



## (51) International Patent Classification:

**C08L 3/04** (2006.01)      **C08L 97/02** (2006.01)  
**C08L 23/06** (2006.01)      **C08J 5/04** (2006.01)  
**C08L 23/08** (2006.01)      **C08J 11/00** (2006.01)  
**C08L 23/12** (2006.01)      **C08K 7/02** (2006.01)  
**C08L 23/14** (2006.01)      **B29C 67/24** (2006.01)  
**C08L 55/02** (2006.01)      **B33Y 70/00** (2015.01)  
**C08L 67/02** (2006.01)      **B29C 67/00** (2006.01)  
**C08L 67/04** (2006.01)      **C08K 7/16** (2006.01)

## (21) International Application Number:

PCT/IB2016/051882

## (22) International Filing Date:

1 April 2016 (01.04.2016)

## (25) Filing Language:

Italian

## (26) Publication Language:

English

## (30) Priority Data:

102015000010935 3 April 2015 (03.04.2015) IT

## (72) Inventor; and

(71) Applicant : **MILAZZO, Giovanni** [IT/IT]; Via Don Milani 35, 95047 Paterno' (CT) (IT).(74) Agent: **BOTTERO, Carlo**; c/o Barzano' & Zanardo Milano S.P.A., Via Borgonuovo, 10, 20121 Milano (IT).

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

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

## Published:

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

(54) Title: COMPOSITE MATERIAL COMPRISING AT LEAST ONE THERMOPLASTIC RESIN AND GRANULAR SHIVE FROM HEMP AND / OR FLAX

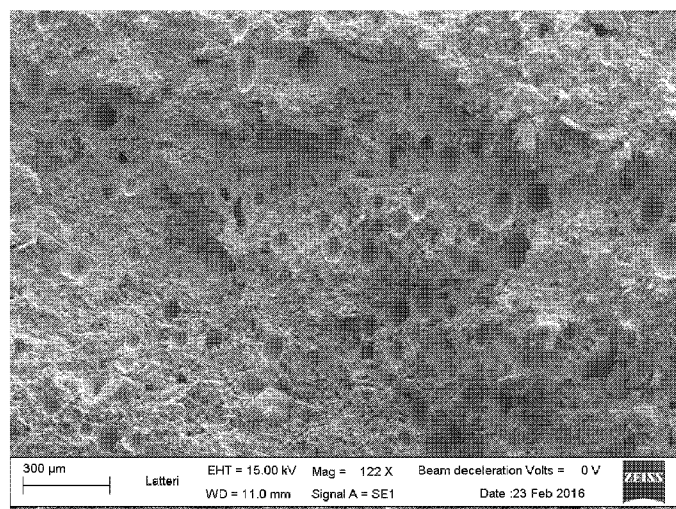


Fig. 2

(57) Abstract: The present invention relates to a composite material comprising at least one thermoplastic resin and granular shive from hemp and/or flax. The invention further relates to a method for preparing the above-mentioned composite material and the use of the latter in the 3D printing technologies, namely the manufacture of three-dimensional objects by additive manufacturing, starting from a digital 3D model.

**COMPOSITE MATERIAL COMPRISING AT LEAST ONE THERMOPLASTIC RESIN AND  
GRANULAR SHIVE FROM HEMP AND / OR FLAX**

-----

5       The present invention relates to a composite material comprising at least one thermoplastic resin and granular shive from hemp and/or flax.

      The invention further relates to a method for preparing the above-mentioned composite material and the use of the latter in the 3D printing technologies,  
10       namely the manufacture of three-dimensional objects by additive manufacturing, starting from a digital 3D model.

      The composite materials are obtained thanks to the combination of two or more materials different from  
15       each other and they are widely used thanks to their enhanced physical-mechanical properties, among which higher resistance and long-duration with respect to those of the single starting materials.

      Various composite materials comprising matrices of  
20       thermoplastic resins and components of natural origin, in particular natural fibers derived from Kenaf, hemp, flax, jute, henequen, leafs of pineapple, sisal, wood and sawdust are currently known. The possibility to recycle the processing scraps of such components of  
25       natural origin raises particular interest.

      Generally, the spread of the processing of hemp or flax and the resulting production of waste products shifted the interest on the recycle of the waste by-products obtained.

30       The products obtained from the processing of the hemp or flax plant are the long fibers (used in the textile industry), the bast fibers or bast (from which

the cellulose is obtained) and the ligneous stems or shive.

These latter two products, which compose the inner core of the long fibers which are removed, can be  
5 obtained by the process of scutching, through which the ligneous core of the stems, after maceration, is broken, so as to obtain the separation of bast from shive.

