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Production of PP Composites Reinforced with Flax and Hemp Woven Mesh Fabrics via Compression Molding

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Abstract: Hemp and flax fibers are among the most interesting vegetable fibers that can be used to reinforce polymeric matrices. In line with the global environmental requests, the use of these fibers especially coupled with thermoforming polymers are increasing more and more in order to expand their applications and replace synthetic fibers and thermosetting plastics. However, one of the major limitations of vegetable fibers is their poor adhesion with polymeric matrices that is often overcome by fibers chemical treatments or by using coupling agents within the matrix. Aiming to produce polypropylene (PP) bio composite laminates reinforced by hemp and flax fibers without additional process steps, this paper deals on the study of their production via the compression molding technique by using woven fabrics characterized by a large mesh size able to ensure a mechanical anchoring between fibers and matrix. Two different forming strategies that differ in the time required for reaching the maximum values of compression pressure and in the dwelling time at this value were used in order to investigate how the yarn impregnation was affected by them. To expand the applications of composites under investigation, tensile, bending, Izod, heat deflection temperature (HDT) and bearing tests were carried out. The results highlighted how the use of a waiting time before the reaching of the maximum moulding pressure allowed a better matrix flow within the vegetable yarn leading to higher mechanical performances.

Keywords: hemp; flax; PP; fabric; vegetable fibres; compression molding

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1. Introduction

In recent years, the rapid growth of interest in the use of natural fibers (NFs) as reinforcement for composites, due to their reduced environmental impact and low-cost, generated several research studies aimed at the study of manufacturing processes of natural fibers polymeric composites and their innovative applications.

Among polymer families, the use of thermoplastic matrices sparked an increasing interest due to their advantages over thermoset in terms of short processing time [1–4], potential recyclability and the possibility to be remodeled at high temperatures. Indeed, thermoplastic composites are used in many industrial fields, such as in automotive, naval and sports [5–8].

Among different types of natural fibers, obtained from seeds, stems or roots, flax and hemp attracted more interest because their highest mechanical properties such as specific stiffness and strength [9].

The natural fibers used as reinforcement in composite materials are generally constituted by a complex microstructure of bundles of twisted elementary fibers and, inside the latter, crystalline microfibrils embedded in an amorphous matrix (pectins, hemicelluloses and lignin) [10]; due to this structures flax and hemp fibers have excellent vibration damping properties and a nonlinear behavior when subjected to external loads.

On the other hand, the presence of the amorphous matrix that covers the cellulose microfibrils confers a hydrophilic behavior that prevents a good chemical compatibility with the hydrophobic molecules of polymers.

As known in a composite system, the adhesion plays an essential role to transfer the stress between matrix and fibers and thus affects the properties of the final composites. Poor surface adhesion due to insufficient wetting is the principal reason for the formation of a weak or ineffective interface between the fiber and the matrix. For improving the adhesion between polymeric matrix and vegetable fibers, several chemical treatments of the fibers were studied [11–13].

Alkaline and silane treatments are the most commonly used but other chemical treatments such as the isocyanate treatment, peroxide treatment, acetylation and the use of maleated coupling agents were also studied.

Alkaline treatment consists in the immersion of fibers into an alkaline solution (usually an NaOH solution), for a specific period of time. This treatment acts by increasing the fibers surface roughness that improves the mechanical bonding with the polymeric matrices, due to the facilitated mechanical interlocking [14,15]. Some alkali treatments, especially those performed at elevated temperatures, are able to selectively degrade cementing materials (i.e., lignin, pectin and hemicellulos) in natural fibers, with a little effect on the cellulose chains [16]. NaOH treatment can completely remove pectins without any residue being left in the hemp fiber also if the rate of lignin removal is dependent from the NaOH concentration [17].

During alkali treatment, a separation of the elementary fibers of the bundles occurs, then the effective surface area available for matrix bonding increases [18,19] as well as the fiber dispersion within the composite.

Silane treatment usually consists in the immersion of the fibers in a silane solution diluted in a water/alcohol mixture. The presence of water allows the hydrolysis of the silane leading to the replacement of the alkoxide groups with –OH groups and so the formation of alcohol and silanol. Therefore, silano reacts with the –OH groups of the cellulose present in the cell walls of the natural fibers by forming stable covalent bonds [13].

The adhesion between the natural fibers and polymeric matrices can also be improved by modifying the matrix with a coupling agent that adheres well to both fibers and matrix. Naturally, the choice of the coupling agent strictly depends on the type of polymeric matrix composing the composite system. Among different thermoforming polymeric matrices noteworthy for coupling with natural fibers, polypropylene (PP) is attractive for its mechanical characteristics, excellent flowability, chemical resistance, low cost and weatherability [20].

Maleic anhydride grafted polypropylene (MAPP) results to be one of the most suitable coupling agents used for polypropylene composite reinforced with natural fibers [21]. MAPP consists of long polymer chains with a maleic anhydride functional group grafted onto one end. It acts as a bridge between the non-polar polypropylene matrix and the polar fibers by ensuring a chemical bond with the cellulose fibers through the maleic anhydride groups, and bonding to the matrix by means of polymer chain entanglement.

The improvement of mechanical properties after the addition of MAPP is well described in literature [21–25]. However, both the fiber treatments and the addition of MAPP to the polymer provide further production steps that sometimes can limit the use of long fibers or fabrics. In addition, their use can also cause the increasing of the manufacturing process time as well as the costs and decreasing the environmental benefits connected with the use of NFs. Indeed, in most research, treatments are conducted on short fibers and using screw extruder processes to blend MAPP polymer and fibers.

In addition to chemical treatments, something else can facilitate the interaction between matrix and fibers: the main idea is to consider what has been performed in some engineering applications to improve the mechanical interaction between incompatible materials, i.e., the use of meshes. This solution is very common in the building construction field [26–28]. The mesh can indeed guarantee a mechanical interlocking inside the matrix, this solution is successfully utilized in some metal foam [29] and building construction works [28,30].

Therefore, the aim of this work is to study the possibility to use woven mesh natural fiber fabrics (characterized by a large mesh size) of flax and hemp to reinforce polypropylene (PP) matrix. The idea is then to use these fabrics that can act as a mesh and then can be anchored the matrix without the use of fibers or matrix treatments. In addition, the use of fabrics with large mesh size can allow an easier and more uniform distribution of the polymer matrix around the fabric yarn than that achievable with common fabrics.

The use of thermoplastic matrices and fabrics, instead of single fibers, makes the production of composites by compression molding technique very attractive, then the study of suitable production strategies represents a research key aspect. Indeed, natural fiber fabrics are different from synthetic ones, so the production strategies have to be optimized and furthermore studied with respect to the well-known strategies adopted for synthetic fibres [31].

Due to the above said limitations, it is very common to find research works where short natural fibers are used together with treated PP [21–25].

Very few works aimed on the use of fabrics are available, one of these is the work of Dobah et al. [32]. They studied the properties of flax fabrics/polypropylene (PP) composites produced by compression moulding using special commingled fabrics. Different thermoforming manufacturing parameters, in particular different values of temperature (in the range of 175–200 °C), dwelling time (ranging between 40–1280 s) and molding pressure (in the range of 2.7 and 4.7 MPa) were considered to evaluate their effects on the overall mechanical performances.

