FLAX WASTE MANAGEMENT USING BIODEGRADABLE POLYMER MATRICES

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In memory of
Acad. Bogdan C. Simionescu

The ongoing development of biodegradable polymer production technologies appears to be a logical step toward reducing the ever-growing piles of plastic waste. Commercially available biodegradable polymers are still expensive and, unfortunately, are not yet a viable alternative to traditional petroleum-based polymers. However, it should be noted that this is slowly beginning to change. One way to reduce the cost of biodegradable polymers is by using additives, such as short cellulose fibers derived from local agricultural crops, which are often by-products of other processes. Of course, to ensure that the newly developed biodegradable composites remain competitive on the market, it is essential to guarantee that their physical and mechanical properties are not significantly diminished. The aim of the research was to produce biodegradable composites based on polymer Mater-Bi with added flax fibers and to determine their mechanical and tensile properties. The melt flow rate (MFR) was also specified for selected composites.

Keywords: Mater-Bi, natural fibers, biodegradation, biodegradable plastic, flax, composites

INTRODUCTION

Growing environmental concerns, driven by the massive increase in the use of plastics, have accelerated the ecological crisis and forced current generations of scientists to search for biodegradable alternatives. Such materials and their composites with renewable reinforcements have attracted the attention of scientists due to their pro-ecological properties.[1] However, the degradation of these composites after use remains a significant challenge. In this context, biodegradable polymers and their composites can play a key role in reducing the overall carbon footprint and plastic waste. Moreover, some leading biodegradable polymers and their composites exhibit excellent mechanical, thermal, and thermomechanical properties, comparable to traditional thermoplastics used in food packaging, medicine, and the automotive industry.[2-4] As a result, biodegradable polymers are experiencing annual production growth of 20%, and significant sales growth is expected by the end of this decade.

Due to the depletion of petrochemical and carbochemical raw materials, as well as economic, political and ecological factors (environmental pollution, especially during chemical coal processing), more and more attention is being paid to natural renewable raw materials, such as polysaccharides, proteins, and fats. The European Union plans a 300% increase in chemical industry production by 2040.[5] This requires a search for new sources of raw materials, as existing resources may prove insufficient to achieve this goal. Increasingly stringent environmental regulations further exacerbate the situation. This has sparked an energetic search for new materials. Plants are considered as raw materials for the chemical industry. Interest in plants as raw materials stems from their availability, chemical composition, renewable nature (especially relevant in the case of annual plants), and biodegradability. They accumulate significant amounts of renewable raw materials in biomass as an alternative to obtaining biodegradable materials. Some of these are widely

used as foodstuffs, and each of these components also has non-food applications. Available domestic resources of polysaccharide raw materials can be utilized, even without extensive processing, adapting them for various purposes through physicochemical, physical, chemical, and enzymatic modifications. An example of such applications is biodegradable plastics.[6-9]

The primary goal of developing fiber-reinforced composites is to achieve materials that exhibit both high strength and an elevated elastic modulus. Reinforcing fibers utilized in these composites can be classified as either synthetic or natural.[10-15] Common synthetic fibers include glass, carbon, aramid, and silicon carbide,[16-23] with glass fiber being the most prevalent due to its cost-effectiveness and excellent strength-to-weight ratio, although it is somewhat less stiff compared to other reinforcing fibers.[24-26] The resins incorporated in reinforced polymer composites can be either thermoplastic or thermosetting.[27-30]

To lessen the environmental footprint of composite materials, there's a growing interest in using natural fibers and bio-based matrices as alternatives to conventional, synthetic reinforcements and petrochemical-based matrices. Recent reviews highlight the extensive research being conducted on the production methods and mechanical properties of these eco-friendly, biodegradable composites made from renewable materials.[31] In fact, the global capacity of bio-based plastics was projected to surge nearly tenfold by 2024 compared to 2010, with starch-based plastics leading the charge in production volume. When it comes to natural plant fibers, bast fibers stand out for their impressive potential to provide reinforcement, thanks to their excellent specific axial mechanical properties that result from a high crystalline cellulose content and a low microfibril angle relative to the fiber axis.[32]

Since ancient civilizations, natural fibers have been used to make ropes, strings, and fabrics, playing an important role in social life. Today, due to their biodegradability, renewable nature, carbon neutrality, and environmental friendliness, they are gaining increasing importance as a raw material for modern material applications.[33] Unlike synthetic fibers, natural fibers are lightweight, low-density, easy to obtain and process, and relatively inexpensive. Their advantages also include reduced wear and tear on machinery during processing and no negative impact on soil and aquatic environments.

