BULETINUL Universităţii Petrol – Gaze din Ploieşti	Vol. LXIII No. 1/2011	135 - 140	Seria Tehnică

On the Methodology of Obtaining some Biodegradable Polymer Matrix Composite Materials

Marian Mareş, Mihai Dărângă, Camelia Mihăilescu,
Cristina Racu, Liliana Buhu

Technical University “Gh. Asachi”, Bd. Mangeron 61, Iaşi
e-mail: mmares@tuiasi.ro

Abstract

The paper is focused on the manufacture of some composite materials, using as constituents raw materials originating in our country and being produced on the basis of some vegetable or animal resources: a proteic polymeric matrix, and woven bast fiber as reinforcement. As a consequence, one can assume that the final material should be almost completely biodegradable (that issue is evaluated in present). Various polymeric matrices were tested, together with various constituents' proportions and structures. As a result, the suitable variant of composition and reinforcement disposition was choosing in order to obtain a good combination of properties for the studied composite materials.

Key words: *biodegradability, polymer matrix composite, properties.*

Introduction

The manufacture and the extended use of biodegradable materials are acting nowadays as important challenges for science and industries, in order to solve, even partially, the problem of permanently increasing environmental pollution and degradation. On the other hand, composite materials are good candidates for being biodegradable [1], on the basis of the large amount of natural raw materials that could be used as constituents, but having also in view the various possibilities of tailoring their composition and structure, for obtaining a desirable combination of properties for the final material.

Materials

Biodegradability is referring to the possibility, for the integrity of some materials, to be affected by the action of some natural organic factors, which usually exist in the environment. In that way, if many of the waste products were biodegradable, an important reduction of the environmental pollution could be possible.

The raw materials originating in vegetable or animal resources are important candidates for being largely biodegradable. The present paper is an investigation on the possibility of obtaining some biodegradable composites, using raw materials from the internal market of our country:

- a polymeric matrix, proteic in nature (having an animal glue, as the main component);
- woven bast fiber, as a natural originating reinforcement.

The resulting composite materials are intended to be used in applications as the manufacture of packaging and ambient products.

Technological aspects

The proteic polymer was choosing, as the basic component of the matrix, because of its accessibility and its cost effectiveness. It must also be said that the animal glue, when used without any additive, lead to a brittle, low mechanical resistant matrix, and it is very sensitive to water attack. Those drawbacks can be, in a good measure, compensated, using some suitable chemical additives as curing agents.

It is well known that the formaldehyde is usually added, as an agent of that kind, but its use is inconvenient, for many reasons: it increases the stiffness of the basic polymer, and has negative effects on its biodegradability, but also it makes difficult the manufacturing process, because of its emphasized volatility and toxicity.

As a result, a particular additive (pre-polymer acetone-formaldehyde, which was synthesized in our laboratory) was preferred as a curing agent for the polymer matrix of the proposed composite materials. The reinforcement consisted of pieces (with proper dimensions) of a textile material (woven flax strands) that is recommended in literature [2, 3] as a good candidate, in order to be used as reinforcements in green composites. It forms, together with a suitable amount of matrix non-cured material, some pre-impregnated laminas. The superposition of such prepregs, with desirable orientations, leads to obtaining the final laminate material.

The laminate sample was then kept at room temperature, till the matrix resin was gellified, and it was finally dried, at 60°C, till constant weight. Subsequent, in order to increase the mechanical properties of the composite material, the samples were also consolidated, by being heated, at 90°C, and simultaneously pressed, at maximum 1.75MPa (as it will be presented into a separate paper work).

Comparison of different structure types

The first step of the present investigation was the choice of the suitable composition and volume ratios of constituents, for the proposed composite materials. It was establish that the best combination of mechanical properties can be obtained for the polymer matrix composed of animal glue, plasticizer, and curing agent, in such a proportion as 82:10:8, and a good ratio for the volume content is 0.7 - for the matrix to 0.3 - for the textile reinforcement.

