

# Industrial paper recycling process: suitable micronization for additive polymer application

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## Abstract

*The traditional paper recycling process has problems related to the disposal of sludge and waste, the use of incinerators and water treatment. Because of that, an interesting alternative proposed from dep. of chemical and materials engineering of Sapienza University of Rome to Carlucci industrial typography, is using paper as filler in thermoplastics or recycled thermoplastic matrix composite. In this way it's possible re-use paper, but it also possible reduces the amount of polymer with equal volume. The paper has to be subjected by grinding. The chosen grinding process is fundamental to obtain a suitable product for composite. After a pre-grinding process obtaining 5-10 mm of paper fragments, the charge have to be subjected to a micronization process. In this study are selected two different type of micronization: the superfine grinding mill SF, a micronization process based on system that uses simultaneously the impact and friction action, and a knife mill that uses instead the cutting and friction action. Thanks to the first process, it is possible obtaining fibres with diameters of about 15-20 microns and lengths of the order of 250-500micron: a product therefore suitable for the application of filler in composites. This process unfortunately causes the production of fluff, as will shown in figure 2, because of the interfibrillar bonds between the fibers: an optimal dispersion of the fibers is necessary to avoid agglomerates which would decrease the composite properties. From the second process it is possible to obtain both fiber than particles as illustrated in figure 3. This morphologies mix allows to achieve a higher fluency preventing agglomerates. The turbomixer equipment realizes the production process of the composite, this process allows to introduce a higher percentage of filler respect the traditional injection moulding technique. The resulting composite is subjected by a morphological and mechanical characterization: look at the SEM analysis of the fracture surface, the fiber-matrix interface is weak, in spite of this, however, from the tensile test there is a constant elastic modulus and in some cases growing respect to the matrix devoid of fibres. From these results, the importance of optimize the grinding and micronization processes is clear and there is the opportunity of additives introduction to improve fiber-matrix interface.*

## Introduction

In Italy almost 9 millions of ton of cellulosic products are consumed, this shows the importance of recycling process to reduce the environmental impact.

From all the cellulosic waste, the 64% is used in recycling process, the 14% is sent to incinerator for thermovalorization and the last 22% is used for other applications (for example fireplace) or it is sent in waste dump. [1,2,4]

Paper recycling, economically speaking, is more advantageous than its incineration or its disposal.

Paper waste cost, avoiding collection costs and features plant used for the disposal, is near to 96-192 euro/ton. The recycling process, instead, allows a cost reduction to 40% less [3].

Paper, without ink, can be subjected to composting with other vegetable and animal materials to produce a fertilizer to improve soil property.

Paper recycling process has advantages like raw material's lower cost, the chance to use waste as fuel for steam production, the reduction of the amount of waste, of the storage

cost and trees' consumption. [4]

There are also some disadvantages: the possibility to reuse paper at least for 5-6 times, after that is not possible another use of the same so the waste paper again becomes a solid urban waste and needs disposal [5], the production of variable amount of slug and waste that have to be recovered and have to be subjected to different treatment, the CO<sub>2</sub> and NO<sub>x</sub> emission if is used gas as fuel and also SO<sub>2</sub>, dust and heavy metal if is used oil or coal.

Another pollution source is the incinerator both for the dust that developed in the process that for the residual material that hasn't completed the combustion [6].

Concluding, despite great economical advantages and the reduction of environmental pollution due to recycling process there are still problems because of disposal of slug and waste, the use of incinerators and water treatment. Moreover ink paper or additivated one (for example with polymeric film) can't be subjected to traditional recycling process and must accordingly be disposed as special waste.

In this contest fit the will of Carlucci, industrial typography, to find a new opportunity to manage its paper scraps; They think that the chance to use different kind of cellulosic waste as dispersed phase in composite materials with HDPE or other thermoplastic polymer matrix is interesting and participate at this project also thanks to a public founding (regional founding VAPETORE). In order of that the micronization process to obtain a product suitable for this application has to be optimized.