The availability of fibers in a given geographic region directly influences their use in composites. In Europe, flax and cereal straw are most commonly used, while in Asia, hemp, cotton, jute, sisal, and rice husk fibers predominate. The popularity of specific fiber types is also related to the scale of their global production – jute, hemp, and flax hold the largest market share, which translates into their widespread use in composites reinforced with natural fibers.

Flax fiber is the strongest of all natural fibers. It is a renewable and biodegradable raw material. In recent years, interest in flax fiber-reinforced plastic composites has increased significantly due to its high strength, stiffness, damage resistance, low density, and biodegradability. Compared to E-glass fiber, flax fiber exhibits a higher specific tensile strength. Various research groups have conducted work on the physical and chemical surface modification of flax fibers. These methods include alkaline treatment, corona discharge, maleic anhydride grafting, aminopropyltriethoxysilane treatment, interfiber polymerization to remove water from the fibers, and other methods.[34]

In general, natural fibers are characterized by low density, renewable nature, and are cheaper than glass fibers. Although their strength is lower than that of synthetic fibers, they outperform synthetic fibers in applications where low weight and stiffness are essential. Due to their biodegradability, renewable nature, and environmental friendliness, natural fibers are an excellent alternative for many applications.

Composites made from Mater-Bi combined with biodegradable fibers, especially plant cellulose, have been successfully created. Incorporating flax cellulose pulp with Mater-Bi enhances both mechanical strength and thermal stability.[35] Moreover, adding sisal fibers in amounts ranging from 5 to 20% significantly improves the composite's resistance to crack initiation and fracture progression compared to the pure matrix. Interestingly, the material's biodegradability is evident, as just a month of soil burial of the composite leads to a decrease in mechanical properties because of damage at the fiber/matrix interface.[36]

EXPERIMENTAL

Materials

Mater-Bi – HF03-A2 (specific gravity = 1.26 g/cm^3, MFR = 2.5 g/10min at 160 °C/5.0 kg) was purchased from Novamont, Italy. Flax fibers – of a

length 2, 4 and 6 mm (specific gravity = 1.5 g/cm^3) were obtained from Ekotex, Poland.

Methods

Mater-Bi and flax fibers were dried prior to processing in order to avoid hydrolytic degradation during the processing. Matrices were melt mixed at three weight ratios: 100/0, 99/1, 98/2. The composites were prepared using Polydrive (HAAKE, USA) by melt mixing for 9 min with a mixing speed of 60 rev/min at a temperature of 140 °C. Plates were prepared using a LabTech LP-20B hydraulic press, with platen pressure of 50 bars and a temperature of 140 °C.

Tensile test samples (type 1 according to ISO 527-2:2012) were made using a micro injection molding machine produced by Proma, Poland.

Melt flow rate was measured with a specific weight of 2.16 kg at the temperature of 150 °C. The testing device was a Melt Flow Junior (Ceast, Italy) in accordance with ISO 1133 standard.

Tensile properties were evaluated using an LR10K LLOYD testing machine, at a speed of 50 mm/min.

Impact test were evaluated using a Dart Tester (Ceast, Italy) in accordance with ISO 7765 standard.

The composition and notation of the samples are presented in Table 1.

Table 1
Notations and compositions of the studied composites

Composite	Filler	Filler content (%)	Length of fibers (mm)
Mater-Bi	-	0	0
Mater-Bi + 10% F 2mm	Flax	10	2
Mater-Bi + 20% F 2mm	Flax	20	2
Mater-Bi + 10% F 4mm	Flax	10	4
Mater-Bi + 20% F 4mm	Flax	20	4
Mater-Bi + 10% F 6mm	Flax	10	6
Mater-Bi + 20% F 6mm	Flax	20	6

RESULTS AND DISCUSSION

Melt flow rate

During the tests, it was observed that, probably due to the fiber length, it was impossible to test some of the experimental materials under the assumed conditions. Consequently, tests were not performed for the following samples: Mater-Bi + 10% F 6mm, Mater-Bi + 20% F 6mm. The results of the MFR test are presented in Table 2 and Figure 1.