Three different variants of reinforcement disposition were then used, in order to determine the type of structure that leads to the best mechanical characteristics of the composite material:

A. having the warp strands running in the longitudinal direction (parallel to the length of the samples);
B. having the fill strands running in the longitudinal direction;
C. composed of alternating layers of the two preceding categories.

The comparison was based on the mechanical characteristics values of the studied composite materials, which were established by tension tests; they were conducted following a specification [4] that is usually used when testing a polymer matrix composite material. The prismatic specimens were cut, at the width of 25mm, from the composite samples, and they had the same thicknesses as the respective samples (4.5-5 mm, corresponding to four plies of pre-impregnated textile reinforcement). According to the above-cited specification, end-tabs were bonded on each specimen (see Fig. 1, where tabs are in grey color), made on 1mm thick aluminum sheet, in order to avoid the negative effects (on the experimental data) of gripping the specimen into the machine fixture.

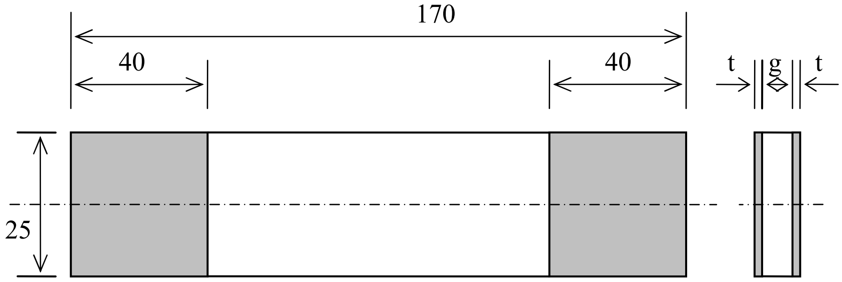

Fig. 1. The composite specimen for tension tests.

The experiments were conducted on a universal, computer-assisted testing machine (Fig. 2), having a maximum loading capability of 50kN. The load-elongation dependence was registered, during the tests, both as numerical data and as the corresponding curves of variation. Uniaxial tensile tests to failure yield the following properties of the studied composites: tensile strength (R_m), ultimate tensile strain (A_r), and Young's modulus (E).

Fig. 2. The achievement of uniaxial tension tests.

Experimental results and discussion

A similar aspect of the stress-strain dependence was observed, for the three categories of composite samples that were evaluated. It is largely assumed, for the laminate composite materials, the fact that, as resulted from uniaxial tension test, the stress-strain curves show two main regions, characterized by different slope values: the big value is present at the beginning of the curve, on its starting quasi-linear segment (on that particular zone is determined the Young's modulus value, for the tested material). The second region of the curve is clearly non-linear, having a down-oriented convexity. It seams to be corresponding to a mechanical response that is dominated by the textile reinforcement of the tested composite sample.

It is interesting to emphasize, for many of the load-elongation dependence curves (see Fig. 3), the fact that, when a certain loading level is exceeded, some discontinuities are observed, leading to a particular ("notched") aspect of the curve, which is keeping, however, its general, above-described, trend. One can consider that phenomenon as corresponding to the beginning of (partial) failure, for some of the matrix layers (see the typical aspect of the failed specimens, in Fig. 4 from below): because of its prominently brittle mechanical behavior, the matrix is not allowing the complete elongation of textile reinforcement, so that its fragmentation leads to the load-transmittance towards some new regions of flax reinforcing strands.