## Materials and Methods

### Materials

Eraclene MP90, commercial name of high density polyethylene (HDPE) from General Electrics (GE), has been chosen as matrix. Its properties are a melt flow index (MFI) of 7.0 g/10 min (190°C/2.16Kg), a density of 0.96 g/cm<sup>3</sup>, a tensile strength of 17 MPa, a flexural modulus of 1.45 GPa, a Shore D hardness of 69.

Paper used in recycling process or in thermovalorization was previously subjected to grinding by knife mill: it was possible to obtain in this way fragments of few centimetres.



Figure 1 – The micronizing unite of the superfine grinding mill type SF: in the process scheme 1 is the feeder, 2 is the mill and 3 is the static classifier.

In order to obtain paper suitable for incorporation in polymeric matrix, micronization treatment is needed: in this study two different kind of micronization process were conducted and below exposed.

#### 1. Superfine grinding mill type SF

Paper used in recycling process or in thermovalorization is previously subjected to grinding, in this way fragments of

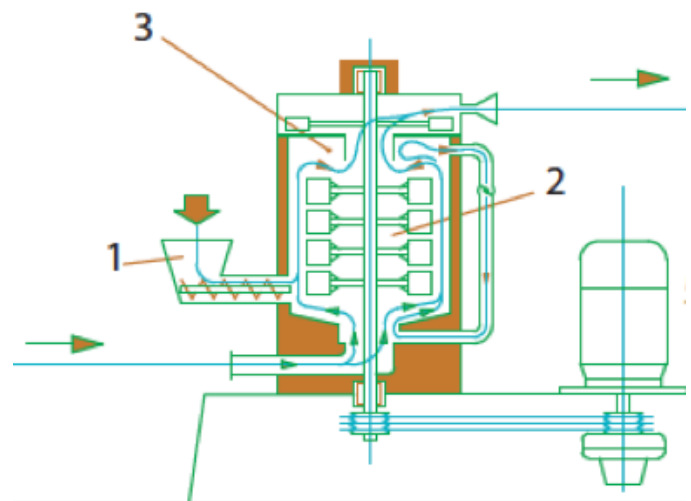
few centimetre are produced.

In order to obtain paper suitable for incorporation in polymeric matrix, micronization treatment is needed: a far from simple process due to the fibrous nature of the same.

A pre-grinding process in hammer mill is previously adopted, obtaining paper fragments of 8-10 millimetres to realize a product suitable for the next step, the charge is then sent to micronization mill that exploits simultaneously the action of impact, shear and turbulence [7]. It is a superfine grinding mill type SF from Cimma, which works in the design and construction of machines and systems for powder technology, in particular machines and grinding plants and pulverizing, screening, air classification, drying, dust filtration, pneumatic conveying and mixing. The figure 1 shows the micronizing unit of the adopted plant.

The process is here presented: the paper is introduced by a screw feeder or through a current of air sucked downward into the grinding chamber. At the exit of the grinding chamber, the ground product, swept upward by the airflow, is classified by the separator placed in the higher part of the mill. The fine material is discharged upwards and collected into a filter, while the reject of the separator is recycled into the mill.

Thanks to this process the complete opening of paper in cellulosic fiber of steady diameters of about 15-20 microns and



lengths of the order of 250-500 micron are obtained: a product therefore suitable for the application of filler in composites. The images below represent the fibres obtained.

#### 2. Knife mill SM 300

After a pregrinding treatment, adopted also in the previous process, to obtain 8-10 mm fragments of paper, the charge is sent to a knife mill. The process adopted is a knife mill SM



Figure 2 - (on the left) Micronized paper observed by stereomicroscope. Figure 3 (on the right) Micronized paper.

300 from Retsch, a German company that is active in the fields of neutral-to-analysis sample preparation and characterization of solids [8]. The figure 4 represents the scheme of the adopted knife mill.