The Applicant considered how to provide a composite  
10 material having enhanced physical-mechanical properties and more lightness, starting from waste material from hemp or flax processing and which, at the same time, has an enhanced workability and is cheaper.

The Applicant was able to obtain such results by  
15 using, into a composite material, a component having a fine particle size derived from waste of hemp or flax processing, to date used as fertilizer, as fuel pellet, in the field of green-building, together with lime and as animal litter.

Particularly, the Applicant found that the above-  
20 mentioned problem is solved through a composite material comprising a thermoplastic resin and shive from hemp and/or flax with fine particle size, wherein resin and shive are present in a certain ratio by  
25 weight.

Therefore, according to a first aspect, the present invention relates to a composite material comprising at least one thermoplastic resin and from 5 to 180 parts by weight of granular shive from hemp and/or flax, with  
30 respect to 100 parts by weight of the thermoplastic resin, with particles having an average particle size lower than 0.2 mm, preferably lower than 0.1 mm.

Advantageously, the composite material according to the present invention has an enhanced workability, as the granular shive from hemp and/or flax with particle size lower than 0.2 mm, preferably 0.1 mm, disperses in  
5 a more homogenous manner with the thermoplastic resin, both with respect to other components derived from hemp with a higher particle size, and with respect to other materials with a particle size lower than 0.2 mm, for example sawdust.

10 Another advantage related to the particle size characterizing the shive according to the invention is to allow the use of high amounts of shive with respect to the amount of resin, determining both a higher lightness of the resultant material and an economical  
15 advantage, as it allows to decrease the amount of resin used in the composite material, without adversely affecting the thermoplastic properties of the composite material obtained. Furthermore, the shive used in the present invention represents a waste material of the  
20 hemp and/or flax processing, and therefore its reuse does not involve additional costs, indeed it represents a manner for using such waste product.

Furthermore, the addition of shive according to the present invention allows to obtain a composite material  
25 with enhanced processability in the molten state with respect to, for example, the addition of short hemp fibers, which tend to increase the viscosity of the melted and to create obstructions when the material is processed by the passage through nozzles with a very  
30 small particle size, for example lower than 2 mm or also lower than 0.4 mm. That makes the material according to the invention particularly suitable for

the manufacture of three-dimensional objects by 3D printing, which requires the use of nozzles of those sizes.

Further features and advantages of the present invention will be evident from the following detailed description.

In the present description and attached claims, the wording "granular shive from hemp and/or flax with particles having an average particle size lower than 0.2 mm, preferably lower than 0.1 mm" (also called "fine shive" or "shive") means a fine powder obtained from the processing of the large shive, for example by grinding. The fine shive can also derive from the suction of powders which disperse during the processing of hemp straws. Similarly, in the present description, the term "large shive" (which generally has a particle size of 1-2 cm) means the shive obtained by separation (for example by scutching) of the shive itself (also known as "woody stem") from the bast fiber (also called "bast").

Particularly, the granular shive from hemp and/or flax in general has substantially spherically-shaped particles, which is not to be confused with the so called short hemp fibers which are characterized by a fibrillary structure. Such structural difference can be appreciated, for example, by optical microscope observation or, preferably, by scanning electron microscope (SEM) observation.

Preferably, the particles of shive have an average particle size from 5  $\mu\text{m}$  to 300  $\mu\text{m}$ , more preferably from 10  $\mu\text{m}$  to 100  $\mu\text{m}$ . The average particle size can be determined according to the known techniques.

Particularly, for average sizes such as those indicated above, the laser diffraction technique, according to the standard ISO 13320-1 (1999), is generally used. As for higher sizes (up to 0.2 mm) techniques based on the analysis of images obtained by microscope can be used.

A parameter which can be used for characterizing particles of shive is the aspect ratio, namely the ratio between the higher diameter and the smaller diameter, perpendicular to the higher diameter, determined on a projection onto the plane of each particle (Feret diameter). Such parameter can be determined through the analysis of images obtained by the microscope, as described in the standard ISO 9276-6 (2008).

According to a preferred embodiment of the present invention, the shive has an aspect ratio from 0.5 to 2.0, preferably from 0.8 to 1.2, even more preferably from 0.9 to 1.1.

According to another preferred embodiment the composite material according to the present invention further comprises from 0.1 to 60 parts by weight of bast fibers (bast) from hemp and/or flax having a length from 0.5 cm to 4 cm, preferably from 0.6 cm to 2 cm.