In the present work, hemp/PP and flax/PP composites were produced by using woven mesh natural fiber fabrics via compression molding techniques without any additional chemical treatment. Two different forming strategies that mainly differ in the time required to reach the maximum values of compression pressure and in the dwelling time at this value were considered. Aiming to expand the application of flax/PP and hemp/PP composites, in addition to usual mechanical characterization, i.e., tensile, flexural and impact properties, also heat deflection temperature (HDT) and bearing tests were carried out.

2. Materials and Methods

2.1. Materials

Woven hemp and flax fabrics (provided by Fidia Srl, San Mariano di Corciano, Italy) characterized by a high value of areal density, 360 and 320 g/m^2 respectively, were used as reinforcement for composites under investigation. Fabrics were safely stored in polymeric bags under vacuum in a store at 20 °C and 45% of humidity level; fabrics were not subjected to any chemical or surface treatments before their use.

Figure 1 shows the fabric (Figure 1a) and a magnification of a single yarn (Figure 1b) constituted by the winding of filaments, with a twist angle equal to about 20°; in turn, each filament consists of several elementary fibers. For the composite matrix, films of neat PP with a thickness of 40 µm, were used.

2.2. Samples Production

Before sample's production, all fabrics were dried in an oven at 60 °C for 12 h to eliminate any trace of humidity. Composite laminates (200 mm × 150 mm) were produced using the film-stacking procedure where layers of fabrics and PP were piled up alternately as shown schematically in Figure 2a. Five layers of PP films and four fiber fabrics with a 0/90 lay-up were used. The PP samples reference (200 mm × 150 mm) were produced by controlling the thickness to avoid an excessive flash production and then an excessive thickness reduction during the molding process, for this purpose a control thickness of 3.00 mm was adopted. All sample types were produced with a conventional compression molding press.

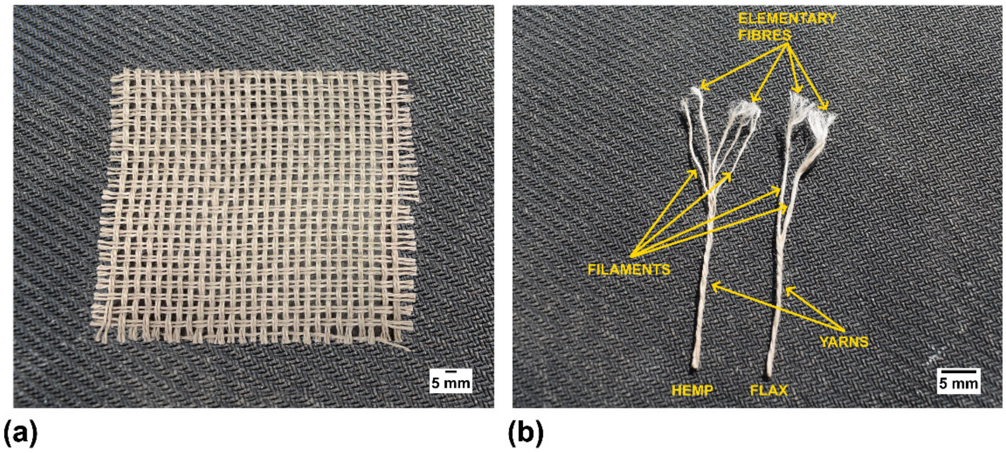


Figure 1. (**a**) Fabric adopted as reinforcement and (**b**) magnification of hemp and flax yarn.

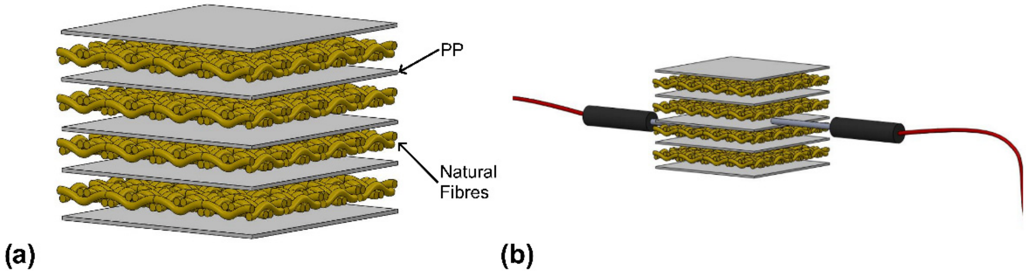


Figure 2. (**a**) Stacking sequence of composite samples and (**b**) position of thermocouples.

The molding temperature was fixed at 200 °C, the pressure at about 4 MPa and the total molding time at 300 s, considering the studies cited in the introduction [29]. Two molding strategies are considered:

(i) The maximum pressure value of 4 MPa was reached after a waiting time of 120 s, before it the molding plates were closed without compressing the laminate;
(ii) The maximum pressure value of 4 MPa was immediately applied to the laminate.

For choosing the waiting time of (i), the molding process was conducted on testing laminates having two thermocouples (Type K supplied by T.M. ELECTRONICS LTD, W. Sussex, UK) close to the laminate middle plane (Figure 2b). The waiting time was chosen as the time required to reach a temperature of around 200 °C (i.e., the mold plate temperature) within the testing laminate where the thermocouples were placed.

The second strategy (ii) was considered in order to further simplify the molding process without any control of the waiting time, the only requirement is that the molding plate have to be at the molding temperature (200 °C) before the process starts.

For each configuration, three laminates (200 mm × 150 mm) were produced. In Table 1, the mean values of laminate thickness, volumetric fiber percentage, and other details for each sample type are listed.

Table 1. Main properties of samples, mean values and standard deviations (between brackets).

Label	Reinforcement Fabric	Stacking Sequence	Molding Strategy	Thickness [mm]	Fibre Volume Percentage [%]
PP	-	-	Waiting time 0 s	2.98 (0.12)	-
H1	Hemp	$[(0/90)_4]$	Waiting time 120 s	3.28 (0.15)	30.5 (0.5)
F1	Flax	$[(0/90)_4]$	Waiting time 120 s	2.98 (0.21)	31.7 (0.7)
H2	Hemp	$[(0/90)_4]$	Waiting time 0 s	3.15 (0.13)	31.8 (0.8)
F2	Flax	$[(0/90)_4]$	Waiting time 0 s	2.70 (0.23)	33.9 (0.4)

2.3. Experimental Procedure

A confocal microscope (Sensofar, Terassa, Spain) was used to investigate the yarns impregnation and their distribution. For each laminate type, three metallographic specimens were mounted in a proper epoxy resin and polished with grinding discs.

The mechanical performances of bio-composite under investigation were studied in terms of: (i) tensile, (ii) flexural, (iii) impact, (iv) bearing and (v) HDT resistance. In detail, tensile and three point bending tests were performed according to ASTM D3039 and D790 standard respectively using a universal testing machine (MTS Aliance RT 50) Torino, Italy) equipped with a 5 kN load cell and an extensometer MTS 634.31F-24 (for evaluating the strain in the case of tensile tests). Both tensile and bending tests were carried out on five specimens for each sample configuration.