The results showed that the higher the fiber content and/or length in the composite, the lower the melt flow rate of the tested material. This is due to the presence of a filler in the liquid medium, which does not melt.

Table 2
Melt flow rate results

Composite	Load (kg)	Temperature (°C)	MFR (g/10 min)
Mater-Bi	2.16		3.16
Mater-Bi + 10% F 2mm	2.16		1.41
Mater-Bi + 20% F 2mm	2.16	150	0.31
Mater-Bi + 10% F 4mm	2.16		1.37
Mater-Bi + 20% F 4mm	2.16		0.30

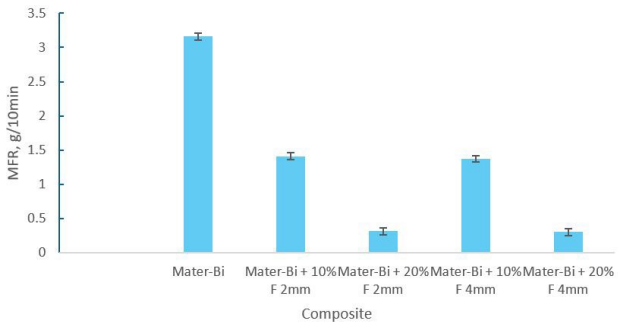

Figure 1: MFR of composites

A relationship can be observed: the addition of any filler reduces the melt flow rate (MFR) by 2.2-2.6 times, compared to the pure Mater-Bi. Increasing the fiber content in the composite from 10% to 20% for fibers of the same length causes a decrease in the MFR value.

Tensile properties

Tensile strength was tested based on the ISO 527 standard. This test allowed determining composite parameters such as Young's modulus, maximum stress at break, stress at maximum load, and elongation at break. The tensile strength test results are included in Table 3. The results are also presented as graphs in Figures 2-5.

A strength test allowed assessing the effect of flax fiber content and length on the strength properties of biodegradable composites. The presence of fibers in the composite increases its stiffness and reduces the elongation at break. This is due to the stiffening of the structure and the restriction of the mobility of the polymer chains. The highest modulus of elasticity was obtained for the Mater-Bi composite with 20% flax fibers, 4 mm long (642.11 MPa).

Table 3
Evolution of mechanical properties of composite materials

Composite	Young's modulus (MPa)	Stress at break (MPa)	Stress at maximum load (MPa)	Percentage strain at break (%)
Mater Bi	215.75	12.82	13.76	119.95
Mater-Bi + 10% F 2mm	346.86	11.28	13.50	26.70
Mater-Bi + 20% F 2mm	598.73	15.22	15.72	11.75
Mater-Bi + 10% F 4mm	322.05	10.99	11.93	24.31
Mater-Bi + 20% F 4mm	642.11	15.25	15.68	11.88
Mater-Bi + 10% F 6mm	296.32	10.49	11.37	23.87
Mater-Bi + 20% F 6mm	580.43	14.52	14.72	11.76

