.

Se recomienda eliminar dicha reivindicación del capítulo reivindicatorio.

- En la reivindicación 17 se plantea un "producto basado en fibras caracterizado porque incluye fibras de polímero cargado de acuerdo con una cualquiera de las reivindicaciones 1 a 15".

Aunque dicha reivindicación se encuentra en la categoría de producto, al analizar la caracterización de dicho producto se observa que solo está definiéndolo porque incluye fibras de las reivindicaciones 1 a 15. Esto da a entender que se espera reclamar el uso o aplicación de las fibras de polímero en cualquier producto pero utilizando expresiones indirectas en la reivindicación; esto no es posible aceptar porque está implícitamente reclamando el uso de las fibras en cualquier producto.

Se recomienda eliminar dicha reivindicación del capítulo reivindicatorio.

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- En la reivindicación 18 se menciona que el producto se encuentra en la forma de una tela, vellón, estera de fibras trozadas, estera de fibras largas, producto no tejido, producto unidireccional, cuerda, red, cinta, material continuo o tira, o de alguna otra manera en la forma de una mezcla de dichas fibras con fibras de un tipo diferente, y preferentemente en forma de fibras entremezcladas.

En la reivindicación 19 se menciona que el producto incluye una matriz mineral, preferentemente un material de fraguado hidráulico, y se elige más preferentemente el producto entre adhesivos, morteros, concretos, lechadas de cemento y cemento de fibras.

En la reivindicación 21 se menciona que el producto tiene la forma de una lámina plana o corrugada.

En la reivindicación 22 se menciona que el producto incluye una matriz de polímero y porque se elige preferentemente entre una matriz de polietileno, polipropileno, poliamida, poliéster, epoxi o fenólica.

Como puede observarse, las reivindicaciones mencionadas incluyen un gran número de productos a los cuales puede incorporarse las fibras de polímero de las reivindicaciones 1 a 15. Sin embargo, con dicha información se da a entender que se espera la protección de los diversos productos en los cuales pueden estar dichas fibras, esto es equivalente a reclamar los usos y segundos usos de las fibras lo cual no es posible aceptar en el capítulo reivindicatorio.

Se recomienda entonces eliminar dichas reivindicaciones o realizar modificaciones de claridad según se menciona en el numeral correspondiente de la presente comunicación.

- En conclusión, las reivindicaciones 16, 17, 18, 19, 21, 22 corresponden a usos de las fibras de las reivindicaciones 1 a 15 y por tanto no pueden ser aceptadas.

6. Determinación del Estado de la Técnica

- La fecha que se ha tenido en cuenta para determinar el Estado de la Técnica es:

Fecha de presentación de la solicitud prioritaria: **25 de Mayo de 2004**

- Teniendo en cuenta la fecha indicada se realizó la búsqueda en el estado de la técnica de documentos relacionados con la solicitud encontrándose los siguientes:

Nº	Documento	Título	Fecha de publicación
D1	JP2180771	"FIBER REINFORCED CEMENT MOLDED BODY"	13 de Julio de 1990
D2	JP5044163	"POLYOLEFIN FIBER HAVING HIGH PRECIPITATION RATE IN WATER"	23 de Febrero de 1993
D3	US2002110686	"FIBERS INCLUDING A NANOCOMPOSITE MATERIAL"	15 de Agosto de 2002
D4	Fibres and Polymers 2004, Vol. 5, No. 1, 59 – 67	"STUDIES ON COMPOSITE FILAMENTS FROM NANOCLAY REINFORCED POLYPROPYLENE"	Marzo de 2004

7. Análisis de patentabilidad (artículo 14 D 486)

7.1. Novedad (artículo 16 D 486)

- Respecto a la Solicitud en estudio

Una fibra de polímero cargado (Reivindicaciones 1 a 15)

En la reivindicación independiente 1 se menciona una fibra de polímero cargado que contiene aditivos dentro de ella, teniendo la fibra de polímero cargado un Módulo de Young superior al de una fibra de polímero sin cargar caracterizada porque los aditivos incluyen aditivos minerales que tienen por lo menos una dimensión submicrónica.

En la reivindicación independiente 27 se menciona una composición para un material de fraguado hidráulico caracterizado porque incluye un ligante hidráulico y fibras de polímero cargado de acuerdo con las reivindicaciones 1 a 15.

En la reivindicación independiente 28 se menciona una composición caracterizada porque incluye una matriz de polímero y fibras de polímero cargado de acuerdo con una cualquiera de las reivindicaciones 1 a 15.

- Respecto al Estado de la Técnica:

En el documento **JP2180771** (D1) se divulgan fibras de refuerzo para cemento. Las fibras son obtenidas a partir de una película de polipropileno que comprende de 15 – 30% en peso de una carga inorgánica de talla submicrónica. Los ejemplos en las páginas 5 – 6 de D1 son efectuados utilizando una sílice de tala media de 230 nm.

En el documento **JP5044163** (D2) se describen unas fibras de refuerzo para cemento. Estas fibras son elaboradas a base de poliolefina y contienen carbonato de calcio en forma de polvo con diámetro de 50 nm (Ver ejemplos y Tabla 2). Estas fibras son igualmente tratadas en su superficie. Las fibras de D2 presentan el mismo título que las de la solicitud (Ver tabla 2).

En el documento **US20020110686** (D3) se divulgan fibras que incluyen un material nanocompuesto. El material incluye una matriz de resina polimérica y una pluralidad de partículas inorgánicas tales como arcillas, en forma de plaquetas con por lo menos una dimensión en nanotamaño y dispersadas en ella (Ver Resumen, párrafo 7 y 8).