In the bending tests, the span was changed according to the actual measured thickness keeping the span-to depth ratio equal to 32.

Izod impact tests were performed to evaluate the capability of reinforced and unreinforced PP to absorb energy. The tests were carried out according to ASTM D256 on notched samples (100 mm $\times$ 12 mm with 2 mm V notch at 45 °C) using a pendulum impact instrument (CEAST resilimpactor, Pianezza, Italy). The tests were carried out at room temperature and five specimens for each category were tested, so the mean value of the absorbed energy obtained was calculated.

Additionally, to this conventional mechanical characterization, to further expand the application fields of the natural fiber composites (NFCs) under investigation, other types of tests can be considered. For instance, it can be taken into account the possibility to produce some parts that can be jointed to other parts, for example by mechanical connections in drilled holes.

Therefore, in order to evaluate the effects of the woven mesh reinforcement into the matrix on the hole mechanical resistance, bearing tests were performed.

As known, three typical joint failure modes of composite exist: net-tension, shear out and bearing modes [33]. Net-tension failure is catastrophic; it occurs when the specimen width to-hole diameter ratio (W/D) is small, therefore an optimal W/D ratio should be selected to avoid this type of failure. Increasing the W/D ratio, shear-out failure occurs, it is a special case of bearing failure. When the W/D ratio is large enough, bearing failure occurs leading to an elongation of the hole. Bearing failure is progressive and less likely to cause serious problems than net-tension and shear-out failure mode [34].

Hao et al. [35] suggested a minimum value of W/D ratio equal to 3 for better analyzing the damage due to the presence of a hole.

In the present paper, W/D values of 6 and 3 were adopted for analyzing the behavior of the composite when the bearing occurs. Hole diameter equal to 5 mm was considered and five samples for each sample configuration were tested.

In line with the aim to expand the applications of PP reinforced by hemp or flax, it has to be considered that in several applications, polymeric composites can be exposed to different types of stresses at different temperatures during their life, then to study the viscoelastic behavior of these materials appears to be necessary. Therefore, for evaluating the influence of the woven mesh reinforcement on the mobility of the polymeric matrix, HDT tests were carried out by using a TA Instruments Rheometric Series RSA III in a flexural loading system configuration. HDT test is a suitable test to evaluate the physical performance of a polymer (or reinforced polymers) under load and elevated temperature. The HDT data allow us to detect the maximum service temperature without large deflection. Indeed, HDT measurements allow the determination of the temperatures at which a specimen undergoes a pre-set deflection under a specific load during heating. The experiments were carried out at a defined heating rate of 20 °C/min following the ASTM E 2092 standard. The stress applied for HDT measurement was 0.45 MPa and the pre-set deflection was fixed at 0.34 mm. Five specimens for each sample type were tested.

At the end of each set of tests, one-way ANOVA tests were performed in order to determine significant differences among all samples.

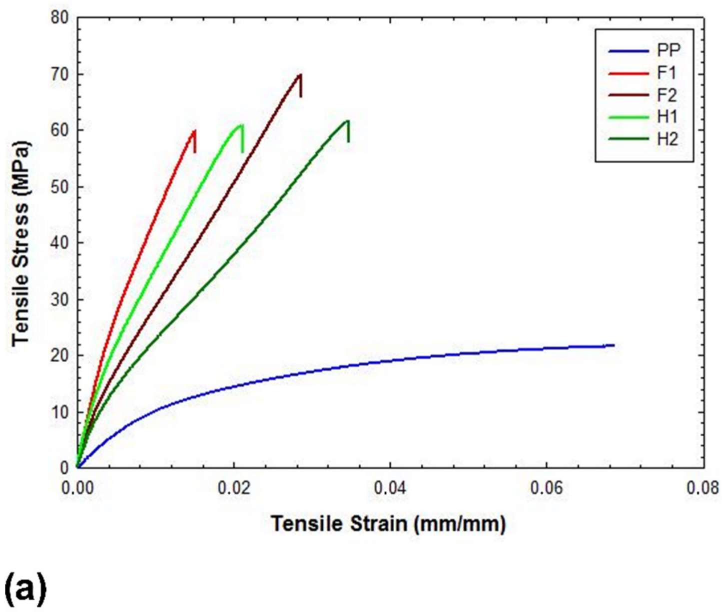
3. Results and Discussion

Tensile, bending and Izod impact tests were performed in order to determine the overall effects of the presence of the fiber reinforcement on mechanical performances with respect to the unreinforced PP samples evaluating how the molding strategy affected them.

Once the molding strategy that maximizes these performances for each fiber type was selected, bearing and HDT tests were performed on these composite categories in order to evaluate how the yarn and the mesh that characterizes the fabrics are able to anchor the polymeric matrix.

3.1. Tensile, Bending and Izod Tests

In Figure 3a, typical tensile stress-strain curves of all specimens were plotted. First of all, it is possible to note the significant improvement of both the tensile strength and the tensile modulus conferred by the reinforcement respect to the only matrix.



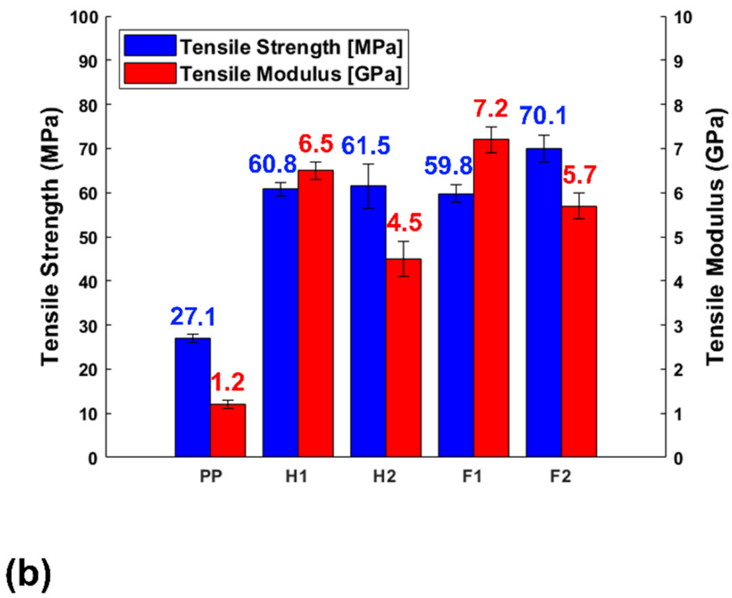


Figure 3. (**a**) Typical tensile stress–strain curves carried out from tensile tests for each sample type and (**b**) mean values of tensile strength and modulus.

From Figure 3a clearly appeared the typical bilinear behavior of bast fiber composite materials correlated with the intrinsic strain behavior of elementary fibers caused by the elementary fibers' kink band defects and micro fibril angle rotation inherited mainly from extraction and weaving methods of the fibers.

