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Analitični model določevanja mehanskih lastnosti biopolimernih kompozitov - mikromehanski postopek

A Model for Predicting the Mechanical Properties of Wood-Plastic Composites - A Micro-Mechanical Approach

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V zadnjem desetletju prejšnjega stoletja se je pričel razvoj novih kompozitov, sestavljenih iz naravnih vlaken in cenejših polimerov, kakor sta npr. HDPE in PP. Zaradi relativno kratkega časa prisotnosti na trgu in naključnih lastnosti vlaken do sedaj še ni bilo poskusov analitičnega določevanja mehanskih lastnosti omenjenih kompozitov. V tem prispevku je uporabljen mikromehanični postopek, imenovan posplošena metoda celic (PMC - GMC) za popis lastnosti injekcijsko brizganih biopolimernih kompozitov, sestavljenih iz polipropilena (PP) ali polistirena (PS) ter lesnih ali celuloznih vlaken. Glavni problem pri analitičnim postopku je, da naravna vlakna nimajo enotne dolžine, prereza ali oblike, zato jih je težko popisati z običajnimi matematičnimi modeli, tako da so v prispevku uporabljene povprečne vrednosti geometrijskih lastnosti vlaken. Kompoziti so bili najprej pregledani z optičnim in elektronskim mikroskopom z namenom določiti lastnosti in razporeditev vlaken. Ti podatki so bili nato uporabljeni pri ugotavljanju elastičnega in plastičnega odziva kompozita ter njegovih porušnih vrednosti, ki so bile izračunane po Tsai-Hillovi porušni hipotezi. Rezultati so bili nato primerjani z eksperimentalnimi podatki, tako da je bilo mogoče oceniti praktično uporabnost te metode. Časovno odvisne mehanske lastnosti materiala tu niso bile upoštevane zaradi zapletenosti in naključnih lastnosti vlaken.

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(Ključne besede: biopolimerni kompoziti, posplošena metoda celic, naravna vlakna, mehanske lastnosti)

The development of a new composite that is made up of natural fibres and low-price polymers, such as HDPE or PP, began in the 1990s. Because this material is relatively new to the market and due to the random characteristics of the fibres, no attempts have been made to analytically define the mechanical properties of this material. In this paper a micro-mechanical approach, called the generalised method of cells (GMCs), is introduced to describe the properties of injection-moulded wood-plastic composites made up of polypropylene (PP) or polystyrene (PS) and wood or cellulose short fibres. The main problem with an analytical approach is that the natural fibres are not uniform in shape and size, which makes them hard to fit into standard mathematical models. In this paper the average values of the fibre sizes were used. The materials were first scanned with optical and electron microscopes to determine the fibre properties and their scatter. These values were then used to determine the elastic and plastic response of the composite together with the maximum strength and elongation of the composite, where the Tsai-Hill failure-criterion was used. The results were then compared to the experimental data in order to evaluate the practical usefulness of this method. The time-dependent mechanical behaviour of the composite was not considered due to the complex and random properties of the fibre.

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(Keywords: wood-plastic composites, generalised method of cells (GMC), natural fibres, mechanical properties)

0 UVOD

V zadnjem desetletju prejšnjega stoletja se je pričel razvoj novih kompozitov, v katerih so bile združene dobre lastnosti tako lesa kakor tudi

0 INTRODUCTION

The development of a new composite began in the 1990s. This composite combined the good properties of wood and polymer [1]. It became polimerov [1]. Izkazalo se je, če čistemu polimeru, na primer polietilenu (PE), dodamo lesna vlakna, se njegove mehanske lastnosti znatno izboljšajo. Povečata se mu elastični modul *E* in natezna trdnost, obdrži pa tudi nekatere dobre lastnosti lesa, ob tem pa je kompozit razmeroma dobro odporen proti vremenskim pojavom.

Ker so naravna vlakna naključno oblikovana v razmeroma zapletene geometrijske oblike, je njihov popis mnogo težavnejši v primerjavi z drugimi kratkimi, npr. steklenimi ali ogljikovimi vlakni, ki so ravna in imajo nespremenljiv prerez. Pri le-teh, namreč lahko, če poznamo splošno usmeritev vlaken ter lastnosti posameznega vlakna, dokaj natančno določimo elasto-plastične lastnosti kompozita [2]. Vendar je pri naravnih vlaknih to problem, saj je nemogoče določiti lastnosti posameznega lesnega ali celuloznega vlakna, vlakna so mnogo prekratka, da bi na njih opravili natezni preizkus. Povrh tega se njihove lastnosti ob toplotnem postopku kompozita spremenijo, saj je za učinkovito brizganje mešanico treba segreti na okrog 190 °C. Vlakna po postopku tudi niso ravna, ampak zvita, kar še dodatno otežuje njihov popis.