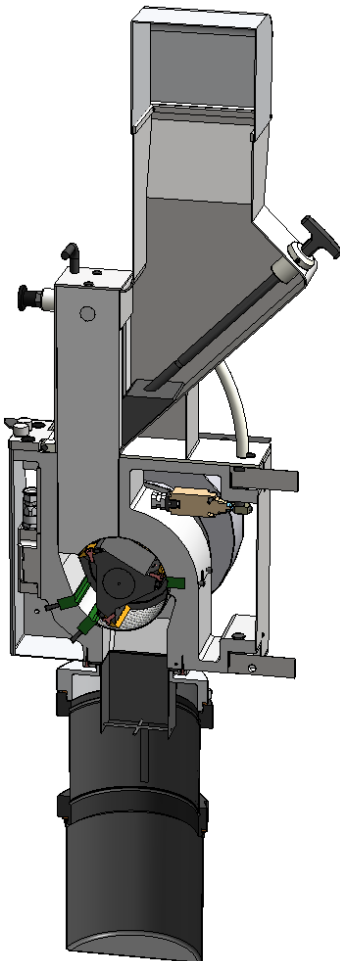


Figure 5 - The scheme of the adopted knife mill SM300.

Cutting bars of stainless steel are used in this mill.

The charge is introduced through a hopper into the grinding chamber. Here it is in contact with the rotor and it is subjected to size reduction as a result of the cutting and friction action. To increase the number of the cutting during the grinding, it is equipped with cutting bar with double cutting effect. The residence time of the charge in the chamber is generally short and depends on the type of sieve adopted: in this study sieve from 1 mm and 0,5 mm are used. the charge, once reached the size to pass through the mesh of the sieve, it is collected in a container. The speed of rotation varies between 700 and 3000 round/min: in this way a rapid reduction in the size is possible. The product obtained from this process is very different from the fibre obtained by the superfine grinding mill: in this case the charge is reduced in fragments from which some fibres are isolated.

The images below represents the product obtained from the knife mill.

The obtained product, using a sieve of 1mm, is composed mainly of fragments of paper, there is instead a lower amount of fibers. The fragments are about 500-800 micron, higher dimension compared to the fibers.

### The composite production

The main problem of the traditional injection moulding process is the low percentages of fibers that can be introduced and their poor dispersion, as we exposed in the previous work presented at 5<sup>th</sup> International Conference on Innovative Natural Fibre Composites for Industrial Applications [9] in which is clear the poor mixing dispersion with one screw extrusion implant. A new batch implant of mixing has developed in order to improve the fibres percentages in the poly-



Figure 5 (on the left) and figure 6 (on the right): the charge obtained from knife mill with a sieve of 0,5mm observed with the stereomicroscope (fig. 5), the knife mill product (fig. 6).

meric matrix: the turbomixer (the results and the implant presented are to be placed in the "Project for the promotion of a culture of innovation and technology transfer - marine sector" approved and funded by the Region of Lazio with determination n. B03034 of 22/05/2012. The actuator of this

Project is the Polo interprovinciale formativo della nautica Rome-Latina and the I.I.S.S. "G. Caboto" (is the leader). The turbomixer is composed of a cylindrical stator and a rotor with attached blades. The figure 7 represents the HDPE matrix and the cellulosic fibers introduction in the turbomixer.