According to another preferred embodiment, the used thermoplastic resin can be of natural or synthetic origin. When the thermoplastic resin is of synthetic origin, it is preferably selected from polyethylene terephthalate (PET), polypropylene (PP), polyethylene (PE), acrylonitrile-butadiene-styrene (ABS) copolymer, polypropylene/polyethylene terephthalate copolymer, ethylene-propylene copolymer. When the thermoplastic

resin is of natural origin, it is preferably selected from polylactic acid (PLA), polyhydroxyalkanoates (PHA), modified starches (such as, for example, those known under the commercial name "Mater B") or  
5 polyethylene from bioethanol (known under the commercial name "BIO PET 30"), more preferably PLA, which are characterized by high biodegradability.

Preferably the composite material according to the invention further comprises a particle-shaped component  
10 having an average particle size lower than 0.2 mm, preferably lower than 0.1 mm, for example a component derived from coconut shell, namely the hard shell of coconut (brownish covering), to which the meat (pulp) adheres and which needs to be broken in order to reach  
15 the meat itself. Such component is preferably obtained by breaking operations of the hard shell of coconut, which are carried out in order to take the pulp. Furthermore, such component can derive by grinding coconut shell residues.

20 According to a preferred embodiment of the invention, the composite material further comprises a binding agent, which allows to improve the mechanical properties of the material itself. It is believed that such binding effect is obtained thanks to the  
25 capability of the binding agent of binding the silicates present in the shive. Preferably the binding agent is selected from selected from alkali metal oxides and/or alkaline-earth metal oxides, preferably calcium oxide.

30 According to a second aspect, the present invention relates to a method for the manufacture of the composite material as defined above comprising the

steps of:

- melting at least one thermoplastic resin;
- mixing said at least one molten resin with from 5 to 180 parts by weight, with respect to 100 parts by weight of the thermoplastic resin, of granular shive from hemp or flax with particles having an average particle size lower than 0.2 mm, preferably lower than 0.1 mm;
- cooling the mixture obtained in order to form said composite material.

According to a third aspect, the present invention relates to the use of the composite material as defined above for the manufacture of three-dimensional objects by 3D printing. As known, the 3D printing is an additive manufacturing of three-dimensional objects, starting from a digital 3D model. One of the most widespread techniques is the so-called "fused deposition modeling" (FDM), which provide the overlapping of thin layers of thermoplastic material in the molten state obtained starting from filaments which are directed to an application head where the filament is melted and placed on a platform by nozzles, thus forming subsequent layers according to the 3D model provided. The composite material according to the invention is in the filament form, for example filament coils or rolls, which are directed to the application head of the 3D printer. The use of the composite material according to the invention in the field of the 3D printing advantageously allows to avoid the formation of obstructions in the application nozzles, which instead are often formed when composite materials containing a fibrous component are used, for example



hemp fibers, which have a marked tendency to adhere to the nozzles walls, thus impeding the material deposition.

The present invention has been described for  
5 illustrative but not limitative purposes, according to its preferred embodiments, but it is to be understood that modifications and/or changes can be introduced by the persons skilled in the art without departing from the relevant scope of protection as defined in the  
10 enclosed claims.

#### Brief description of the drawing

The present invention will be now described, for illustrative, but not limitative purposes, according to its preferred embodiments, with particular reference to  
15 the figures of the enclosed drawings, wherein:

- figure 1 shows the results of the tensile tests carried out on the composite material according to the invention;
- figure 2 shows the SEM image related to the  
20 composite material according to the invention;
- figures 3 and 4 show the SEM images related to the composite material containing hemp fibers in place of granular shive.

The present invention will now be further  
25 illustrated by certain example of embodiments as reported below.

#### EXAMPLE 1

The composite material according to the invention was prepared by placing into a vessel made of aluminum  
30 about 2g of PLA (equal to about twenty granules of PLA)

and then heated on a plate to a temperature of 300 °C in order to obtain the PLA melting. Then, about 1 g of shive constituted by particles having an average particle size lower than 0.1 mm was added, namely equal  
5 to about 40% by weight with respect to the total weight of the composite material obtained and then the whole was homogenized by mixing for at least 5 minutes, decreasing the plate temperature. The composite material obtained had good workability and it was  
10 placed on an aluminum foil and a sample in the plate-form having a thickness equal to about 3 mm was obtained by pressing, which was left to cool up to hardening. The sample was subjected to several tests in order to test the physical-mechanical properties of the  
15 sample itself. The results showed that the material tested is rigid, hard and it has properties of flexural, tensile and impact strength, equal to or higher than the thermoplastic resin alone. Furthermore, once hardened, the sample was subjected to conditions  
20 of water washout mechanical stress keeping intact its properties, without undergoing degradation.