Looking at Figure 3b, it appears that by comparing samples produced using the same molding strategy, the use of flax allowed to reach a higher value of the elastic modulus than samples reinforced with hemp fibers. Indeed, F1 sample type has a mean elastic modulus (7.2 GPa) around 10% higher than H1 (6.5 GPa) and F2 sample type shows a mean elastic modulus (5.7 GPa) around 26% higher than H2 (4.5 GPa).

The significant differences among samples produced with the same molding strategy were clearly evident and confirmed looking at Table 2 in which the *p*-values carried out from the ANOVA tests were listed. Samples produced with a different molding strategy or samples produced using a different reinforcement showed elastic tensile modules statistically different (p-value < 0.05).

Table 2. *p*-values carried out from one-way ANOVA test on the results of tensile, bending and impact tests. Method: null hypothesis; significance level: $\alpha = 0.05$. σ_{ts}: tensile strength; E_t: tensile modulus; σ_{fs}: flexural strength; E_f: flexural modulus; U: specific adsorbed energy.

Condition	Comparison	*p*-Value of E_t vs. Sample Type	*p*-Value of σ_{ts} vs. Sample Type	*p*-Value of E_f vs. Sample Type	*p*-Value of σ_{fs} vs. Sample Type	*p*-Value of U vs. Sample Type
Different molding strategy and same material	H1–H2	0	0.747	0	0.029	0.491
Different molding strategy and same material	F1–F2	0	0	0.002	0.295	0.048
Different material and same molding strategy	H1–F1	0.003	0.416	0.175	0.028	0.101
Different material and same molding strategy	H2–F2	0.001	0.004	0.033	0.007	0.092

Higher tensile modulus reached by flax can be justified considering the better adhesion and impregnation of flax with the PP polymer than hemp fibers; looking at the confocal micrograph images of Figure 4, it appears that: differently from what happen for hemp yarn (Figure 4b,c), in the case of flax fibers the PP polymer was able to impregnate also the yarn core (Figure 4d,e); this deeper yarn impregnation allowed to reach higher elastic modulus.

The flax composites produced by adopting the first molding strategy (F1, i.e., with the presence of a waiting time) allowed to reach higher value of elastic modulus (7.2 GPa, +500% than PP) than F2 due to the better impregnation of the yarn as clearly evident by comparing Figure 4d,e.

On the other hand, F1 samples are characterized by a lower tensile strength (59.8 MPa) in comparison with F2 (70.1 MPa). Table 2 highlights the significant statistic differences among the tensile strengths of these two sample types. This strength reduction (almost 14.7%) was justified by considering the higher value of F1 sample thickness, it was +9.4% thicker than F2 sample.

Indeed, the load at breaking is conferred by the fibers content that is exactly the same for both F1 and F2 typologies, so the tensile strength (given by the ratio between the load at breaking and the cross-section area) is higher for F2 since its thickness is lower.

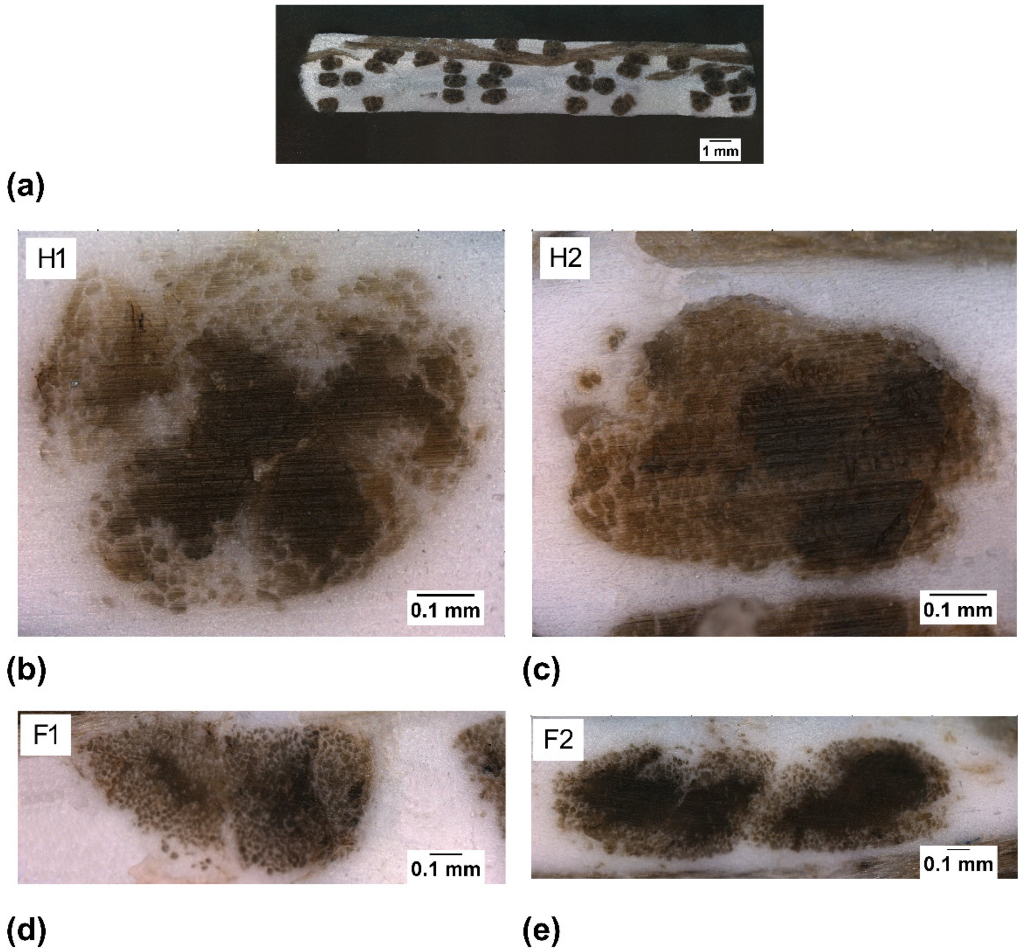


Figure 4. (**a**) Typical cross section of H 1 sample, (**b**) 10× image magnification of typical yarn of H1, (**c**) H2, (**d**) F1 and (**e**) F2 samples.

Considering hemp as reinforcement and looking at Figure 3b and Table 2, it is possible to observe that, as for flax, the use of a waiting time during the molding process (i.e., H1 sample type) allowed to obtain composites with higher tensile modulus (+44.5% if compared to H2) thanks to the better yarn impregnation (see Figure 4a,b). Contrary to what occurred for flax, the tensile strength of H1 and H2 were very close to each other since the thickness of these sample types was almost the same. Indeed, looking at Table 2, no significant statistical differences were detected by comparing the tensile strengths of H1 and H2 samples (p-value = 0.747). The reason is that the hemp fibers are less deformable than flax fibers, so there were not observed significant thickness variation between H1 and H2 (see Table 1). This is also testified by no significant yarn deformation differences between these two hemp sample types (see Figure 4b,c).