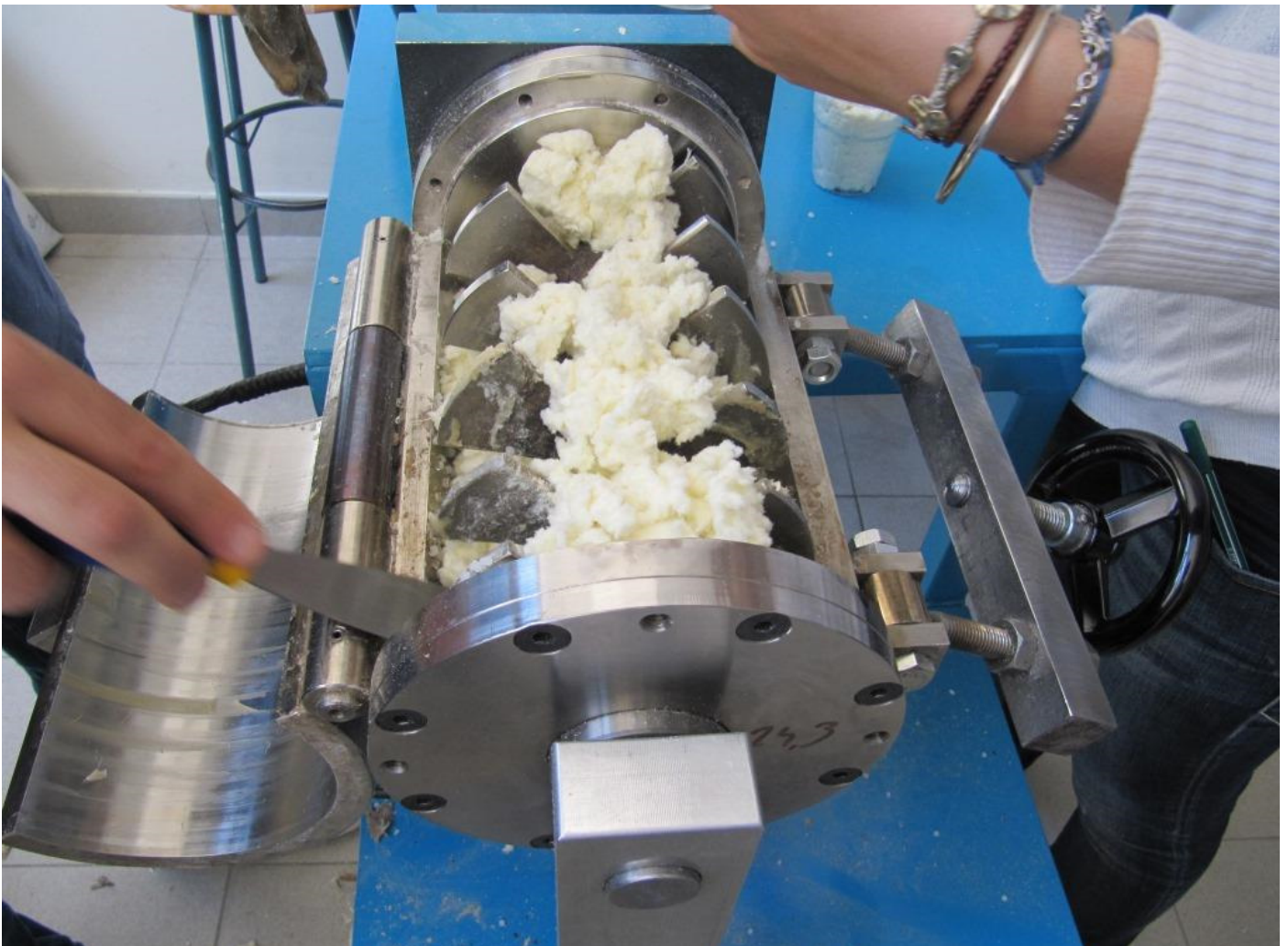


Figure 7 - The HDPE grains and cellulosic fibers introduction in the turbomixer.

The blades are oriented with different angles, because of the rotation in this way a suitable mixing is possible. The heat develops thanks to the conversion of mechanical energy into thermal one due to Joule effect.

There are three thermocouples to record the temperature: one of them is on the cylinder wall, one in the peripheral area and the last one in the centre zone.

Even if the heat develops in the same way of the endless screw of the extrusion system, in this case it is possible to reach a speed up to 3000 round/minute due to the only mixing of the charge without the possibility of progress in the mixer.

With this process the charge reaches the melting point, using HDPE the temperature is near to 170-180°C thanks entirely to heat develops from friction, the presence of additional heat source like electric heaters, is not necessary.

The turbo mixer has batch working: at the beginning there is the charge of the material, after that the closer of the cylinder, the activation of the mixing and in the end the hand discharge of the material.

The melted charge is then subjected to moulding under pressure, in this way it is possible to obtain tables from which specimen are derived.

The specimens are obtained with different percentages of filler: it is possible to introduce up to 20% of paper and it is also possible a better dispersion of the same.