En D3 se enseña que tales partículas tienen un diámetro promedio entre 10000 Angstrom y 50 Angstrom (1000 nm – 5 nm) y una relación de longitud/espesor en el rango de 1000 a 1. Las partículas tienen típicamente un espesor promedio de menos que 20 Angstrom (2 nm) y preferiblemente de menos de 10 Angstrom (1 nm). Esto en contraste a los materiales rellenos convencionales tales como caolín, dióxido de titanio, carbono y similares, los cuales tienen grandes tamaños y típicamente son sustancialmente esféricas (Párrafo 8).

En D3 se explica que las fibras resultantes pueden presentar resistencia mejorada, por ejemplo resistencia a la tensión y módulo flexural comparado con las fibras que no tienen nanocompuestos.

Las partículas de arcilla pueden ser de diversa naturaleza, entre las cuales se puede encontrar la montmorillonita (Párrafo 15). También, dichas partículas se cargan en el rango de 0,1 a 5% en peso (Párrafo 16).

Las resinas de poliolefina que forman la matriz pueden ser polipropileno, polietileno, polibuteno y polimetil penteno, poliamidas, incluyendo nylon 6, poliésteres incluyendo polietilen tereftalato, politrimetilen tereftalato y poliésteres alifáticos como ácido poliláctico (Párrafo 22).

Las fibras pueden incluir otros componentes como antioxidantes, estabilizantes, pigmentos, y similares (Párrafo 23) y dichas fibras pueden ser utilizadas para hacer telas tejidas o no tejidas (Párrafos 29 a 31).

En el documento **"STUDIES ON COMPOSITE FILAMENTS FROM NANOCCLAY REINFORCED POLYPROPYLENE"** (D4) se divulgan nanocompuestos de polipropileno/montmorillonita. Las organoarcillas utilizadas se dispersaron en polipropileno a escalas de nanómetros.

En D4, se describen filamentos poliméricos a base de polipropileno que incluyen arcillas de dimensiones nanométricas (Resumen, página 59, párrafo "Preparación de mezclas maestras", Tabla 1, página 65, párrafo "SEM"; Figuras 1 – 8).

Las arcillas de D4 reciben tratamiento superficial mediante sales de amonio. En dicho documento se enseña que dichas arcillas son utilizadas para mejorar las propiedades mecánicas de las fibras (Resumen, párrafo de introducción y de conclusión, Figura 5).

D4 se divulga un estudio sobre compuestos de filamentos de polipropileno reforzados con nanoarcillas. En este estudio, nanoarcillas de montmorillonita de sodio pura fue modificada usando bromuro de hexadecil trimetil amonio (HTAB) vía reacción de intercambio iónico. Esta arcilla fue mezclada con polipropileno en presencia de un agente.

El compuesto de filamentos a partir de PP/nanoarcillas fue preparado en diferentes porcentajes de peso de nanoarcillas. Los compuestos obtenidos fueron caracterizados por sus propiedades mecánicas, morfológicas y térmicas.

En D4 se enseña que la preparación del compuesto se realizó mediante la dispersión de nanoarcillas en una matriz de polipropileno mediante mezclado por fusión. El mezclado y la extrusión de filamentos fueron llevados a cabo en un extrusor de tornillo sencillo.

El rango de tamaño de partícula de las nanoarcillas es de 100 a 500 nm pero se menciona que idealmente, el tamaño de partícula debería estar por debajo de 100 nm.

- Comparación elemento por elemento

El elemento característico de la fibra reivindicada es la incorporación de aditivos minerales que tienen por lo menos una dimensión submicrónica. Sin embargo, cualquiera de los documentos D1 a D4 tomados por separado sin ningún tipo de combinación, divulgan claramente que ya era conocido incorporar aditivos minerales tales como arcillas de montmorillonita con por lo

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menos una dimensión submicrónica a las fibras de polipropileno para obtener compuestos y productos con propiedades físicas y mecánicas mejoradas.

En conclusión, las fibras de polímero cargado, los productos y los compuestos de la solicitud en estudio no son Nuevos y por ello no cumplirían con el artículo 16 de la Decisión 486.

8. Conclusión

Los requisitos de patentabilidad de la presente solicitud se encuentran afectados así:

N°	Documento N°	Reivindicaciones afectadas	Requisito que afecta
D1	JP2180771	1 – 15; 27	Novedad
D2	JP5044163	1 – 15; 28, 29	Novedad
D3	US2002110686	1 – 15; 28, 29	Novedad
D4	Fibres and Polymers 2004, Vol. 5, No. 1, 59 – 67	1 – 15; 28, 29	Novedad

En cumplimiento del artículo 45 de la Decisión 486 de la Comisión de la Comunidad Andina, se le indica que debe dar respuesta dentro del término de **60 días** contados a partir de la fecha de notificación de la presente comunicación. **En caso de no dar respuesta al presente requerimiento, o si a pesar de la respuesta subsistieran los impedimentos para la concesión, se denegará la patente.**

Notifíquese el presente oficio conforme a lo dispuesto en el numeral 5.2, literal e), del capítulo quinto del título primero de la Circular Única N° 10 de 2001.


ALIX CARMENZA CÉSPEDES DE VERGEL
Directora de Nuevas Creaciones (C)

El anterior requerimiento fue notificado por fijación en lista, de fecha 26 MAR 2010

CONCEPTO TÉCNICO
No. 0167

EXAMINADOR TÉCNICO
Código No. 202042