[1.]Teodor Gabriel FODOREAN, [1.]Elena Mihaela NAGY, [1.]Nicolae CIOICA, [2.]Lucian FECHETE, [1.]Valentin VLADUT

RESEARCH REGARDING THE POSIBILIETIES OF USING NATURAL FIBERS AND WOOL FOR FABRICATION OF BIOCOMPOSITE MATERIALS

[1.]INMA Bucharest, ROMANIA
[2.]UT Cluj Napoca, Facultatea de Autovehicole, Mecatronica si Inginerie Mecanica, ROMANIA

Abstract: Biocomposite materials are a family of new materials that differentiate from the classic composite materials by the fact that are partially or completely obtained from biogenic raw materials and are biodegradable in natural environments. Products from plastic and composite materials became important components of the human's life in the last 60 – 70 years. Their main disadvantage consists is that they are not biodegradable, creating big environment issues. Biocomposite materials start to be recognized as a positive investment for the chemical industry, offering various opportunities and presenting a real interest for all industrial and social sectors. In present it exists the trend of continuing the research on the already known directions, but in the same time we have the horizon of some new researches in other domains of possible applications, that were not touched till now. The evolution of researches in the field of biocomposite materials conduct to the development of new bio materials and bio fabrication technologies, with applicability in agriculture, agriculture equipment's manufacturing, packaging industry, biomedicine, passengers and public transportation vehicles industry, civil construction industry etc. The researchers performed in this paper were orientated on the study of the possibilities of fabrication by extrusion of a few blends using biopolymers, natural fibers or wool. As raw materials have been used starch, PBAT biopolymer (ecoflex F BX 7011 is an oil–based, biodegradable polymer designed for film extrusion and extrusion coating, produced by BASF) and natural fibers as miscanthus, hemp or sheep wool. The mechanical properties of the new obtained biocomposite materials were also evaluated.
Keywords: biocomposite materials, sustainability, starch, natural fibres, wool

INTRODUCTION

Biocomposite materials are biodegradable materials obtained from biogenic raw materials originating from agricultural/natural resources.

Natural fibers are biogenic raw materials that are more and more introduced in many industries during the last two decades. Through the most used natural fibers in industry are hemp, curacao, mischantus, cocos etc. The most viable renewable resources used in the development and fabrication of biocomposite materials are: starch, cellulose, sugar or sucrose, casein, chitin and chitosan, gelatine, vegetable oils, protein from cereals, seaweeds, wool, hemp fibers, miscanthus fibers etc.

In Figure 1 is presented the ideal life cycle of renewable row materials that can be used at the biocombisite materials production.

As evolution of appearance of bioplastic and biocomposite materials we point that in 1869 Hyatt brothers opened the first cellulose factory, in 1923 was started the mass production of cellophane, in 1941 Henry Ford was uncover the fist car with plastic body made from biogenic row materials, in 1988 was discovered termoplastic starch (being after one of the most commune used raw material for obtaining the new bioplastic and biocomposite materials).

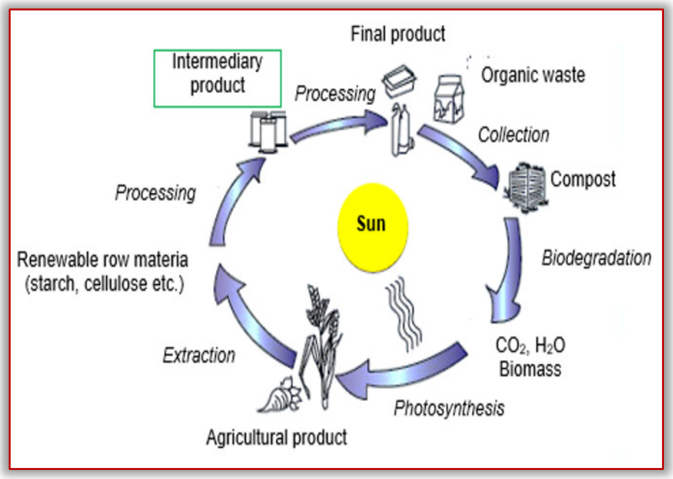

Figure 1 – Loop of the ideal life cycle of renewable raw materials in the case of their use for biocomposite materials production.

The global volume of plastic materials production is 390.7 million tones. From this 98.5% are fossil–bases, recyclable 8.3%, non–recyclable 91.7%. From this only 5.86 million tones (1.5%) are fabricated based on renewable resources.