Summarizing, even though the total molding time was the same (300 s), when the molding load was immediately applied (second molding strategy), the flax or hemp fibers were immediately compressed and the flow of molded PP within the yarn was hindered. This means that in this case, PP flow finds the molding plane as a preferential way involving the production of PP edge flash, then a reduction of samples thickness and so an increase of the fiber volumetric fraction content was obtained (see Table 1). Looking at Figure 4, it is clear how the presence of PP within the yarn was more evident when a waiting molding time was adopted (i.e., Figure 4b,d).

In Figure 5, typical stress-strain curves of all sample typologies obtained from bending tests are plotted and a typical image of the top surface of a representative sample is reported. From the bending tests appeared that all samples showed compression failure, and looking at Figure 5b appears that in any cases the presence of fibers improved the bending performances than PP (between +90.9% and +181.8% for the modulus in case of F2 and H1 respectively and +34.8% and +63.4% in flexural strength in case of H2 and F1 respectively).

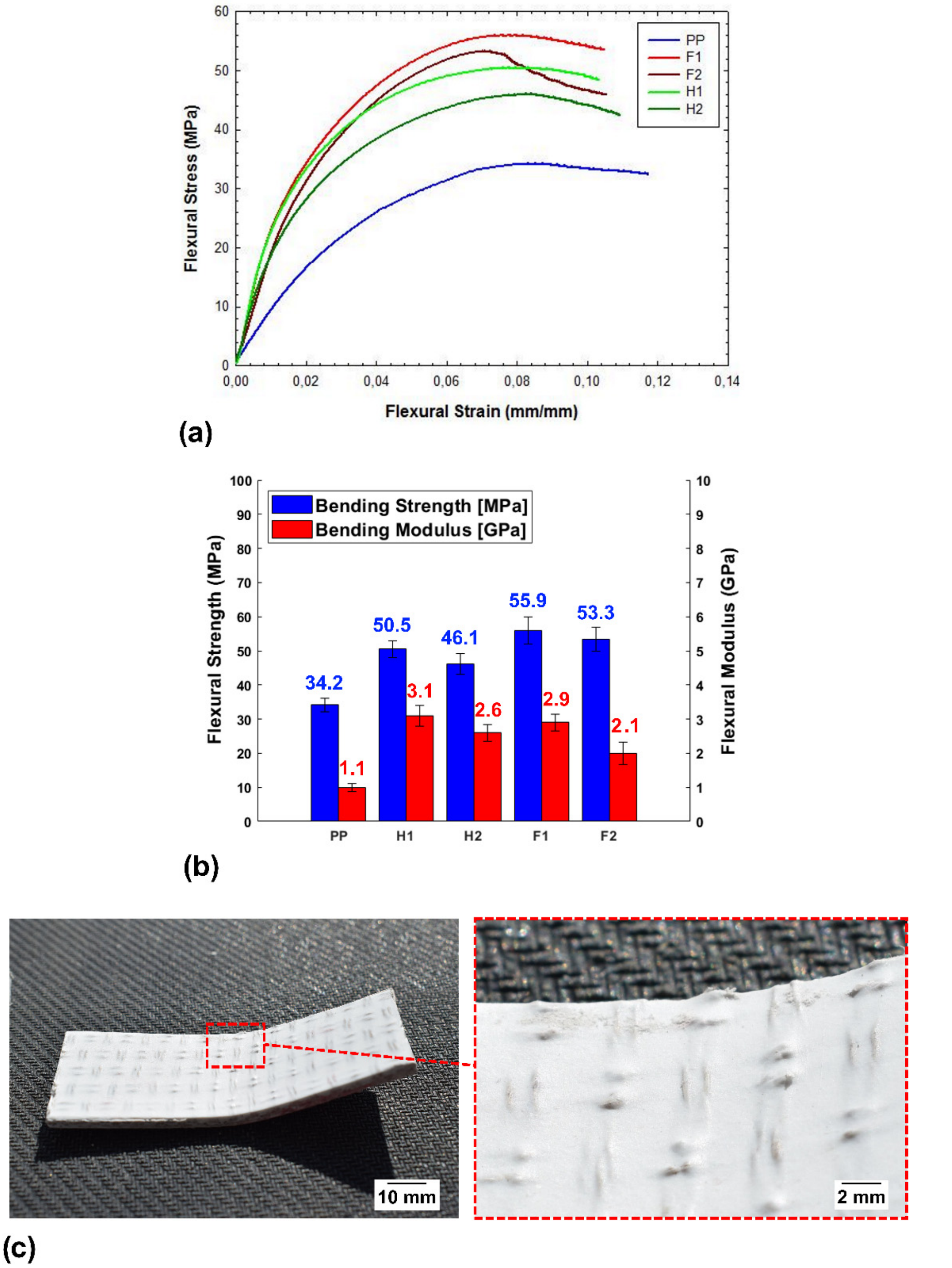


Figure 5. (**a**) Typical flexural stress-strain curves carried out from bending tests for each sample type, (**b**) their mean values of strength and modulus and (**c**) H1 sample at the end of the test.

On the compression side of all specimens (see Figure 5c), it was observed a compression instability of the fibers that emerged from the surface. This phenomenon was more evident for samples produced according to the second molding strategy (without waiting time, i.e., H2 and F2 sample types). This occurred because of, as explained before, during the molding process without waiting time, PP easily flows in the molding plane then samples produced according to the second molding strategy showed a lower amount of matrix that covers the fibers on the external side surfaces. An effect of this phenomenon is the lower value of the flexural modulus reached by the above said sample types (i.e., those produced with the second molding strategy). The significant statistical differences that proved the lower flexural modulus of H2 than H1 and of F2 than F1 were also confirmed looking at the p-values (<0.05) in Table 2.

Regarding the flexural strength, looking at Figure 5a,b and at Table 2, it appears that independently from the molding strategy (so comparing samples produced with the same molding strategy in Table 2), it was observed a significant higher value of flexural strength of samples reinforced by flax fibers (i.e., F1 and F2) than those reinforced by hemp (i.e., H1 and H2), due to a better adhesion between flax and PP.

Comparing flexural and tensile strength of the same sample type, it is clear that the first one appeared to be lower than tensile (see Figures 3b and 5b). Usually, in composite systems the flexural strength should be higher than tensile due to the beneficial contribution of the side subjected to compression loads during the bending: higher is the in-plane compression modulus and strength of the specimen, higher the contribution of the compression side should be. In the cases under investigation, the resistance offered by the compression side (constituted by few NF yarns) is low, so the flexural resistance is lower than tensile. This aspect was more evident for composite produced according to the second molding strategy (i.e., without waiting time) especially those reinforced with hemp fibers (H2 sample type, see Figure 5b).

Looking at Figure 5b and Table 2 and comparing the molding strategies, it appeared that the bending strength of H1 and H2 showed significant differences (p-value < 0.05), differently from the differences between F1 and F2 (p-values > 0.05). Even though the first molding strategy allowed the reaching of a higher mean bending strength, the effect was significant for hemp fibers only. This is because of the hemp fibers are less deformable than flax fibers, then the use of the second molding strategy implied a more evident presence of fibers on the external side surfaces that did not work well during compression.