#### EXAMPLE 2

A further test was carried out starting from the sample obtained in the example 1 in order to verify the  
25 possibility to rework the thermoplastic resin already produced with shive so as to reuse it without causing the separation between the phases.

Particularly, the sample obtained in the preceding example was melted into a 100 ml beaker on a heating  
30 plate, at a temperature of 300°C (mixture 1). In order to avoid the degradation of the thermoplastic polymer, the material was subjected to stirring. Simultaneously,

2g of PLA were melted on a heating plate into a 100 ml beaker, and then shive 1 g was gradually added. At the same time of any shive addition, the whole was mixed. After having obtained an homogeneous mixture (mixture 2), the latter was combined with mixture 1 and the whole was mixed, maintaining the plate temperature at 300°C. The obtained composite material had good workability and it was placed on an aluminum foil and a sample in the plate-form having a thickness of 2-3 mm was obtained by pressing, which was left to cool up to hardening. The sample was subjected to several tests in order to test the physical-mechanical properties of the sample itself. The results showed that the material tested has surprising hardness, tensile, flexural mechanical properties and it has good properties of resilience in addition to a low weight.

#### EXAMPLE 3

A test, in which polypropylene (PP) was used as thermoplastic resin, was carried out. 2 g of PP were placed into a 100 ml beaker and melted on a heating plate at 300 °C. Simultaneously, about 1 g of shive, with an average particle size lower than 0.1 mm, was weighted in a crucible. After having achieved the temperature of 100°C, namely when PP was almost completely melted, the shive was gradually added. The sample was subjected to several tests in order to test the physical-mechanical properties of the sample itself, particularly the obtained material was subjected to water flow (for 10 min), showing a great resistance to water. Furthermore, such material showed an excellent tensile strength.

#### EXAMPLE 4 (comparison)

The examples 1-3 were repeated using sawdust in place of shive, wherein said sawdust had a particle size comparable to the particle size of the shive used in examples 1-3, namely a particle size lower than 0.1 mm, wherein the sawdust was present in an amount of 50% by weight of the total composition with respect to the resin. As in the preceding examples, the obtained material was subjected to several tests in order to test the physical-mechanical properties thereof. The results showed that the tested material has worse workability than the materials obtained in the examples 1-3.

#### EXAMPLE 5

Tensile tests were carried out on the composite material of the invention obtained in the example 1 in order to determine certain mechanical characteristics. The properties considered and the results are reported in Figure 1.

#### EXAMPLE 6

Scanning electron microscope (SEM) analyses were carried out in order to structurally characterize shive from hemp. The shive considered was in powdery and volatile form and, for this reason, it was not possible to carry out a direct analysis through electronic microscopy as the SEM device requires high vacuum to be applied within the chamber containing the sample, which is evidently incompatible with a sample of that type. Therefore, in order to overcome such problem, the analysis was carried out on the composite material of the present invention containing PLA as thermoplastic resin and shive as filler, in correspondence of the fracture areas of the material itself. Mixtures of PLA

and shive were made in several ratios from each other, in particular 5, 10, 15, 20 and 25 parts by weight of shive respectively, with respect to 100 parts by weight of PLA, wherein the shive has an average particle size of about 50  $\mu\text{m}$ . For this purpose a Brabender Plastograph mixer was used at a temperature of 170°C for 10 minutes. Samples of material thus obtained were fractured and analyzed by scanning electron microscope SEM. As showed in figure 2, the microscope analysis highlighted as the samples containing shive have cavities attributable to the granular particles of shive which were extracted from the matrix after the fracture.

From the microscope analysis it is evident as the nature of the shive is of the granular type.

EXAMPLE 7 (comparison)

The example 5 was repeated using, in the place of shive, hemp fibers having an average length of 2 mm and the scanning electron microscope (SEM) analysis highlighted (see Figures 3 and 4) the presence of fibrillar structures only and the absence of cavities in the samples of material obtained. Furthermore, the fibrillar structures have poor adhesion with respect to the matrix of thermoplastic resin.