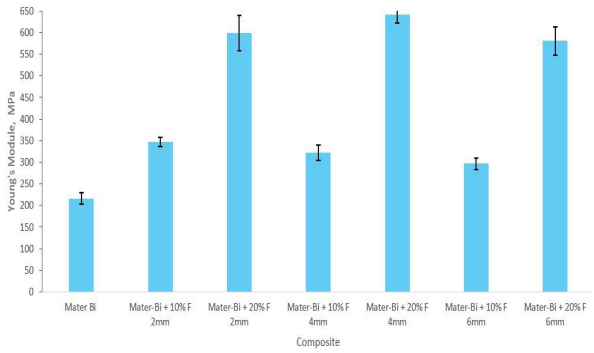

Figure 2: Young's modulus of composites

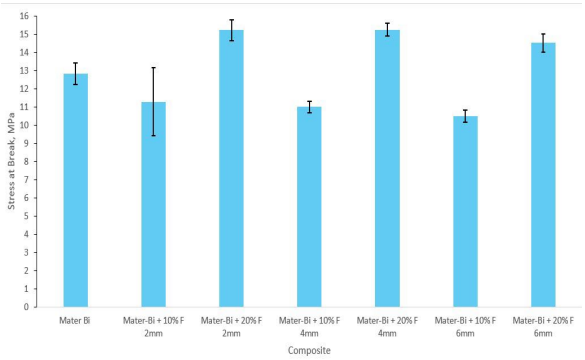

Figure 3: Stress at break of composites

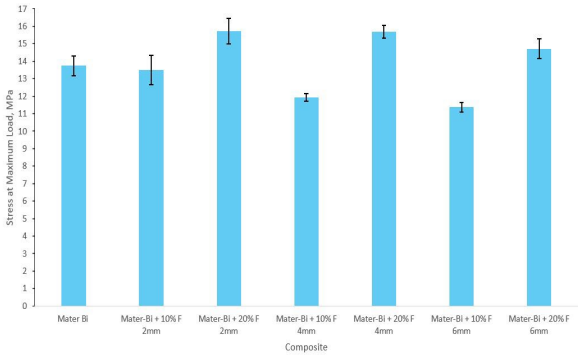

Figure 4: Stress at maximum load of composites

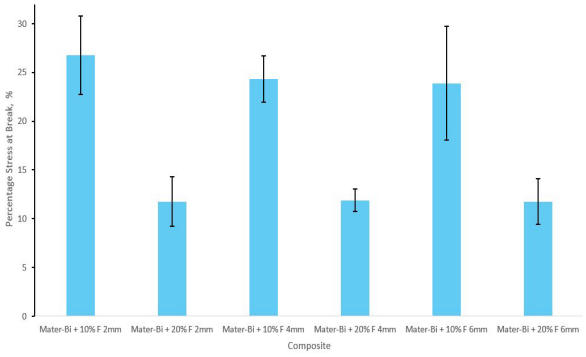

Figure 5: Percentage stress at break of composites

The addition of natural fibers significantly increased the material's stiffness, for the composite with the addition of 20% fibers – an almost three-fold increase. It was found that the fiber length is of secondary importance – the percentage of fibers is more important. The highest stress at break was achieved by composites with 20% fiber content (approximately 15 MPa), suggesting good strength. However, it is worth noting that increasing fibers does not always improve strength – with weak adhesion between the fiber and the matrix, the structure may be weakened.

Table 4
Evolution of mechanical properties of composite materials – bending

Composite	Young's modulus of bending (MPa)	Maximum deflection (mm)	Maximum bending stress at maximum load (MPa)
Mater Bi	956.19	23.22	10.48
Mater-Bi + 10% F 2mm	598.95	22.83	13.79
Mater-Bi + 20% F 2mm	697.96	22.46	17.58
Mater-Bi + 10% F 4mm	360.02	23.23	11.93
Mater-Bi + 20% F 4mm	759.05	18.32	18.43
Mater-Bi + 10% F 6mm	618.97	22.69	13.32
Mater-Bi + 20% F 6mm	572.55	22.35	16.23