The specimens are produced both with the product of the superfine grinding mill and the knife mill. The family samples tested are 10% of fibers derived from the superfine grinding mill and 20% of fibers derived from both the superfine grinding mill and the knife mill, the higher percentages of fibers has been chosen to compare the product from two different mill. Each family of different fibers percentages is composed

of five samples.

### Methods

#### The tensile test

The tensile test is carried out to measure the breaking load and the elastic modulus of both fibers produced with the superfine grinding mill and particles and fibers obtained from the knife mill.

The test was conducted in accordance with the norm ASTM D 3039 using a Zwick/Roell Z010 and a 1.8 MPa preload was applied. The specimens, obtained from 100mmx200mmx2mm (LxWxT) plates, have dimension of 200mmx12,5mmx2mm (LxWxT). The crosshead speed was 5 mm/min.

#### The scanning electron microscopy

The specimens were observed with SEM (Philips XL40) in order to analyse the fibers and particles dispersion in the matrix and the interface quality.

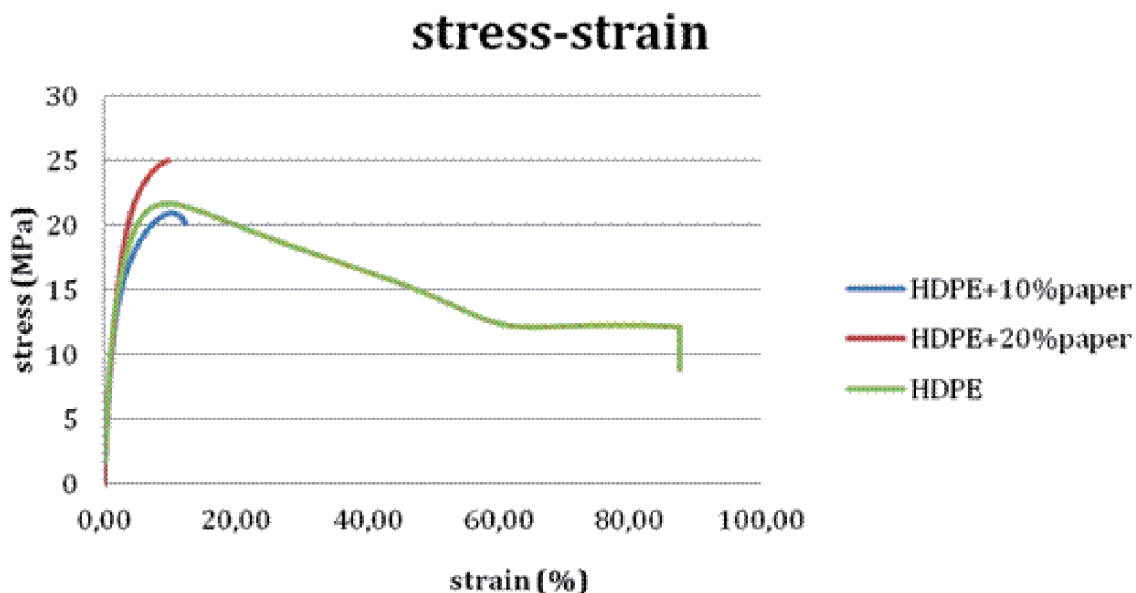
The samples were sputter-coated with gold particles before the surface characterization.

## Results and Discussion

#### The tensile test

The specimens obtained from the turbomixer are subjected to tensile test.

The graphic below represents the stress and strain curve of the specimens with different percentages of fibres produced with the superfine grinding mill compare to polyethylene without fibers.



Graphic 1 – The stress and strain curve conduct with 1,8 MPa preload with fibers from the superfine grinding mill and without them.

The elastic modulus slightly increases or is constant with the introduction of fibers. The same behaviour is shown by the breaking load, which is affected of a visible improvement with the higher percentage of fibers. This leads, however, a marked loss of material toughness.

The table 1 summarizes the main results of the tensile tests.

Table 1 – the main results of the tensile test for the samples with fibers from the superfine grinding mill. For each family, five samples were tested.