Metoda celic in njena posodobljena različica, tj. posplošena metoda celic (PMC), ki ju je v 90. letih razvil Jacob Abudi [3], je analitični mikromehanski model za določevanje elastičnih in plastičnih lastnosti vlaknatih kompozitov. Temelji na predpostavki, da so vlakna periodično urejena, med njimi pa je osnovni material. Periodična narava kompozita omogoči, da ga razdelimo na med seboj enake gradnike ali celice, nato pa obravnavamo le posamezno celico. Lastnosti posamezne celice so tako veljavne za celotni kompozit. Tako lahko popišemo kompozite z dolgimi vlakni (dvojna periodičnost celic), kratkimi vlakni (trojna periodičnost), laminate ter tudi porozne materiale [4]. V nadaljevanju prispevka so predstavljeni teoretični model PMC, eksperimentalno delo ter primerjava med eksperimentalnimi in teoretičnimi, s PMC pridobljenimi podatki.

1 OPIS POSPLOŠENE METODE CELIC

Ker opisujemo prekinjani kompozit s kratkimi vlakni, je treba uporabiti metodo celic s trojno periodičnostjo. Tu so vlakna periodično urejena v osnovi v identične kvadre, kar prikazuje evident that if wood particles are added to a polymer matrix such as polyethylene (PE) the mechanical properties of the new composite are significantly improved. The Young's modulus E and the tensile strength are increased and, additionally, the composite is also weather resistant.

Natural fibres have a much more complex and random geometry than other short fibre fillers. such as glass or carbon fibres, which are straight and have a constant cross-section. With these types of fibres, knowing the general orientation and properties of an individual fibre, means that the elasto-plastic properties of the composite can be determined with a high degree of accuracy [2]. On the other hand, it is nearly impossible to determine the properties of an individual wood or cellulose fibre because it is much too short for tensile tests. Furthermore, their properties are changed after thermally processing the composite due to heating to at least 190°C, which is required for an effective injection. In addition, the fibres are not straight after the processing; they are curled, which makes their description even more difficult

The method of cells and its modernised version, the generalised method of cells (GMCs), were developed in the 1990s by Jacob Abudi [3]. They represent an analytical micro-mechanical model for determining the elastic and plastic properties of the composites. It is based on the assumption that the fibres are arranged in a periodic array and the space between them is occupied by the matrix material. The periodic nature of the composite makes it possible to divide it into equal building blocks or cells, and then to study only an individual cell. The properties of the one cell are then representative of the whole composite. Thus, composites with long fibres (double periodicity) or short fibres (triple periodicity), laminates and porous materials can be described [4]. In the remainder of the paper a theoretical model of GMCs is presented, as well theoretical work and a comparison between the experimental and theoretical results based on the GMCs.

1 DESCRIPTION OF THE GENERALISED METHOD OF CELLS

Since a discontinuous composite with short fibres is being described, the method of cells with a triple periodic array has to be used. Here, the fibres are periodically arranged in identical rectangular



Sl. 1. (a) Kompozit z vključki vlaken, urejenimi v trojno periodično razporeditev, (b) predstavitvena celica z osmimi podcelicami α , β , $\gamma = 1,2$

Fig. 1. (a) Composite with fibre inclusions, arranged in a triply periodic array, (b) Representative cell with eight subcells α , β , $\gamma = 1,2$

slika 1a. Kvader $d_i h_i l_i$ označuje vlakno in skupaj s parametri d_{y} h_{z} in l_{z} , ki označujejo razmik med vlakni, predstavljajo eno celico, kakor jo prikazuje slika 1b. Izmere te celice so $d_i + d_z$, $h_i + h_z$ in $l_i + l_z$ v ustreznih smereh x_i , x_z in x_3 . Celica je razdeljena v osem podcelic za α , β , $\gamma =$ 1,2, v središču katerih so lokalni koordinatni sistemi $\begin{pmatrix} z_{1}^{(\alpha)} & z_{2}^{(\gamma)} & z_{1}^{(\gamma)} \end{pmatrix}$, katerih smeri so vzporedne z glavnimi sorednicami. Glede na sliko 1b lahko pomik v katerikoli točki znotraj podcelice $\alpha\beta\gamma$ prikažemo z enačbo (1) po teoriji prvega reda: parallelepipeds, as shown in *Fig. 1a.* The parallelepiped $d_i h_i l_i$ represents the fibre, and together with the parameters d_{χ} , h_2 and l_{χ} which denote the spacing between the fibres, it represents a single cell, as shown in *Fig 1b.* The dimensions of this cell are $d_i + d_2$, $h_i + h_2$ and $l_i + l_2$ in the corresponding directions x_i , x_2 and x_j . The cell is divided into eight subcells, where α , β , $\gamma = 1,2$. In the centre of each subcell a local system of coordinates $\begin{pmatrix} r(\alpha) & r(\beta) & r(\beta) \\ x_1 & x_2 & x_3 \end{pmatrix}$ is introduced, which is oriented in parallel with the main coordinate system. Referring to *Fig. 1b*, the displacement at any point within the subcell $\alpha\beta\gamma$ can be expressed in the framework of the first-order theory by equation (1) [3]:

$$u_{i}^{(\alpha\beta\gamma)} = w_{i}^{(\alpha\beta\gamma)} + x_{2}^{-(\alpha)} \phi_{i}^{(\alpha\beta\gamma)} + x_{2}^{-(\beta)} \chi_{i}^{(\alpha\beta\gamma)} + x_{3}^{-(\gamma)} \psi_{i}^{(\alpha\beta\gamma)}, \ i = 1, 2, 3$$
(1),

kjer je $w_i^{(\alpha\beta\gamma)}$ premik središča podcelice in $\phi_i^{(\alpha\beta\gamma)}, \chi_i^{(\alpha\beta\gamma)}$ in $\psi_i^{(\alpha\beta\gamma)}$ so mikrospremenljivke, ki ponazarjajo premo odvisnost pomika $u_i^{(\alpha\beta\gamma)}$ glede na lokalne koordinate $x_1^{-(\alpha)}, x_2^{-(\beta)}, x_3^{-(\gamma)}$. Komponente deformacijskega tenzorja so:

$$\varepsilon_{ij}^{(\alpha\phi\gamma)} = \frac{1}{2} \left(\partial_{i} u_{j}^{(\alpha\phi\gamma)} + \partial_{j} u_{i}^{(\alpha\phi\gamma)} \right), \quad i, j = 1, 2, 3$$
⁽²⁾

kjer so parcialni odvodi $\frac{\partial}{\partial x_1}$ in $\frac{\partial}{\partial y_2}$ podani po pravilu: $\frac{\partial}{\partial x_1} = \frac{\partial}{\partial x_1}$, $\frac{\partial}{\partial x_2} = \frac{\partial}{\partial x_2}$ in $\frac{\partial}{\partial x_3} = \frac{\partial}{\partial x_3}$. Za oba materiala, tako osnovo kakor vlakna, določimo Where the partial derivations ∂_i and ∂_j are $\partial_1 = \partial/\partial x_1$, $\partial_2 = \partial/\partial x_2$ and $\partial_3 = \partial/\partial x_3$. Both materials, the matrix and the fibres, are treated as

popolnoma elastične la<u>s</u>tnosti. Povprečno napetost v celotnem kompozitu σ_{ij} lahko zapišemo v obliki: perfectly elastic. The average stress in the whole composite σ_{ij} is given by:

kjer so $V = (d_1 + d_2)(h_1 + h_2)(l_1 + l_2),$ $v_{\alpha\beta\gamma} = d_{\alpha}h_{\beta}l_{\gamma}$ in σ_{ij} povprečna napetost v podcelici. Podobno velja tudi za povprečno deformacijo ε_{ij} :

$$= \frac{1}{V} \sum_{\sigma=1}^{2} \sum_{\theta=1}^{2} \sum_{\nu=1}^{2} v_{\alpha\beta\gamma} \overline{e}_{ij}^{-(\alpha\beta\gamma)}$$
(4).

Pri prečno izotropnem materialu, kjer je x_i smer anizotropije, so napetosti in deformacije povezane takole:

E.

For transversely isotropic material, where x_i is the anisotropy direction, the stresses are related to strains in the following form:

$$\left\{\sigma^{(a\beta\gamma)}\right\} = \left\{C^{(a\beta\gamma)}\right\} \left(\left\{\varepsilon^{(a\beta\gamma)}\right\} - \left\{\varepsilon^{(a\beta\gamma)}\right\}\right) - \left\{\Gamma\right\} \Delta T$$
(5),

composite E !! :

kjer sta stolpec komponent tenzorja napetosti in deformacij: where the columns of stress and strain tensor components:

$$\left\{ \boldsymbol{\sigma}^{(\alpha\beta\gamma)} \right\} = \left\{ \boldsymbol{\sigma}_{11}^{(\alpha\beta\gamma)}, \boldsymbol{\sigma}_{22}^{(\alpha\beta\gamma)}, \boldsymbol{\sigma}_{33}^{(\alpha\beta\gamma)}, \boldsymbol{\sigma}_{12}^{(\alpha\beta\gamma)}, \boldsymbol{\sigma}_{13}^{(\alpha\beta\gamma)}, \boldsymbol{\sigma}_{23}^{(\alpha\beta\gamma)} \right\}$$