The ideal solution to follow will be, to increase the percentage of biodegradable plastic and composite materials in correlation with creation of new muti–component completely biodegradable composites, with dedicated performance characteristics.

In the same time are regions worldwide with big production of sheeps. One of the resulting raw materials of this business is the sheep wool. The climatic conditions and the industry was passing to some changes in the past 30 – 40 years. Based on this are regions as Transylvania, in Romania, where is produced a lot of wool and is not used in a big proportion in textile industry resulting many wool residues. Our goal is to use this wool residues fabrication of biocomposite materials from one side. From other side, we are also looking to other opportunities where is possible to use this wool residues.

In the context described above we was developing and testing two recipes:
— Starch – PBAT – miscanthus fibers,
— Starch – PBAT – sheep wool.

For the above two bleeds we was trying to establish the influence of blending conditions (thermal regime, rotation speed, residence time) to the mechanical properties of the new obtained composite material. In the same time we establish will be optimized and established the best processing conditions in order to obtain the expected mechanical proprieties of the new obtained blank biobased composite material.

MATERIALS AND METHODS

The materials used in this experiment was alimentary starch, PBAT (ecoflex F BX 7011) produced by BASF and PA (platamid) produced by ARKEMA.

The starch used was having a density of 1.78 g/cm³.

Table 1. Chemical composition of starch depending of the source of provenience

Chemical composition	Wheat	Potato	Rice	Corn
Lipids (% dry substance)	0.8	0.05	0.8	0.77
Protein (% dry substance)	0.4	0.06	0.45	0.38
Ash (% of dry substance)	0.15	0.4	0.5	0.06
Phosphorus (% dry substance)	0.06	0.08	0.1	0.02
Amylose %	26–31	23	14–32	28
Amylopectin %	72	79	83	67–69

Souce: Banks and Greenwood 1975:263; Blanshard 1987:17; Swinkels 1985:25,27

Ecoflex F BX 7011 comes closer than any other biodegradable plastic to the processing properties of a classic polymer. A flexible plastic designed for film extrusion and extrusion coating. Blown film extrusion is a particular area where PBAT shows well–balanced processing properties and the resin can be used in extrusion coating applications.

The characteristics of PBAT (ecoflex F BX 7011) used, supplied by BASF are shown in table 2. In Figure 2 are presented some row granules of PBAT.

Table 1. PBAT (ecoflex F BX 7011) material proprieties

Property	Value	Test Method
Density (g/cc)	1.25 to 1.27	ASTM D792
Melt Index, g/10min (190°C/2.16Kg)	2.7 to 4.9	ASTM D1238
Melting Point, °C	110 to 120	DSC
Tensile Strength, (MPa)	34	ASTM D638
Elongation, %	700	ISO 527
Water Permeation Rate, g/(m²*d)	140	DIN 53122


Figure 2 – PBAT raw granules.

In our bled we was using also natural fibers – miscanthus and sheep wool.

The miscanthus fibers (figure 3) was imported from South America and the sheep wool (figure 4) was obtained from sheep farms located in Fagaras, Transylvania region, Romania. The density of miscanthus fibers was 0.7–1 g/cm³. The sheep wool was having a density of 3.45 g/cm³.


Figure 3 – Miscanthus fibers


Figure 4 – Sheep wool

Processing was carried out using a Brabender mixer (figure 5) equipped with a 30 cm³ mixing chamber and two counter–rotating rotors. The mixing took place for 10 minutes, at a temperature of 140°C and having a rotors speed of 60 rpm. The amount of biopolymer used in the starch matrix (60%) was 30% PBAT in (mass/volumetric) percentage, miscanthus fibers and wool being incorporated in 10%. The materials were dried before processing at 75°C for 5 hours.

Two experiments/recipes were conducted:

— **Experiment 1** – Starch 60% – PBAT 40% – Wool 10% – temperature 140°C, mixing time 10 min, rotation speed 60 rpm,

Figure 5 – Brabender internal mixer 30 cm^3

— **Experiment 2** – Starch 60% – PBAT 40% – Miscanthus 10% – temperature 140°C, mixing time 10 min, rotation speed 60 rpm.