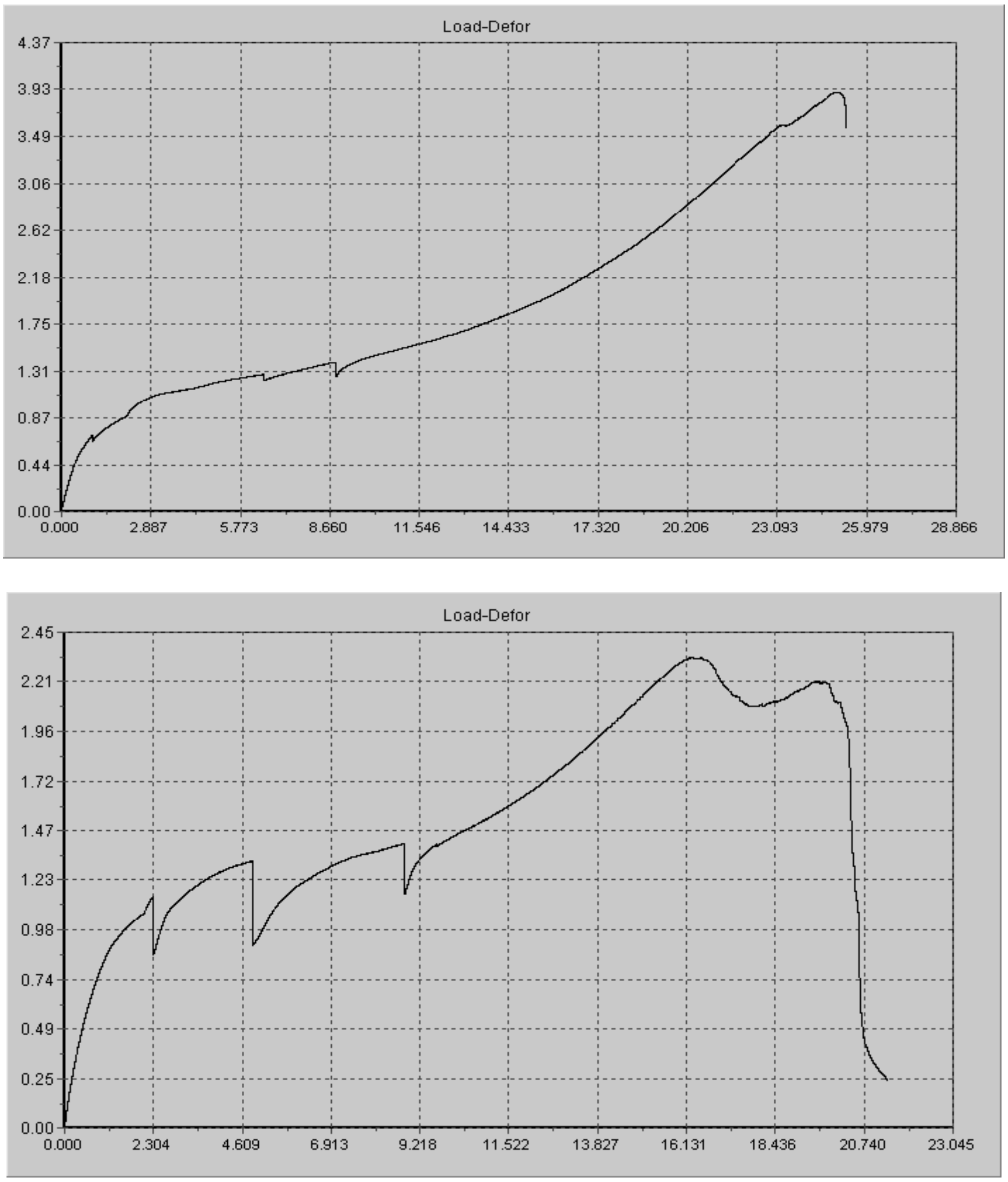

Fig. 3. Typical aspects of load [kN] – elongation [mm] curves, for the studied composite materials.

As it can be observed from the following table (Tab. 1), some different values were obtained, corresponding to the three types of investigated composites; the best combination of tensile strength and ductility was exhibited by the composite samples of type A, for which the load was oriented on the principal direction of the textile reinforcement.