25

## CLAIMS

1. Composite material comprising at least one thermoplastic resin and from 5 to 180 parts by weight of granular shive from hemp and/or flax, with respect  
5 to 100 parts by weight of the thermoplastic resin, with particles having an average particle size lower than 0.2 mm.
2. Composite material according to claim 1, wherein the  
10 particles of shive have an average particle size lower than 0.1 mm.
3. Composite material according to any one of the preceding claims, wherein the particles of shive have an aspect ratio from 0.5 to 2.0, preferably from 0.8 to 1.2, even more preferably from 0.9 to 1.1.
- 15 4. Composite material according to any one of the preceding claims, wherein the composite material further comprises from 0.1 to 60 parts by weight of bast fibers from hemp and/or flax having a length from 0.5 cm to 4 cm, preferably from 0.6 cm to 2 cm.
- 20 5. Composite material according to any one of the preceding claims, wherein the thermoplastic resin is of natural or synthetic origin.
6. Composite material according to claim 5, wherein the thermoplastic resin of synthetic origin is selected  
25 from polyethylene terephthalate (PET), polypropylene (PP), polyethylene (PE), acrylonitrile-butadiene-styrene (ABS) copolymer, polypropylene/polyethylene terephthalate copolymer, ethylene-propylene copolymer.
7. Composite material according to claim 5, wherein the  
30 thermoplastic resin of natural origin is selected from polylactic acid (PLA), polyhydroxyalkanoates (PHA), modified starches or polyethylene from bioethanol,

preferably PLA.

8. Composite material according to any one of the preceding claims further comprising a component with an average particle size lower than 0.2 mm, preferably  
5 lower than 0.1 mm, derived from the coconut shell.

9. Composite material according to any one of the preceding claims further comprising a binding agent selected from alkali metal oxides and/or alkaline-earth metal oxides, preferably calcium oxide.

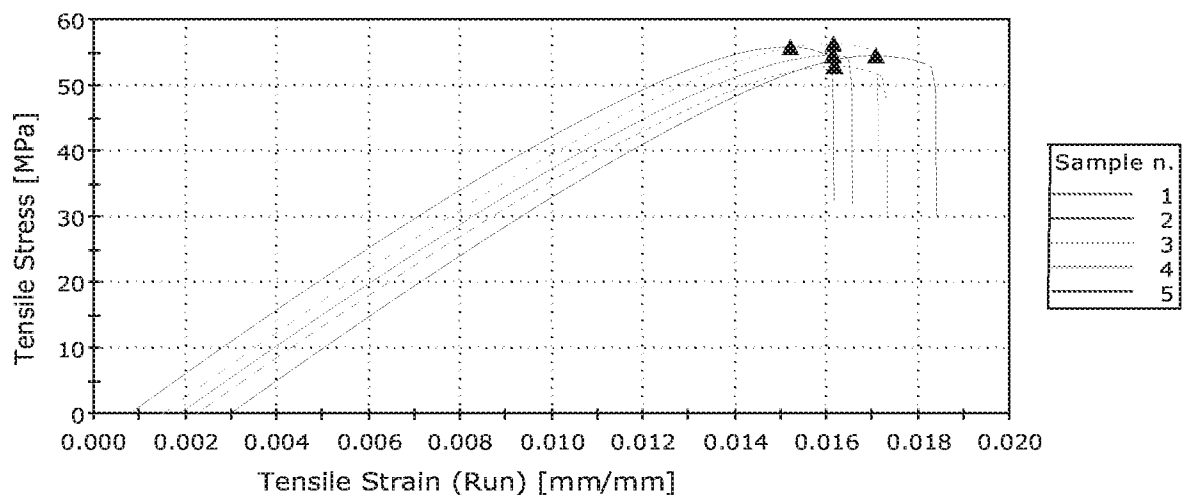
10 10. Method for the manufacture of a composite material as defined in claims 1-9 comprising the steps of:

- melting at least one thermoplastic resin;
- mixing said at least one molten resin with from 5 to 180 parts by weight, with respect to 100 parts by  
15 weight of the thermoplastic resin, of granular shive from hemp and/or flax with an average particle size lower than 0.2 mm, preferably lower than 0.1 mm;
- cooling the mixture obtained in order to form said composite material.

20 11. Use of the composite material according to any one of the preceding claims from 1 to 9 for the manufacture of three-dimensional objects by 3D printing.

1/4

## PLA shive



		Young's Modulus [GPa]	Stress at Yielding [MPa]	Strain at Yielding [%]
	1	4.861	-----	-----
	2	4.951	-----	-----
	3	4.979	-----	-----
	4	4.844	-----	-----
	5	4.914	-----	-----
Average		4.910	-----	-----
Standard				
Deviation		0.05748	-----	-----

		Stress at break [MPa]	Strain at Break [%]	Energy at Break [J]
	1	54.185	1.646	0.180
	2	54.227	1.533	0.198
	3	55.394	1.553	0.203
	4	51.299	1.500	0.184
	5	52.732	1.533	0.195
Average		53.567	1.553	0.192
Standard				
Deviation		1.58051	0.05544	0.010