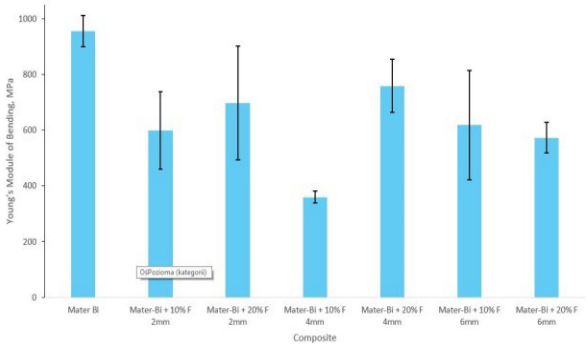

Figure 6: Young's modulus of bending results of composites

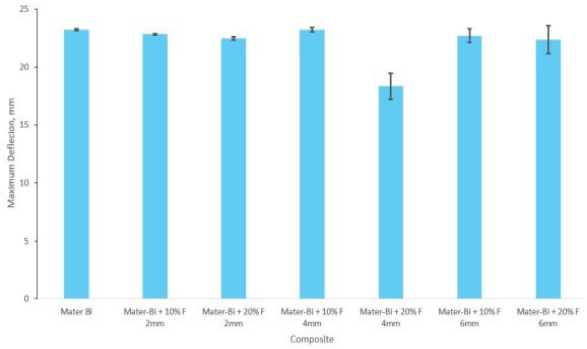

Figure 7: Maximum deflection results of composites

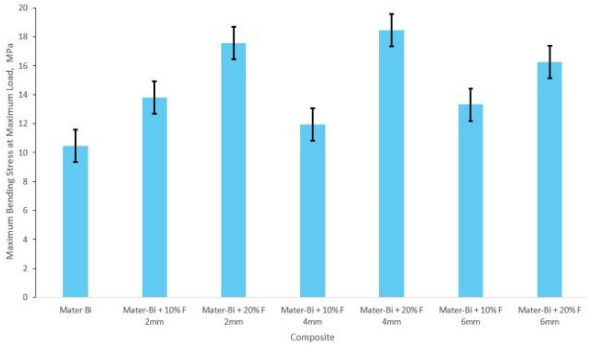

Figure 8: Maximum bending stress at maximum load results of composites

Bending strength

Flexural strength was tested based on BS EN ISO 14125:1998. This test allowed determining composite parameters such as Young's modulus, maximum deflection, and maximum bending stress at maximum load for flax fiber-filled composites. The samples were obtained by injection molding. The flexural strength test results are included in Table 4 and Figures 6-8.

The highest elastic modulus was obtained for pure Mater-Bi (956.19 MPa). The addition of natural fibers resulted in a decrease in Young's modulus in most cases, which may be due to suboptimal interphase adhesion and the presence of micro-voids and structural defects. The

exception is the sample with 20% F 4 mm, where E = 759 MPa, which may be due to better structural integration or more uniform fiber distribution. In terms of bending stress values, all composites with added fibers outperformed pure Mater-Bi (10.48 MPa). The best result was obtained for Mater-Bi + 20% F 4 mm (18.43 MPa), suggesting that this fiber ratio and length achieved a compromise between effective reinforcement and processability. Similar results were observed for the 20% F 2 mm (17.57 MPa) and 20% F 6 mm (16.23 MPa) versions. Maximum deflection did not differ significantly among the composites, ranging from 18.3 to 23.2 mm. Only the 20% F 4 mm sample stood out, demonstrating the lowest deflection value (18.32 mm), which may indicate increased stiffness and reduced formability. The addition of fibers did not significantly affect the deflection value (differences of less than 5% relative to each other). However, the noticeable reduction in deflection at 20% F 4 mm (18.32 mm) may indicate increased stiffness and reduced deformability.

Impact strength

The test was conducted in accordance with ISO 7765. This test examines the ability of a material to withstand the impact of a falling object, helping to determine whether the material will break, tear, or absorb the impact. The test results are included in Table 5 and presented in Figures 9-11.

Composites with added fibers showed a significant decrease in impact strength compared to pure Mater-Bi (787.13 J/m). The highest impact strength values among the composites were achieved for: Mater-Bi + 10% F 2 mm (188.52 J/m) and Mater-Bi + 20% F 4 mm (183.93 J/m). The lowest results were obtained for: Mater-Bi + 20% F 2 mm (72.94 J/m) and Mater-Bi + 20% F 6 mm (78.88 J/m). It can be argued that shorter fibers (2 mm and 4 mm) promote better force distribution within the structure, allowing for partial impact energy absorption. Longer fibers (6 mm) can lead to a more non-uniform force distribution, reducing energy absorption capacity. Pure Mater-Bi exhibits by far the best impact resistance, as seen both in the test results and empirically by observing the sample after the test. When the composites were pulled apart, pure Mater-Bi stretched and tore. Higher fiber content reduces impact resistance, which may mean that excessive fiber concentration can lead to structural defects.

