APPROACHES TO THE EVALUATION OF THE MECHANICAL PROPERTIES OF SINGLE-LAYER COMPOSITE PLATES MADE OF RECYCLABLE POLYMERIC AND PROTEIN MATERIALS

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This paper deals with the theoretical and experimental mechanical characteristics of composite plates obtained from recyclable polymer and protein matrix and fibrous reinforcement. The definition of the theoretical model of the monolayer composite material with its structural elements and the physical-mechanical evaluation of its characteristics leads to the optimal and efficient design and use of all products made of such materials. By the theoretical and experimental determination of the mechanical characteristics that define the properties of the composite material, it can be decided on its use in specific industrial technical applications.

Keywords: composite, recyclable waste, matrix, fibrous material.

INTRODUCTORY CONSIDERATIONS

In the context of ensuring the sustainable development of today's society, as a fundamental desideratum of humanity, the concept of integrated waste management of any kind has been developed. Thus, on the background of solving the national strategic problems of waste management and prioritizing actions to minimize waste production, recycling, composting, energy recovery and ecological storage, there was a need to use urban and / or industrial waste in order to making new materials, such as, for example, single-layer composite materials of the plate type obtained from recyclable polymer waste (PET, PVC, etc.) reinforced with fibrous waste of a protein nature (natural leather, textile fibers, etc.) (Bold, 2003; Căpățână, 2003; Nemeș, 2013).

The theoretical model of the monolayer composite material, made by different manufacturing processes (injection, extrusion, etc.) consists of a single composite layer with the possibility of being able to demonstrate its efficiency by evaluating the mechanical characteristics of its structural components (Hadăr, 2002; Durbacă *et al.*, 2019).

The basic unit for composite materials with fiber reinforcements is the reinforced lamella, (see Figure 1), it confers multiple possibilities of composition and correct introduction of the characteristics in the calculation of the composite.

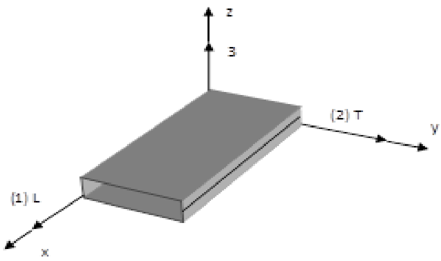

Figure 1. Theoretical model of the single-layer composite plate-type lamella (Hadăr, 2002)

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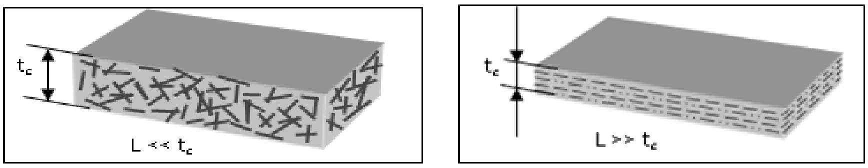
Within the lamella, the fibers that make up the reinforcement are arranged and oriented so as to lead to the achievement of the desired characteristics in the composite structural element.

The reinforced lamella is the elementary part of the composite material which consists of a sample of polymeric matrix reinforced with fibrous material in which the fibers are arranged according to the arrangement of these components in the structure of the composite assembly (random in space or plane), according to Figure 2. In this case, such structures are considered quasi-isotropic in space when the length of the fiber *L* is much smaller than the thickness of the composite, t_c (see Figure 2, a), and in the case of most composite structural elements, the length of the fibers it is much larger than the thickness, achieving quasi-isotropy in the plane (see Figure 2, b).



a. b.

Figure 2. The arrangement of the reinforced fibers in the polymeric matrix of the composite lamella: a - random orientation in space; b - random orientation in the plan

In structural applications where the state of tension is unpredictable, the use of composites with unidirectional reinforcement is insufficient and it is advantageous to use quasi-isotropic layers in the plane, obtained by using short fibers with random orientation.

MECHANICAL CHARACTERISTICS OF SINGLE-SHEET PLATE COMPOSITES

The physical-mechanical characteristics of composite materials reinforced with randomly oriented fibers are determined by parameters such as: fiber diameter *df*, fiber length *L*, volumetric reinforcement fraction *wf*, fiber placement in relation to product axes and manufacturing process.

In order to establish the physical-mechanical characteristics of the composite lamella with polymeric matrix and reinforcement made of fibrous materials, a system of lamella axes is initially chosen. Therefore, in Figs. 1, above, schematically shows a lamella with unidirectional reinforcement: the direction parallel to the fibers *L (1)* is called longitudinal, and the one perpendicular to the fibers *T (2)* is called transverse direction and can correspond to any direction in the plane *(2 ,3)*. Axes *1, 2, 3* are called the main axes of the material.