Fig. 1



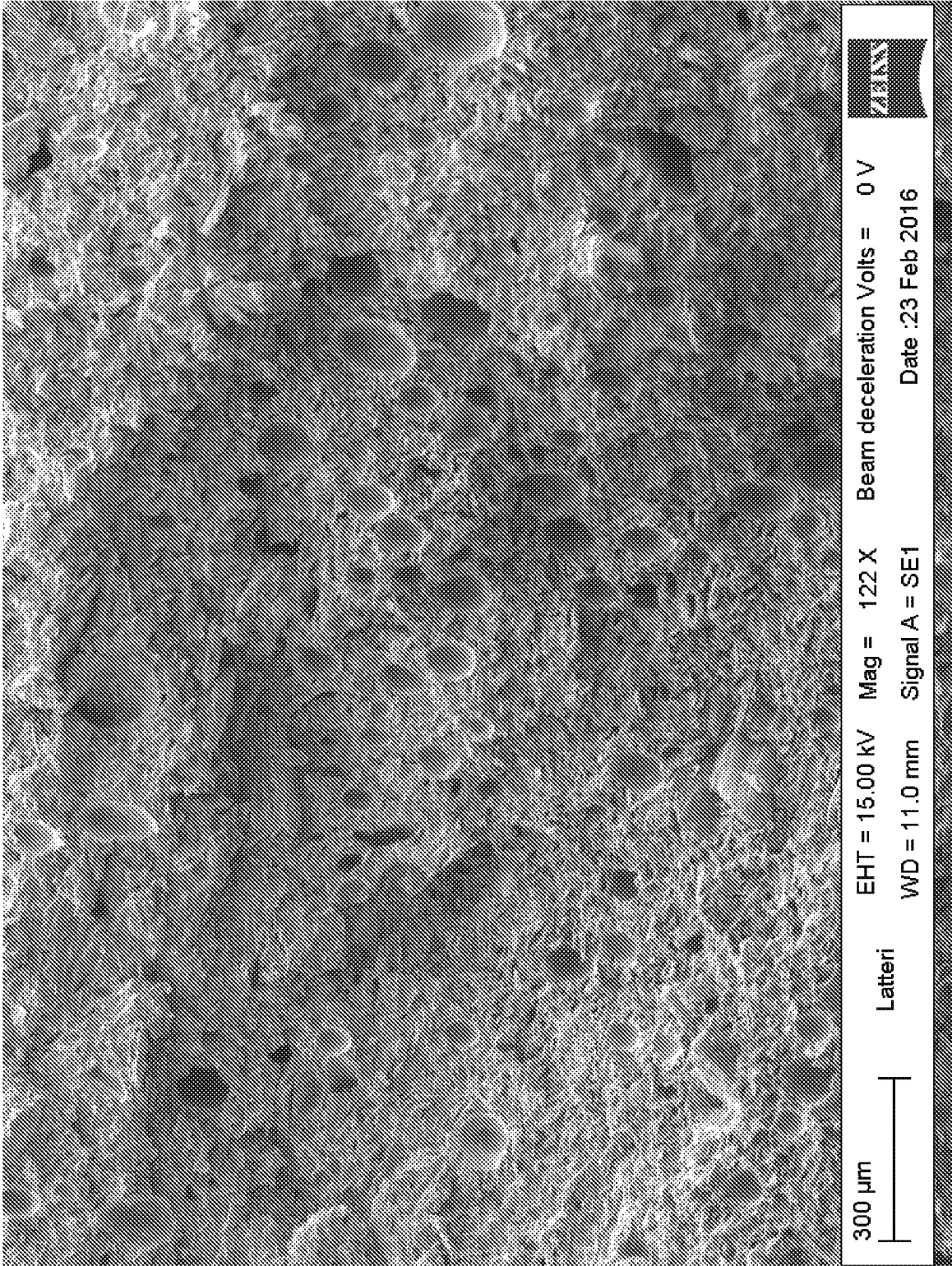
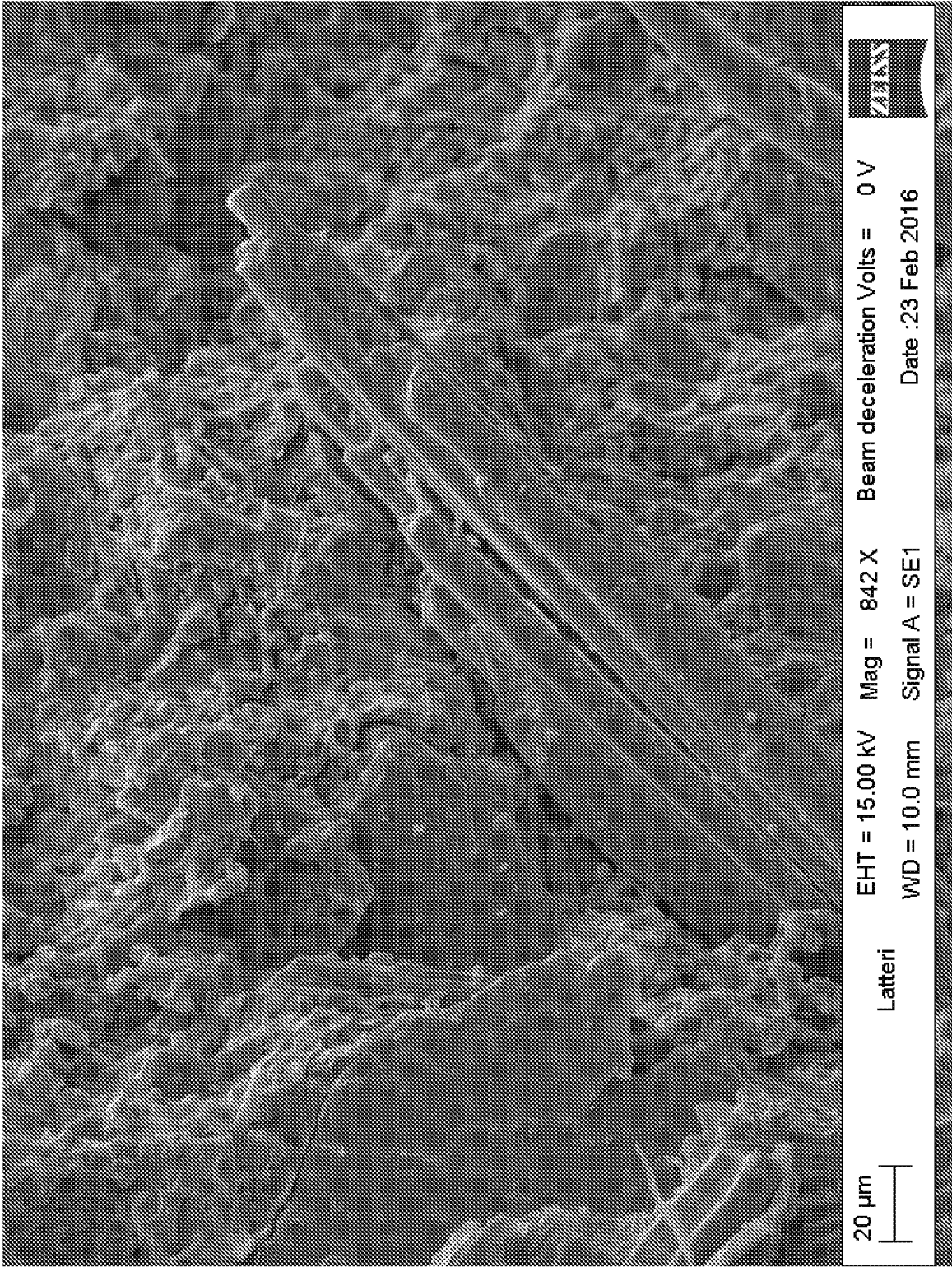