	Elastic modulus (MPa)	Breaking load (MPa)
HDPE	1160±86,6	21,59±0,18
10% fiber	1090±31,62	20,89±0,67
20% fiber	1250±18,26	24,61±1,22

The graphic 2 represents the stress and strain curve of the specimens with 20% of fibres produced with the knife mill compare to polyethylene without fibers and the composite with 20% of fibers produced with the superfine grinding mill. The s after paper in the graphic means the paper produced

but the breaking load is higher, the value is near to the composite produced with fibers from the superfine grinding mill with the same amount of fibers.

The table 2 summarizes the main results of the tensile test.

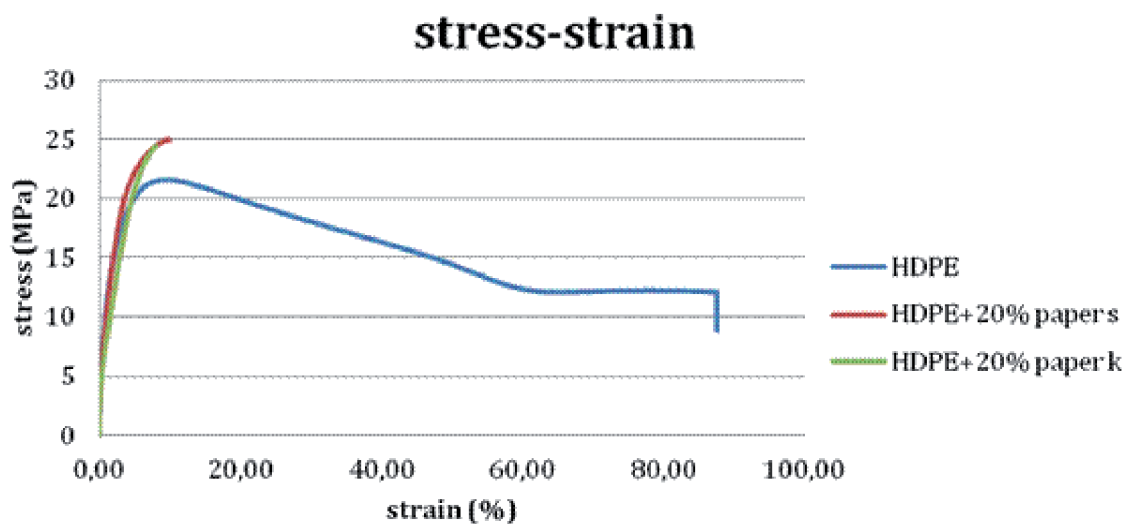
Table 2 the main results of the tensile test of the composite produced with 20% of fibers from the superfine grinding mill and the knife mill. In each family five samples were tested.

	Elastic modulus (MPa)	Breaking load (MPa)
HDPE	1160±86,6	21,59±0,18
20% fiber s	1250±18,26	24,61±1,22
20% fiber k	1067±77,6	24,75±1,68

### SEM analysis

All the specimens are observed by scanning electron microscopy (SEM).

The SEM fracture surface analysis underlines the poor fiber matrix interface: a predictable result considering the hydrophilic nature for the fiber and hydrophobic one for the matrix. This behaviour is shown both from the specimens



Graphic 2 – Stress-strain graphic comparing the mechanical properties of the composite produced with fibers from superfine grinding mill and knife mill.

with the superfine grinding mill, the k instead is for paper treated with the knife mill.

The composite obtained with the knife mill product (paper k) is more particulate, as underline before, this morphology doesn't change the elastic modulus while the mix of fibers and particles seems to contribute to the breaking load. The paper k moreover will lead to achieve higher percentages of filler thanks to its best ease of mixing.

The elastic modulus of the composite produced with fibers from the knife mill is lower then polyethylene without fibers

with the fibers of the superfine grinding mill and from the knife mill.

Thinking of paper s, we expected to obtain a better superficial grip because of the high mechanical activity on the surface of fibers during the micronizing process. Unfortunately this result wasn't found in the analysis performed.