Interesting results were also achieved from the Izod impact tests. Firstly, it was observed that all sample typologies reached the failure, then all the impact energy was absorbed during the impact event. From the tests, it was clearly revealed that the presence of natural fibers in PP matrix, led to a mean improvement of the in-plane impact properties of around six times the one of unreinforced PP (see Figure 6a), this is due to additional energy dissipation mechanisms such as debonding and fiber slipping [36–39]. At this scope, in Figure 6b PP and H1 broken specimens are reported.

Focusing the attention on H1, it is possible to observe the typical brittle failure surface, as well as the PP sample, with the presence of fiber pull-out and broken fibers. This was detected for all reinforced samples.

Differently from the other tests, looking at Figure 6b and Table 2 it is possible to note that the Izod tests did not revealed a significant difference between flax and hemp or between the molding strategies. This because of the adopted fabrics had a large mesh size then, considering the samples size, few yarns participated in the conferring of the impact resistance. This aspect together with the fact that (differently from previous quasi static tests), the impact test velocity was high and also considering that the energy dissipation includes different mechanisms, involved high values of standard deviation, so the differences among reinforced samples were not so significant.

The only significant differences were detected for flax fibers, as for tensile and bending tests, the adoption of a waiting time during the molding process, positively affected the final properties, indeed F1 showed a significant higher adsorbed energy than F2 (see Table 2), it showed an improvement of around 6.5 times than PP.

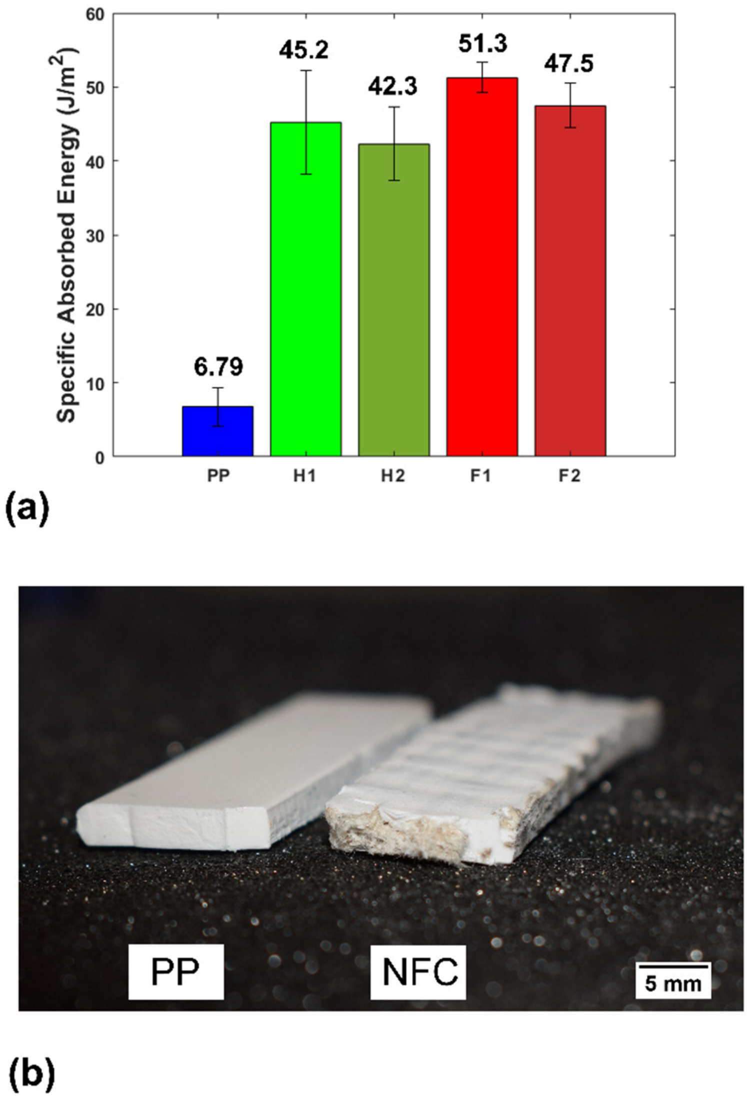


Figure 6. (**a**) Specific adsorbed energy for each sample type and (**b**) example of sample cross section at the end of the Izod tests.

It is difficult to compare tensile, flexural and impact resistance obtained in the present investigation (see Table 3) with those reported in literature because most of the works concerning PP/natural fibers composites focused the attention on the use of short fibers. For example, from the review of Vigneshwaran et al. [40] or in the work of Beckermann et al. [41] appeared that treated hemp or flax composites (30–40 wt.% of fibers) reinforcing treated PP possess lower tensile, flexural and impact resistance than those produced in the present investigation.

Comparable tensile and flexural performances were instead achieved in [32], however in this case commingled fabrics (flax/PP) were adopted, this results in an additional step for the manufacturing of starting reinforcing materials.

3.2. Bearing and HDTU Tests

As discussed in previous section, the presence of a waiting time in the molding process allowed to obtain samples with higher tensile modulus, flexural and impact performances due to the better impregnation of the yarn, so bearing and HDTU tests were carried out on these sample types only (i.e., H1 and F1).

Bearing tests were conducted on samples with two different values of W/D ratio equal to 3 and 6.

Table 3. Main tensile, flexural and impact properties of samples, mean values and standard deviations (between brackets). σ_{ts}: tensile strength; $\%\sigma_{ts}$: percentage increase of σ_{ts} than PP; E_t: tensile modulus; $\%E_t$: percentage increase of E_t than PP; σ_{fs}: flexural strength; $\%\sigma_{fs}$: percentage increase of σ_{fs} than PP; E_f: flexural modulus; $\%E_f$: percentage increase of E_f than PP; U: specific adsorbed energy; %U: percentage increases of U than PP.

Sample Type	Tensile Test				Bending Test				Izod Test	
	σ_{ts} [MPa]	$\%\sigma_{ts}$ [%]	E_t [GPa]	$\%E_t$ [%]	σ_{fs} [MPa]	$\%\sigma_{ts}$ [%]	E_f [GPa]	$\%E_f$ [%]	U [J/m^2]	%U [%]
PP	27.1 (1.1)	-	1.2 (0.1)	-	34.2 (2.1)	-	1.1 (0.1)	-	6.8 (2.6)	-
H1	60.8 (1.5)	+125.3	6.5 (0.2)	+441.7	50.5 (2.5)	+47.7	3.1 (0.3)	+181.8	45.2 (7.1)	+565.7
H2	61.5 (4.4)	+126.9	4.5 (0.4)	+275.0	46.1 (2.8)	+34.8	2.6 (0.2)	+136.4	42.3 (5.2)	+523.0
F1	59.8 (2.1)	+120.7	7.2 (0.3)	+500.0	55.9 (3.7)	+63.4	2.9 (0.2)	+163.6	51.3 (1.9)	+655.5
F2	70.1 (2.6)	+158.7	5.7 (0.3)	+375.0	53.3 (3.5)	+55.8	2.1 (0.3)	+90.9	47.5 (3.1)	+599.6

In Figure 7a are reported the typical bearing stress-pin displacement curves for reinforced and unreinforced samples under investigation when W/D = 6 was adopted. The bearing stress σ_b, was calculated by dividing the load recorded during the test by the respective loaded area, so according to Equation (1):

$$\sigma_b = \frac{F}{d \cdot t}, \quad (1)$$

where, F is the measured load, d the pin diameter and t its thickness. Looking at Figure 7b, it appears that flax fibers were able to manifest the best bearing resistance thanks to its better adhesion with the polymeric matrix (+47.2% than PP). Anyway, also the use of hemp reinforcement allowed to improve the bearing resistance with respect to pure PP (+24.5%). Comparing the bearing strength of PP, F1 and H1 for W/D = 6, the ANOVA tests showed a p-value equal to zero, then their differences were significant.