Fig. 3



# INTERNATIONAL SEARCH REPORT

International application No  
PCT/IB2016/051882

<b>A. CLASSIFICATION OF SUBJECT MATTER</b> INV. C08L3/04 C08L23/06 C08L23/08 C08L23/12 C08L23/14 C08L55/02 C08L67/02 C08L67/04 C08L97/02 C08J5/04 C08J11/00 C08K7/02 B29C67/24 B33Y70/00 B29C67/00				
According to International Patent Classification (IPC) or to both national classification and IPC				
<b>B. FIELDS SEARCHED</b> Minimum documentation searched (classification system followed by classification symbols) C08L C08J C08K B29C B33Y				
Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched				
Electronic data base consulted during the international search (name of data base and, where practicable, search terms used) EPO-Internal, CHEM ABS Data				
<b>C. DOCUMENTS CONSIDERED TO BE RELEVANT</b>				
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.		
Y	WO 00/05294 A1 (RAVACHOL ANDRE [FR]) 3 February 2000 (2000-02-03) claims 1, 2, 5, 6 page 7, lines 2-15 -----	1-11		
Y	EP 1 939 253 A1 (CHEIL IND INC [KR]) 2 July 2008 (2008-07-02) claim 5 examples 1-4 paragraphs [0025], [0026], [0041] -----	1-11		
Y	US 6 133 348 A (KOLLA FRANCIS A [CA] ET AL) 17 October 2000 (2000-10-17) claims 12, 18 column 4, lines 1-7 ----- <div style="text-align: right;">-/-</div>	1-11		
<div style="display: flex; justify-content: space-between;"> <span><input checked="" type="checkbox"/> Further documents are listed in the continuation of Box C.</span> <span><input checked="" type="checkbox"/> See patent family annex.</span> </div>				
<div style="display: flex; justify-content: space-between;"> <div style="width: 45%;"> <p>* Special categories of cited documents :</p> <p>"A" document defining the general state of the art which is not considered to be of particular relevance</p> <p>"E" earlier application or patent but published on or after the international filing date</p> <p>"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)</p> <p>"O" document referring to an oral disclosure, use, exhibition or other means</p> <p>"P" document published prior to the international filing date but later than the priority date claimed</p> </div> <div style="width: 45%;"> <p>"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention</p> <p>"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone</p> <p>"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art</p> <p>"&amp;" document member of the same patent family</p> </div> </div>				
Date of the actual completion of the international search  <div style="text-align: center; font-size: 1.2em;">21 June 2016</div>		Date of mailing of the international search report  <div style="text-align: center; font-size: 1.2em;">05/07/2016</div>		
Name and mailing address of the ISA/ European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016		Authorized officer  <div style="text-align: center; font-size: 1.2em;">Gerber, Myriam</div>		