RESULTS

After the evacuation of the materials from the mixing chamber, they were inserted inside a Carver 4394 press, heated to 140°C, pre–melted for 2 minutes and then pressed in the form of plates for 2 minutes, the pressure used being 200 bars. After pressing, cooling the mold and obtaining plates with a thickness of 1 mm (± 0.05 mm) for tensile testing and bars with dimensions 1 mm x 4 mm x 80 mm for shock tests. The material was inspected to observe the dispersion of the fibers and subsequently prepared for mechanical tests. Good dispersion was observed in all cases.

Tensile test – the characteristics evaluated in the tensile tests (Young's modulus, tensile strength, elongation at break) were recorded according to the SR EN ISO 527–2/2012 standard, using an Instron 3345 machine, with a test speed of 10 mm/min, the distance between the clamps being 40 mm. All samples were initially conditioned at 23°C and 50% relative humidity.

In figure 6 we have presented the tensile strength curve for the three different processed mixtures made following the biocomposite material recipe with miscanthus (starch 60% – PBAT 30% – 10% miscanthus, extrusion at 140°C, 60 rpm, time 10 min.).

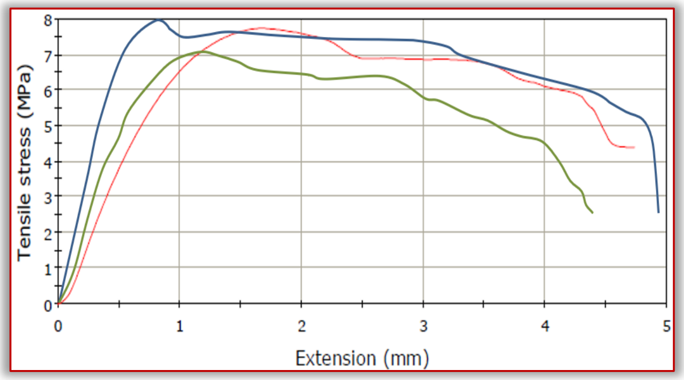

Figure 6 – Tensile strength curve for starch mixture 60%–PBAT30%–10%miscanthus, extrusion at 140°C, 60 rpm, time 10 min. Red curve – mix 1, blue curve – mix 2, green curve – mix 3.

In figure 7 are presented the tensile strength curve for the three different processed mixtures made following the biocomposite material recipe with sheep wool (starch 60% – PBAT 30% – 10% wool, extrusion at 140°C, 60 rpm, time 10 min.).

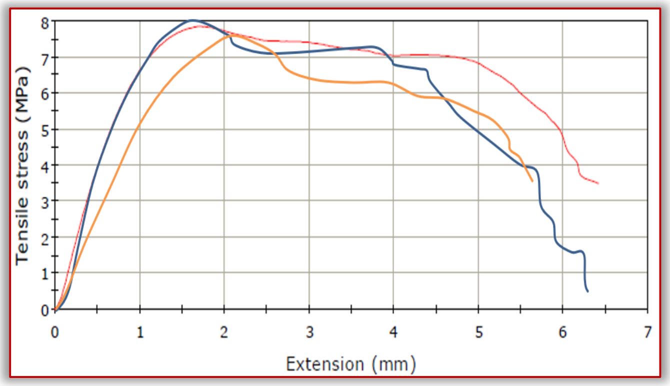

Figure 7. Tensile strength curve for starch mixture 60%–PBAT30%–10%sheep wool, extrusion at 140 °C, 60 rpm, time 10 min. Red curve – mix 1, blue curve – mix 2, orange curve – mix 3.

CONCLUSIONS

Plastic and composite products have become, in the last 70–80 years, important components of everyday life, characterized by good resistance, easy to be processed into different shapes, being much lighter and cheaper than other materials. Their big disadvantage is that they are not biodegradable, creating big environmental problems. An alternative to conventional plastic materials are bioplastic and biocomposite materials. The current research was focused on the development of optimal processes for production of biocomposite materials that meet the functional requirements of users as well as environmental ones. The applied research carried out within the work sought to highlight the connection between the parameters of the biocomposite materials production process (temperature, rpm, extrusion time), from one side, and the mechanical properties of the new material obtained, on the other side.

New biocomposite material recipes have been established, based on starch, biopolymers (PBAT) and natural fibres – miscanthus and sheep wool waste. In this work we analysed the mechanical properties (tensile strength) of the new obtained material. Following the obtained results for tensile strength we can conclude that the new obtained biocomposite materials can be used forward for new applications for agriculture, for agriculture equipments, for automotive industry etc.