The presence of stress peaks in the curves of Figure 7a are due to the breaking of single yarn by the pin; these peaks are more evident in hemp reinforced samples due to the lower adhesion with the matrix of these fibers than flax.

The whole breaking area is characterized by an evident fiber pull out, it can be assumed that the yarns were firstly pulled and then broken. This aspect is highlighted in Figure 8a, where the bearing failure is clearly evident.

The breaking mode is different when a W/D equal to 3 was adopted, in this case firstly bearing and then net tension failure mode were observed. This is highlighted in Figure 8b where H1 tested samples with W/D equal to 6 and 3 are compared. In Figure 9a, the stress-displacement curves for each sample type tested with W/D = 3 is plotted and the mean results are reported in Figure 9b. Comparing the bearing strength of PP, F1 and H1 for W/D = 3, the ANOVA tests showed a p-value equal to zero, then their differences were significant. It appears that:

- The bearing strength (43.2 MPa and 51.3 MPa for H1 and F1 respectively) is lower than those detected in tensile tests (60.8 MPa and 59.8 MPa for H1 and F1 respectively), this means that in any case bearing is generated;
- The bearing strength is lower than that identified in the bearing tests with W/D equal to 6 (75.8 MPa and 90.8 MPa for H1 and F1 respectively). This is due to a shorter length of the yarns arranged close to the hole and orthogonally to the pull direction.

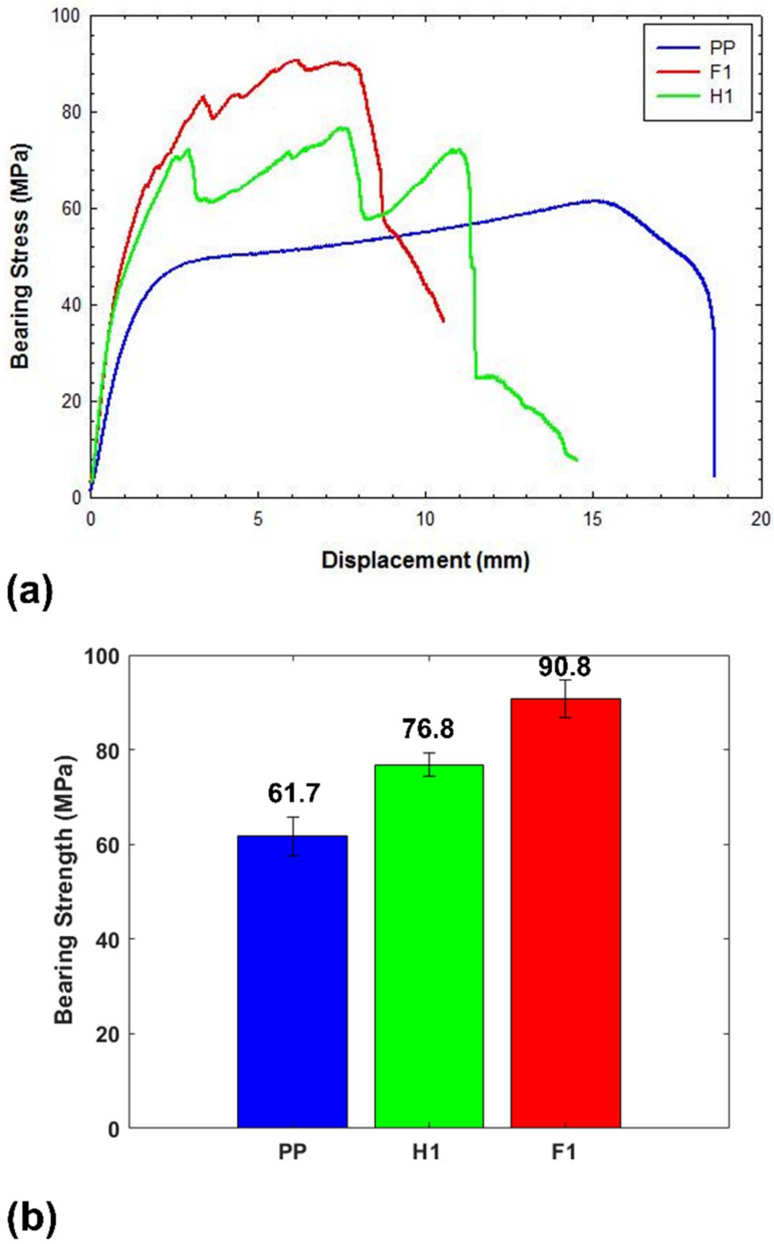


Figure 7. (**a**) Typical bearing stress-displacement curves and (**b**) bearing stress strength for specimens with W/D = 6.

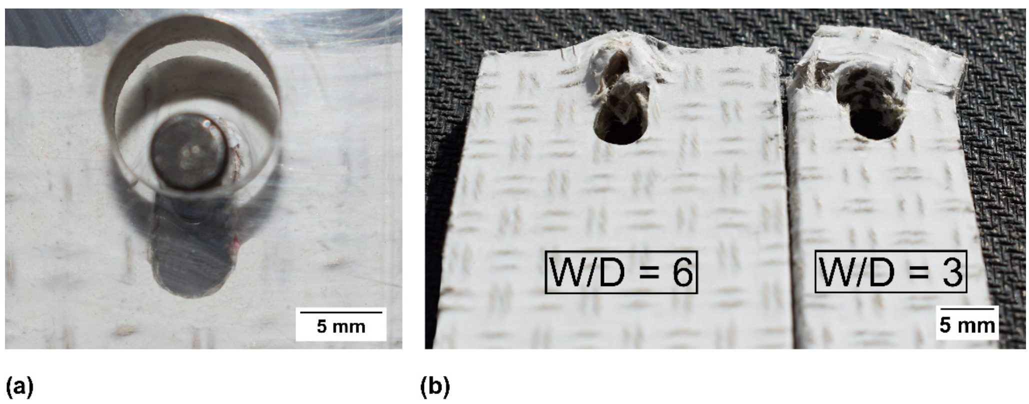


Figure 8. (**a**) Magnification of area close to the hole in the bearing tests with W/D = 6 of H1 sample type and (**b**) comparison of H1 sample tested with W/D equal to 6 and 3.