## INTERNATIONAL SEARCH REPORT

International application No

PCT/IB2016/051882

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	<p>MERKEL K ET AL: "Processing and characterization of reinforced polyethylene composites made with lignocellulosic fibres isolated from waste plant biomass such as hemp", COMPOSITES PART B: ENGINEERING, ELSEVIER, UK, vol. 67, 22 June 2014 (2014-06-22), pages 138-144, XP029055734, ISSN: 1359-8368, DOI: 10.1016/J.COMPOSITESB.2014.06.007 *"2.3. Preparation of micro or nano-cellulose fibres (MCF/NCF-HS)" on pages 139-140*; abstract</p> <p>-----</p>	1-11
Y	<p>BAKRADZE G G ET AL: "Correlation between the mechanical properties and the amount of desorbed water for composites based on a recycled low-density polyethylene and linen yarn production waste", MECHANICS OF COMPOSITE MATERIALS, KLUWER ACADEMIC PUBLISHERS-CONSULTANTS BUREAU, NL, vol. 43, no. 5, 1 September 2007 (2007-09-01), pages 427-432, XP019555142, ISSN: 1573-8922, DOI: 10.1007/S11029-007-0040-0 *"Introduction" on page 427* *"Materials and Investigation Methods" on page 428*; abstract</p> <p>-----</p>	1-11
Y	<p>A. ETAATI ET AL.: "The study of fibre/matrix bond strength in short hemp polypropylene composites from dynamic mechanical analysis", COMPOSITES: PART B, vol. 62, 25 February 2014 (2014-02-25), pages 19-28, XP002750604, *"2. Experimental" on pages 20-22*; abstract</p> <p>-----</p>	1-11
Y	<p>GB 2 509 757 A (STAGE ONE CREATIVE SERVICES LTD [GB]) 16 July 2014 (2014-07-16) page 4, lines 4-5; claim 11</p> <p>-----</p> <p style="text-align: center;">-/--</p>	1-11

## INTERNATIONAL SEARCH REPORT

International application No

PCT/IB2016/051882

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	<p>TOENSMEIER P A: "CARNEVALLI DIVERSIFIES LINES IN BID FOR MARKET GROWTH", MODERN PLASTICS INTERNATIONAL, MCGRAW-HILL, INC. LAUSANNE, CH, vol. 29, no. 2, 1 February 1999 (1999-02-01), pages 30-32, XP000954751, ISSN: 0026-8283 *page 31, right-hand column, first full paragraph* *page 32, left-hand column, second and third full paragraphs* -----</p>	1-11

# INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No

PCT/IB2016/051882

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
WO 0005294	A1	03-02-2000	AU 4916199 A 14-02-2000
			FR 2781492 A1 28-01-2000
			WO 0005294 A1 03-02-2000
-----			
EP 1939253	A1	02-07-2008	CN 101200581 A 18-06-2008
			EP 1939253 A1 02-07-2008
			JP 4726887 B2 20-07-2011
			JP 2008150599 A 03-07-2008
			KR 100816679 B1 27-03-2008
			US 2008145656 A1 19-06-2008
-----			
US 6133348	A	17-10-2000	CA 2269408 A1 22-10-1999
			US 6133348 A 17-10-2000
-----			
GB 2509757	A	16-07-2014	NONE
-----			