The figures 8 and 9 represent the absence of fiber-matrix interface for specimens produced with 10% of fibers derived from the superfine grinding mill (Figure 8) and with 20% of fibers from the knife mill (Figure 9).

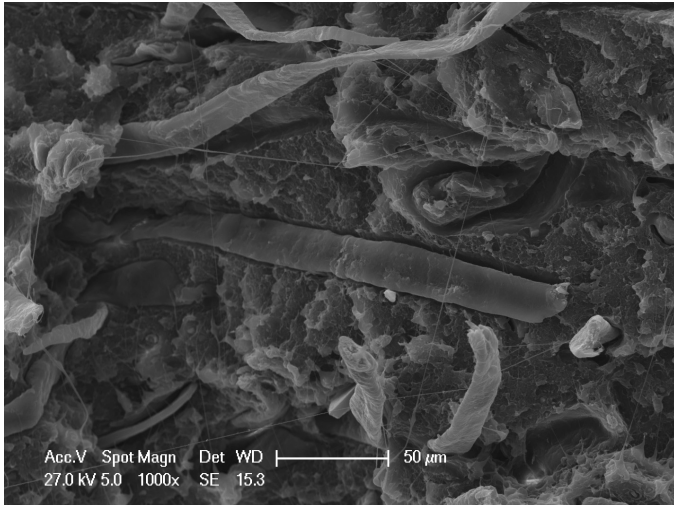


Figure 8 – SEM analysis of the specimens with 10 % of fibers S.

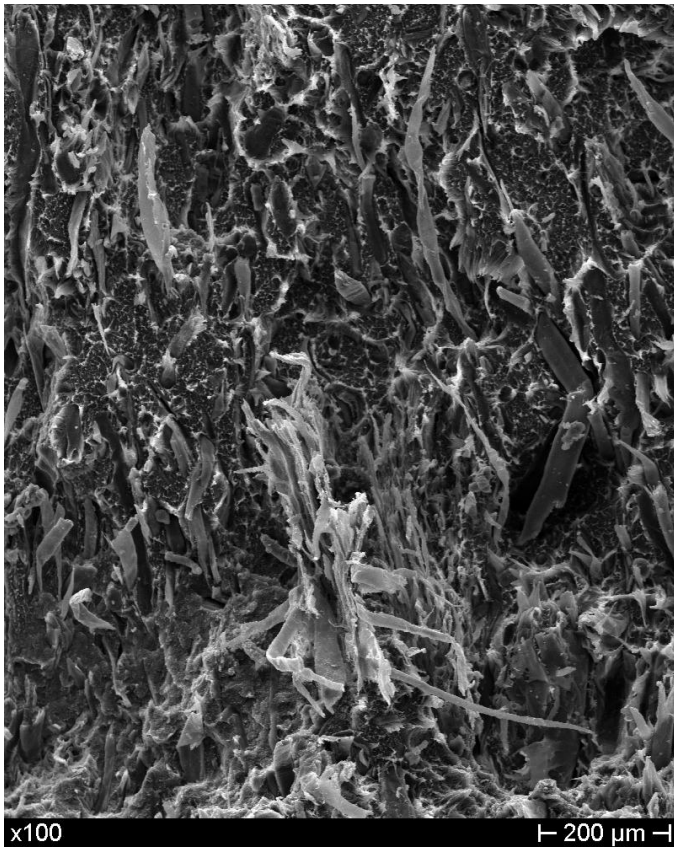


Figure 9 – SEM analysis of the specimens with 20% of fibers k.

The SEM analysis is also useful to study the fibers and particles dispersion.

A good dispersion of fibers is evident for the composite produced with fibers of superfine grinding mill with 10% and 20% of fibers. The composite with fibers derived from the knife mill is characterized by the presence of particles and isolated fibers. There are some agglomerate of fibers near the fragments. As future development the target is to achieve a better dispersion of the fibers avoiding this agglomerate.

In order to improve this result, it is necessary to introduce an additive, which would work both on better dispersion of the

fibers and on improving fiber-matrix interface. The chosen additive is polybond 3029 from Addivant, it is specifically designed for use in wood and natural fiber-filled polyethylene composites. It is a polyethylene with a very high maleic anhydride functionality [10].