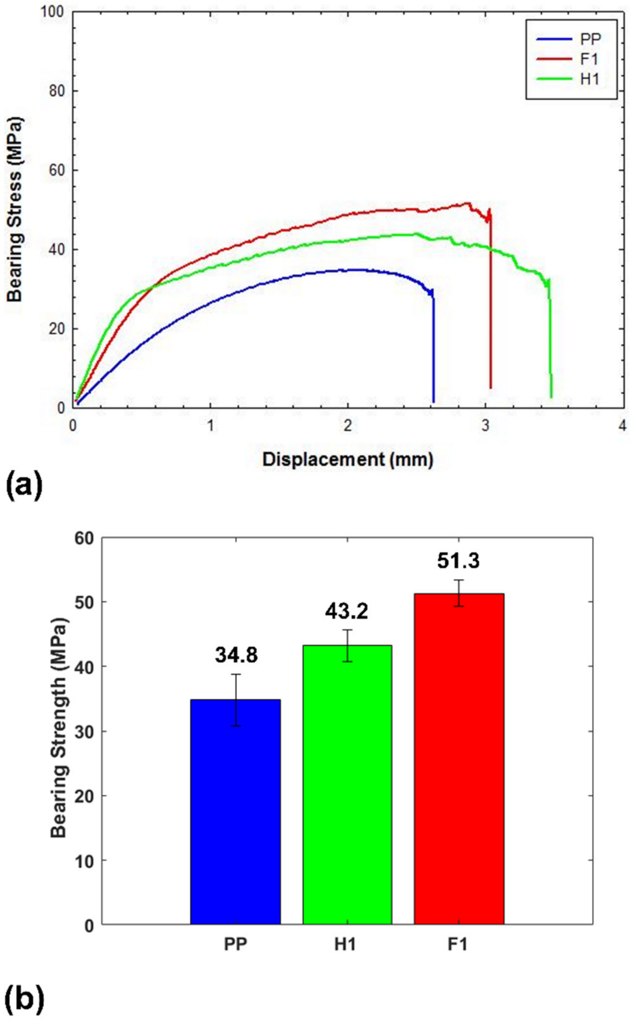


Figure 9. (**a**) Typical bearing stress-displacement curves and (**b**) bearing stress strength for specimens with W/D =3.

Therefore, after a first movement of yarns orthogonally disposed to the load direction, it was observed a failure of the yarns parallel to the load, whose resistance was influenced by the movement of the first ones and by a partial debonding.

In any case, also for the bearing properties there was still a better behavior of the composite compared to the unreinforced PP. Mean and standard deviation of bearing strengths are listed in Table 4.

Table 4. Bearing strength, mean values and standard deviations (between brackets) of PP, H1 and F1 sample type.

Sample Type	Bearing Strength, W/D = 6 [MPa]	Percentage Increase of Bearing Strength than PP, W/D = 6 [%]	Bearing Strength, W/D = 3 [MPa]	Percentage Increase of Bear-ing Strength than PP, W/D = 3 [%]
PP	61.7 (4.1)	-	34.8 (4.2)	-
H1	76.8 (2.5)	24.5	43.2 (2.3)	24.4
F1	90.8 (4.1)	47.2	51.3 (2.1)	47.4

The results of HDT tests are reported in Figure 10 where it is possible to detect the temperature able to involve a strain of 0.2% (about 2 mm in displacement) when 0.45 MPa of stress was applied.

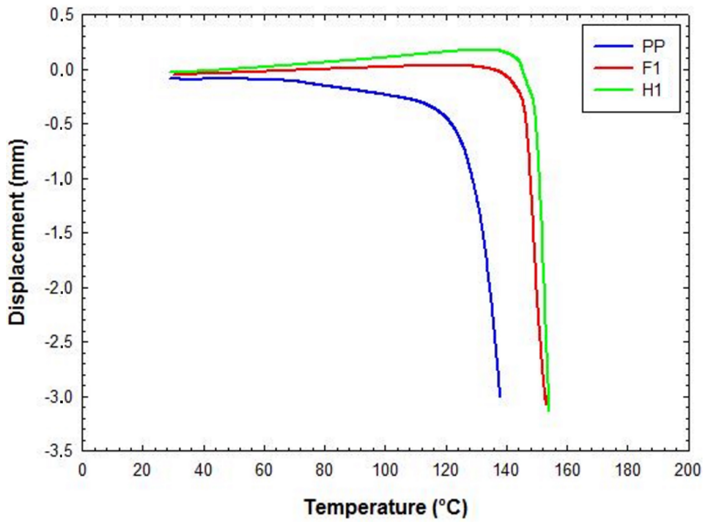


Figure 10. Displacement vs temperature curves carried out from HDT tests.

Comparing reinforced and unreinforced sample typologies, it is possible to note that the presence of the reinforcement leads to a mean improvement of this temperature of almost 15 °C (±1). This aspect can be mainly attributed to the action of the mesh of the fabric that was able to trap the polymer and block the scrolling of the single plies.

Therefore, the mobility of the polymer chains was hindered by the presence of the mesh fabrics. Such restriction on the polymer chain mobility allowed the improvement of the upper service temperature or HDT of the resulting composites.

All curves show a positive increase of the displacement when the temperature increases, this is due to the volumetric expansion of both the matrix and vegetable fibers with the temperature; this aspect is more evident for reinforced samples due to their higher rigidity than unreinforced PP.

No significant differences in the behavior of composites reinforced with different fibers were observed, this aspect indicated that in the investigated temperature conditions, the effect of the geometry of the reinforcement layers was more predominant than the fibers/matrix adhesion.

4. Conclusions

In this paper, hemp and flax fibers in forms of woven mesh fabrics are used to reinforce PP matrix via compression molding technique.

The use of woven mesh fabrics characterized by large mesh size was able to act similar to a containing mesh guaranteeing a mechanical anchoring with the polymeric matrix without any additional process steps, similar to the use of a chemical treatment or a coupling agent.

Two molding strategies were considered that mainly differ in the presence of a waiting time before the application of the molding pressure, it was proved that without waiting time the flowing of molded PP within the yarn was hindered then the PP flow find the molding plane as preferential way, involving the reduction of samples thickness.

This phenomenon negatively affects tensile and flexural modulus and the impact resistances. Otherwise, the use of the waiting time before the application of the molding pressure lead in a better yarn impregnation allowing to reach increases of around +500%; +164%; +655% than unreinforced PP respectively for tensile modulus, bending modulus and specific adsorbed energy when flax fibers were used. Independently from the molding strategy, tensile and bending strength of reinforced samples were higher than only PP.

In bearing tests, the advantage of using the mesh as reinforcement increases as the ratio W/D increases due to the positive effects of the length of the transversal yarn. The higher bearing properties were obtained from flax fibers due to the better fiber/matrix adhesion.

The HDT tests revealed that the mobility of the polymer chains was hindered by the presence of the mesh fabrics. Such restriction on the polymer chain mobility improved the upper service temperature of almost 15 °C independently from the fiber nature.

Since the obtained performances appeared to be interesting considering the non-use of any chemical treatments, future next studies could aim on the study of the drapability of the proposed woven mesh fabrics and their use for the production of complex shape products via hot forming processes.

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