In regards to the future research directions we are planning to follow the next steps:

- Production by extrusion of other recipes of starch–based biocomposites, containing natural fibers;
- Realization by injection or thermoforming of some elements for agriculture, agriculture equipments, interior of vehicles;
- Carrying out other tests to evaluate the mechanical properties of the new obtained materials.
- Carrying out tests to evaluate the behaviour in time and the biodegradability of new materials.

Acknowledgement

This work is financed by Ministry of Research, Innovation and Digitalization through Program 1 – Development of the national research–development system, Subprogram 1.2 – Institutional performance – Projects for financing excellence in RDI, Contract no. 1PFE/30.12.2021,, and by NUCLEU program contr. 9N/01.01.2023, project PN 23 04 02 02.

References

[1] Agassant J.–F., Coupez T., Demay Y., Vergnes B., Vincent M., Thermomechanical modelling of polymer processing, in: Advances in the Flow and Rheology of Non–Newtonian Fluids, D. A. Siginer, R. P. Chhabra, D. De Kee Eds., Elsevier (1999).
[2] Bastioli, C. Global status of the production of biobased packaging materials. Conference Proceedings: The Food Biopack Conference, Denmark, 2000, Copenhagen.
[3] Cai Z, Al Faruque MA, Kiziltas A, Mielewski D, Naebe M.(2021), Sustainable lightweight insulation materials from textile–based waste for the automobile industry, Materials, 14(5):1241.
[4] Fu Y., Fodorean G., Navard P., Peuvel–Disdier E. – Study of the partial wetting morphology in polylactide/poly[(butylene adipate)-co-terephthalate]/polyamide ternary blends: case of composite droplets, Polymer International (impact score 3.44), pp. 1378–1385, 2018, https://mc.manuscriptcentral.com/pi–wiley
[5] G. Della Valle, B. Vergnes, D. Lourdin, Viscous properties of thermoplastic starches from different botanical origin, Intern. Polym. Proc., 22, 471–479, 2007.
[6] J.F. Agassant, P. Avenas, J. Ph. Sergent, P.J. Carreau, Polymer Processing. Principles and Modeling, Oxford University Press, 1991.
[7] James E. R.Couper, Richard Erdlac, Handbook of bioplastics and biocomposites engineering applications, Srikanth Pilla, 2011.
[8] McGlashan, S. A., and Halley, P. J., Preparation and characterization of biodegradable starch–based nanocomposite materials. PolymerInternational, 2003.
[9] Mohanty A K, Misra M, Hinrichsen G, "Biofibres, biodegradable polymers and biocomposites: An overview", Macromolecular Materials and Engineering, 2000.
[10] Parra D.F., Tadini C.C., Ponce P. and Lugão A.B., Mechanical properties and water vapor transmission in some blends of cassava starch edible films, Carbohydrate Polymers, 2004.
[11] Remadevi R., Md Abdullah Al Faruque, Jizhen Zhang, Maryam Naebe,(2020), Electrically conductive honeycomb structured graphene composites from natural protein fibre waste, Materials Letters, 264:127311.
[12] *** Bioplastics: Plants, Raw Materials, Products, Fachagentur Nachwachsende Rohstoffe, Gülzov, Germany, 2005.
[13] *** Rosato, D. Global Bioplastics Legislative Drivers Recap, www.omnexus.com

Note: This paper was presented at ISB–INMA TEH' 2023 – International Symposium on Technologies and Technical Systems in Agriculture, Food Industry and Environment, organized by University "POLITEHNICA" of Bucuresti, Faculty of Biotechnical Systems Engineering, National Institute for Research–Development of Machines and Installations designed for Agriculture and Food Industry (INMA Bucuresti), National Research & Development Institute for Food Bioresources (IBA Bucuresti), University of Agronomic Sciences and Veterinary Medicine of Bucuresti (UASVMB), Research–Development Institute for Plant Protection – (ICDPP Bucuresti), Research and Development Institute for Processing and Marketing of the Horticultural Products (HORTING), Hydraulics and Pneumatics Research Institute (INOE 2000 IHP) and Romanian Agricultural Mechanical Engineers Society (SIMAR), in Bucuresti, ROMANIA, in 5–6 October, 2023.

ISSN: 2067-3809