The figures 10 and 11 shows the fibers dispersion in the matrix.

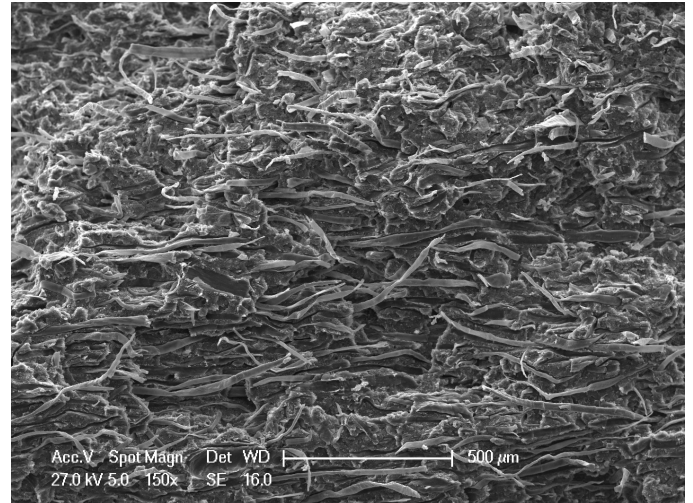


Figure 10 – SEM of the composite produced with 20% of fibers from the superfine grinding mill.



Figure 11 – SEM of the composite produced with 20% of fibers from the knife mill. A paper particle in the HDPE matrix with other paper fibers is shown.

## Conclusion

The analysis of the results obtained shows that the paper can be used as reinforce in composite with polymeric matrix thanks to encouraging tensile test data.

A different morphological product is obtained from two grinding process: both of them could be used as filler in polyethylene matrix improving different properties, for example future studies will focused on the use of knife mill output in order to improve damping and acoustic isolation.

The superfine grinding mill allows to obtain fibers as agglomerates which are more difficult to disperse homogeneously in the matrix, the knife mill instead produces a mix of fibers

and particles which allows to achieve a better fluency and higher percentages of charge introduced.

There is also the possibility of improving the quality of the product using a double step process: the first step is the turbo-mix in order to achieve the high percentage of fiber and the good dispersion, there is an intermediate grinding process, and then the second step is the injection moulding to produce the specimens.

The SEM analysis shows the very poor of the interface both with the superfine grinding mill and the knife mill. There is the possibility of interesting improvements for mechanical properties with this filler thanks to interface's modifier and mixing ceramic recycled fiber and wood flour [11].

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## References

- [1] M. R. Doshi, J. M. Dyer, Encyclopedia of Materials: Science and Technology, Paper: recycling and recycled materials, pp 6711-6720.
- [2] L. Gaines, Encyclopedia of Energy, Volume 5, Recycling of paper, pp 253-261.
- [3] [www.zanettiarturo.com](http://www.zanettiarturo.com), raccolta differenziata carta e cartone, 24 Settembre 2014.
- [4] [www.gestione-rifiuti.it/smaltimento-carta-cartone](http://www.gestione-rifiuti.it/smaltimento-carta-cartone)
- [5] [www.ecoage.it/carta-da-macero](http://www.ecoage.it/carta-da-macero)
- [6] Best Available Techniques (BAT) Reference Document for the Production of Pulp, Paper and Board.
- [7] information on [www.cimma.it](http://www.cimma.it)
- [8] information on [www.retsch.it](http://www.retsch.it)
- [9] M. Valente, J. Tirillò, A. Quitadamo, C. Santulli, "Use of recycled milled-paper in HDPE matrix composites".  
Electronic proceeding of: 5th International Conference on Innovative Natural Fibre Composites for Industrial Applications ISBN 9788890924002
- [10] W. Singworth, D.H. Roberts, compatibilizers for recycled products.
- [11] M. Valente, F. Sarasini, F. Marra, J. Tirillò, G. Pulci: "Hybrid recycled glass fiber/wood flour thermoplastic composites: Manufacturing and mechanical characterization." Composite part A (2011), 649-657.