Tab. 1. Average values of mechanical properties for the studied composite materials

Composite type	Tensile Strength [MPa]	Ultimate Tensile Strain [%]	Young's Modulus [MPa]
A	24.8	32.7	315
B	11.5	17.5	448
C	14.5	24.9	496

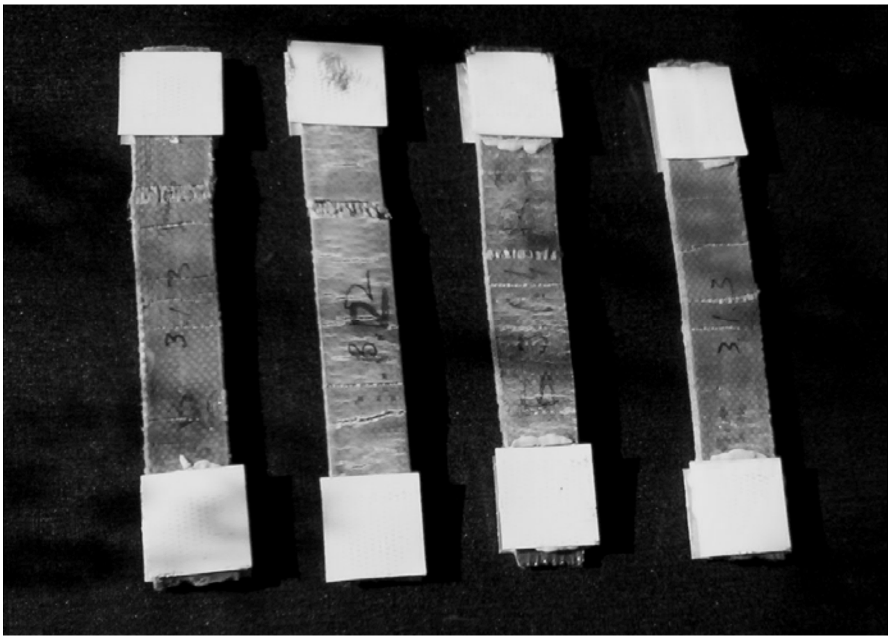

Fig. 4. Typical aspects of failed uniaxial tension tested specimens.

Concluding remarks

1. Many different types of laminate composite material samples have been manufactured based on animal glue, as the polymeric matrix, and various oriented textile reinforcements (consisting of woven flax strands). The total amount of raw materials was produced in our country, and was obtained from the internal market.
2. The raw materials are almost exclusively originating in vegetable and animal resources, and as a consequence an important degree of biodegradability can be estimated for the final material (that issue is experimented in present).
3. The mechanical characteristics, as resulted from uniaxial tension tests, were compared, for three types of reinforcement orientation. On that basis, one can assume that the suitable combination of properties could be obtained for the samples having the warp (reinforcing) strands running in the longitudinal direction (parallel to the length of the samples). It is the structural model that was preferred for being used for the following experiments.

Acknowledgements

The present work was fully supported by PN2 Research Project 72-200/2008.

References

1. Goda K., Cao Y. – Research and Development of Fully Green Composites Reinforced with Natural Fibers, *Journal of Solid Mechanics and Materials Engineering*, Vol. 1, No. 9, pg. 1073-1084, 2007.
2. J.A. Foulk e.a. – Analysis of Flax and Cotton Fiber Fabric Blends and Recycled Polyethylene Composites, *Journal of Polymers and the Environment*, Vol. 14, No. 1, 2006, pg. 15-22.
3. S. Marais e.a. – Unsaturated Polyester Composites reinforced with Flax Fibers: Effect of Cold Plasma and Autoclave Treatments on Mechanical and Permeation Properties, *Composites* Part A, 2005, pg. 975-986.
4. ASTM Standard D 3039 – Standard Test Method for Tensile Properties of Polymer Matrix Composite Materials.

Aspecte metodologice privind obținerea unor compozite biodegradabile cu matrice polimerică

Rezumat

Lucrarea se referă la obținerea unor materiale compozite, folosind materii prime de producție autohtonă, provenind din surse vegetale sau animale: matrice polimerică, pe bază de clei animal şi armare cu țesături din fire de plante liberiene. Pe această bază se poate estima că materialele finale vor avea un grad însemnat de biodegradabilitate (acest aspect este în curs de experimentare). S-au analizat mai întâi diverse variante de matrice polimerică, de proporții între constituenții compozitului, respectiv de dispunere a ranforsantului în structură, fiind stabilită ca definitivă varianta pentru care s-a obținut cea mai bună combinație de proprietăți mecanice.