The main mechanical properties involved in the structural analysis of composite elements are *strength and rigidity*. These properties can be determined experimentally, but the tests are valid for a single fiber-matrix system, obtained with a certain manufacturing process (Hadăr, 2002; Alămoreanu and Chiriță, 1997; Durbacă *et al*., 2017; Păunescu, 2002; Tudorachi, 2007). Therefore, it is recommended to use theoretical and semi-empirical models that allow the evaluation of mechanical characteristics based on parameters that influence the properties of the composite structure. Theoretical models are not always applicable, some direct corrections are necessary for the direct design of the

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elements, especially in the transverse direction, however for the study of mechanical characteristics in the longitudinal direction it is considered that the existing models for composites with continuous unidirectional reinforcement are sufficiently accurate (Iatan *et al.*, 2017; Bere *et al.*, 2012; Boboc, 2019).

Depending on the axis system adopted, for composite materials reinforced with fibers, the following mechanical characteristics necessary in the design are defined: $E_L = E_1$ - longitudinal modulus of elasticity of the lamella (in direction parallel to the fibers); $E_T = E_2$ - transverse modulus of elasticity of the lamella (in a direction perpendicular to the fibers); $G_{LT} = G_{12}$ - modulus of shear elasticity of the lamella in the plane *(L, T)* or *(1,2)*; $\nu_{LT} = \nu_{12}$ and $\nu_{TL} = \nu_{21}$ - Poisson's ratios in the plane *(L, T)* or *(1,2)*; R_{tL} - tensile strength of the blade in the longitudinal direction; R_{tT} - tensile strength of the blade in the transverse direction; R_{cL} - compressive strength of the lamella in the longitudinal direction; R_{cT} - compressive strength of the transverse direction blade; $R_{f\,(LT)} = R_{f\,(12)}$ - the shear strength of the lamella in the plane *(L, T)* or *(1,2)*.

For theoretical analysis, it is considered a composite material with volume v_c, in which the fibers occupy the volume v_f, and the matrix the volume v_m. The same material has the weight w_c, the fibers have the weight w_f, and the matrix weight w_m. Noting with V and W the volumetric and gravimetric fractions, respectively, their definition is made with the help of the relations:

$$v_c = v_f + v_m; \qquad V_f = v_f / v_c; \qquad V_m = v_m / v_c \tag{1}$$

Expressing the masses with the help of the corresponding densities:

$$\rho_c\, v_c = \rho_f\, v_f + \rho_m\, v_m \tag{2}$$

where: ρ_c, ρ_f and ρ_m are the densities of composite, fiber and matrix.

The presence of gaps in the composite element significantly influences some of its mechanical properties. By increasing the gap content, the effects of properties degradation over time are generated and the results on the mechanical characteristics are scattered. A good quality composite must have less than 1% gaps, while an unsuitable one can reach a relative volume of gaps $V_g = 5\%$ (Hadăr, 2002).

An acceptable estimate for the longitudinal modulus of elasticity E of the quasi-isotropic composite in the plane, at axial stress, is obtained based on the relationships established by Gibson (Hadăr, 2002) through a series of simple relations, developed based on Cox's models (Hadăr, 2002) and allowing evaluation of the elasticity modules, distinct for the two cases:

- spatial quasi-isotropic composite:

$$E = \frac{E_f V_f}{6} \tag{3}$$

- quasi-isotropic composite in plan:

$$E = \frac{E_f V_f}{3} \tag{4}$$

The relations determined by Cox (Hadăr, 2002) for the evaluation of the transversal modulus of elasticity G, in the case of the composite reinforced with short fibers arranged at random are:

- spatial quasi-isotropic composite:

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$$G = \frac{E_f V_f}{15} \tag{5}$$

• quasi-isotropic composite in plan:

$$G = \frac{E_f V_f}{8} \tag{6}$$

For flat products made of composites reinforced with random short fibers, the following were used (Hadăr, 2002):

$$G = \frac{E}{2(1+\nu)} \tag{7}$$

where, the Poisson coefficient has the following values ν = 1/4, for spatial quasi-isotropic composite and ν = 1/3, for plane quasi-isotropic composite.

For the calculation of the tensile strength (axial) stresses of short fiber composites randomly distributed in the plane, the following relations are used (Hadăr, 2002):

$$R_t = \frac{2R_{f(LT)}}{\pi}\left[1 + \frac{R_{tT}}{(\sigma_m)_{\varepsilon_f^*}} + \ln\frac{R_{tT}(\sigma_m)_{\varepsilon_f^*}}{R_{f(LT)}^2}\right] \tag{8}$$

where, $R_{f\,(LT)}$, R_{tT} are the resistances of the unidirectional composite with continuous reinforcement calculated with the expressions (Hadăr, 2002).

For the calculation of the values of the mechanical characteristics of the composite reinforced with short fibers randomly distributed, other than those presented above, there are currently no acceptable mathematical models to allow their evaluation at values comparable to those determined experimentally.

CASE STUDY

The mechanical characteristics of a PVB / leather composite sample are analyzed, in which the matrix is represented by the PVB polymer (polyvinyl butyral), made by polycondensation of vinyl acetate with aldehydes, and the reinforcement / reinforcement material is represented by short semi-coarse crushed leather fibers; the polymer is in the form of a white-cream powder, with a density of approximately 1,2 g/cm³, and the processing temperature is between 120 ÷ 170 ºC.