Table 5
Impact strength of composite materials

Composite	Peak force (N)	Energy at peak (J)	Resilience (J/m)
Mater-Bi	57.44	0.66	787.13
Mater-Bi + 10% F 2mm	44.55	0.20	188.52
Mater-Bi + 20% F 2mm	25.11	0.06	72.94
Mater-Bi + 10% F 4mm	41.81	0.20	161.58
Mater-Bi + 20% F 4mm	27.13	0.08	183.93
Mater-Bi + 10% F 6mm	36.35	0.14	122.94
Mater-Bi + 20% F 6mm	29.60	0.07	78.88

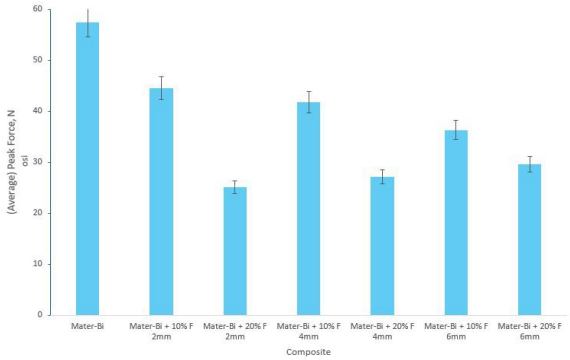

Figure 9: Peak force results for composites

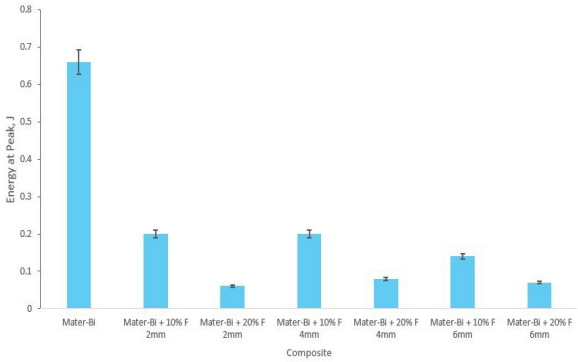

Figure 10: Energy at peak results for composites

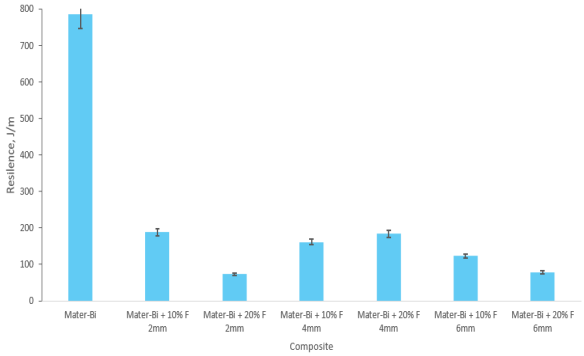

Figure 11: Resilience results for composites

In the case of polymer composites, the addition of short fibers can lead to a change in the fracture behavior. Instead of the brittle fracture characteristic of ceramic materials, composites with short fibers exhibit more plastic behavior, which may be desirable in some applications.

CONCLUSION

Biodegradable composites based on the Mater-Bi polymer with short flax fibers demonstrate improvements in some mechanical properties compared to the pure polymer. It can be seen that the composites exhibit similar strength properties when comparing composites with the same fiber content – 10% and 20%, which can be considered a more important factor than the fiber length in the composite.

The addition of plant fibers to the composite contributed to an increase in tensile strength – the composite became stiffer and less flexible. The composite's tensile strength decreased with increasing fiber content.

The fiber content in the composites affected the thermoplasticity of the composite. The melt flow rate (MFR) decreased with higher fiber content. Furthermore, the longer the fibers, the lower the melt flow rate. The presence of fibers in the composite increased its stiffness and reduced the elongation at break. The addition of short flax fibers increased the flexural strength of Mater-Bi composites, but had a negligible effect on deflection.

Pure Mater-Bi exhibited significantly higher impact resistance. Higher fiber content reduced impact strength. In the case of polymer composites, the addition of short fibers can lead to a change in fracture behavior. Instead of the brittle fracture characteristic of ceramic materials, composites with short fibers exhibited more plastic behavior, which may be desirable in some applications.

Mater-Bi blended with plant fibers loses its characteristic thermoplastic properties and elasticity, although its strength properties are improved. Along with the stiffening of the composite, this opens up possibilities for the production of biodegradable packaging, containers, flowerpots, and disposable cutlery, including plates and cups.

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