Due to the presence of chromium in the skin fibers, which requires use at relatively low processing temperatures, this makes PVB a very interesting polymer matrix for the development of the case study for PVB / leather composite. Figure 3 below shows the microfractrography of some breaking surfaces of a PVB / leather composite sample, if the skin fibers are unevenly distributed in the PVB polymer matrix.

These composites were obtained by the same process used for the preparation of PVB / wood flour composite. Although there are some gaps in the polymer matrix, probably due to the defibration of the leather waste and the agglomeration of the skin fibers, a good adhesion can be given by the continuity of the agglomeration of the skin fibers in the polymer matrix PVB (de Almeida Lucas *et al*., 2011). As mentioned above, good adherence is usually difficult to achieve between the thermoplastic matrix and the natural fibers, due to the differences in polarity between the hydrophilic fibers (composed of collagen macromolecules) and the hydrophobic thermoplastic matrix. The PVB copolymer contains vinyl hydroxyl alcohol groups that can interact with the -OH group and -C(=O)-OH groups of collagen macromolecules in the skin fibers. The interaction between these groups can create an interface that manages to transfer the stress from the PVB polymer matrix on the cryogenic fracture of the skin fibers (de Almeida Lucas *et al.*, 2011; Wehry 2002).

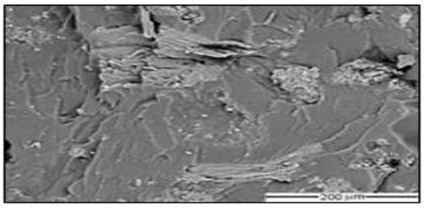

Figure 3. Microfractrography of some breaking surfaces
of a PVB / leather composite sample (de Almeida Lucas *et al.*, 2011)

Figure 4 below shows the results of tensile tests for PVB composite with leather fibers obtained from waste recycling. The modulus of elasticity increases considerably, with the increasing content of leather fibers in the composite. The modulus of elasticity increases from 4 MPa for PVB to 270 MPa for composite with 70% skin fiber. Such a rapid and nonlinear growth of the modulus of elasticity with the fiber content is not frequently observed in thermoplastic composites reinforced with natural fibers. As previously presented, PVB contains plasticizer which gives it improved flexibility in composites.

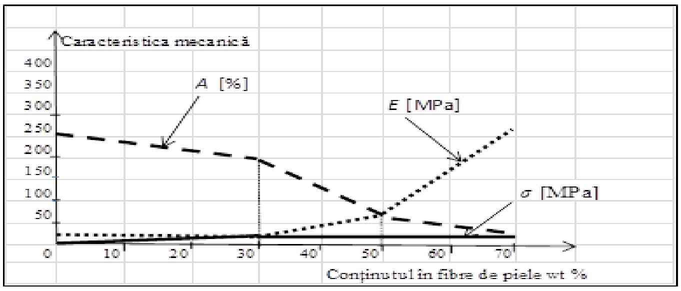

Figure 4. Variation of the mechanical properties (E, σ_r and A) of the composite depending on the content of skin fibers.

In the literature there are a number of studies on thermoplastic composites containing skin fibers (de Almeida Lucas *et al.*, 2011). Polymer matrices may include plasticized PVC, acrylonitrile-butadiene-styrene (ABS) and methyl polymethacrylate (PMMA), if the modulus of elasticity indicates different behavior. While ABS / leather and PMMA / leather composites show decreases in modulus E values or a slight increase depending on the ascending content of leather fibers, PVC / leather composites show a similar behavior to PVB / leather composites, namely, the modulus elasticity is strongly influenced by the fiber content, especially in composites that exceed the skin fiber content $w_t = 30\%$.

It is pointed out that the polymer/leather fiber composite material, if the modulus of elasticity increases considerably with increasing amount of skin fibers, had a plasticized

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thermoplastic matrix, respectively by the existence of PVC and PVB polymeric materials used in this paper. The reduction in the tensile strength of PVB / leather composites can be attributed to a reduction in the deformation capacity of the polymer matrix, due to the concentration of leather fibers.

CONCLUSIONS

Despite the fact that the mechanical properties of the experimentally determined monolayer composite plate do not agree acceptably with those analyzed analytically, it nevertheless showed appropriate levels of use.

Therefore, the use of PVB / leather composite for industrial production, such as shoe soles and other parts of its structure (insole roof, heel, toe, etc.) it is supported by a number of mechanical properties (abrasion resistance, hardness, tear strength, adhesion to commercial adhesives, flexibility or folding capacity, resistance to repeated bending, etc.), as well as design, and have now been successfully tested in footwear companies.

By means of a simple and inexpensive processing technique (for example, an extrusion flow equipped with a single auger), PVB / leather composites have been manufactured with properties suitable for use in the footwear industry, to be returned as a raw material to the industrial chain and to facilitate the approval of recycled products by technical regulatory requirements.

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