$$\left\{ \boldsymbol{\varepsilon}^{(\alpha\beta\gamma)} \right\} = \left\{ \boldsymbol{\varepsilon}_{11}^{(\alpha\beta\gamma)}, \boldsymbol{\varepsilon}_{22}^{(\alpha\beta\gamma)}, \boldsymbol{\varepsilon}_{33}^{(\alpha\beta\gamma)}, \boldsymbol{\varepsilon}_{12}^{(\alpha\beta\gamma)}, \boldsymbol{\varepsilon}_{13}^{(\alpha\beta\gamma)}, \boldsymbol{\varepsilon}_{23}^{(\alpha\beta\gamma)} \right\}$$

$$\left\{ \boldsymbol{\sigma} \right\} = \mathbf{C} \left\{ \boldsymbol{\varepsilon} \right\}$$

$$\left\{ \boldsymbol{\sigma} \right\}$$

$$\mathbf{C}^{(\alpha\beta\gamma)} = \begin{pmatrix} c_{11}^{(\alpha\beta\gamma)} & c_{12}^{(\alpha\beta\gamma)} & c_{12}^{(\alpha\beta\gamma)} & 0 & 0 & 0 \\ c_{12}^{(\alpha\beta\gamma)} & c_{22}^{(\alpha\beta\gamma)} & c_{23}^{(\alpha\beta\gamma)} & 0 & 0 & 0 \\ c_{12}^{(\alpha\beta\gamma)} & c_{23}^{(\alpha\beta\gamma)} & c_{33}^{(\alpha\beta\gamma)} & 0 & 0 & 0 \\ 0 & 0 & 0 & c_{44}^{(\alpha\beta\gamma)} & 0 & 0 \\ 0 & 0 & 0 & 0 & c_{44}^{(\alpha\beta\gamma)} & 0 \\ 0 & 0 & 0 & 0 & 0 & c_{66}^{(\alpha\beta\gamma)} \end{pmatrix}$$

 $\{\varepsilon^{(\alpha\beta\gamma)}\}$ pomeni deformacijski stolpec v plastičnem področju. Lastnosti elastičnosti za transverzno izotropni material v podcelici $\alpha\beta\gamma$ so predstavljene v enačbi (7) [5]. Za oba materiala, osnovo in vlakna, določimo popolnoma elastične lastnosti, tako da predpostavimo $\{\varepsilon^{(\alpha\beta\gamma)}\}=0$, čeprav bomo v nadaljevanju določevali lastnosti kompozita tudi v plastičnem področju. To dosežemo s t.i. prirastkovnim popisom, pri katerem krivuljo $\sigma-\varepsilon$ v plastičnem področju ponazorimo z nizom premic [6]. ΔT v enačbi (5) označuje spremembo temperature. Ker v naslednjih izračunih predpostavimo, da je ta nespremenljiva, se ta del enačbe nadalje zanemari.

Premiki in napetosti na mejah podcelic morajo biti enaki, tako da se vzpostavi mikroravnotežje. Primer takega ravnotežja med prvima dvema podcelicama $\alpha = 1,2$ je prikazan v enačbi (8): $\{\varepsilon^{(\alpha\beta\gamma)}\}\$ denotes the strain column in a plastic area. The elastic compliance inverse for the transversely isotropic material in the subcell $\alpha\beta\gamma$ is presented in equation (7) [5]. Because both materials, the matrix and the fibres, are treated as perfectly elastic, it can be assumed that $\{\varepsilon^{(\alpha\beta\gamma)}\}=0$. Nevertheless, the plastic properties of the composite will also be described in what follows. This is achieved with an incremental presentation, where the curve σ - ε in the plastic area is described by a series of linear lines. ΔT in Equation (5) denotes the temperature difference. Since it is assumed that the difference vanishes, it can be omitted in subsequent calculations.

Displacements and stresses have to be continuous at the subcells' $\alpha = 1,2$ interfaces, where a micro-equilibrium is established. An example of such an equilibrium between the first two subcells is presented in Equation (8):

$$\begin{split} {}^{(1\beta\gamma)} \Big|_{\vec{x}_{1}^{(1)} = \frac{d_{1}}{2}} &= u_{i}^{(2\beta\gamma)} \Big|_{\vec{x}_{1}^{(2)} = -\frac{d_{2}}{2}} \\ {}^{(1\beta\gamma)} \Big|_{\vec{x}_{1}^{(1)} = \pm \frac{d_{1}}{2}} &= \sigma_{1i}^{(2\beta\gamma)} \Big|_{\vec{x}_{1}^{(2)} = \pm \frac{d_{2}}{2}} \end{split}$$
(8).

Podobno naredimo na vseh mejah med podcelicami β , $\gamma = 1,2$. Na podlagi teh izrazov izločimo mikrospremenljivke $\phi_{1}^{(\alpha\beta\gamma)}, \chi_{2}^{(\alpha\beta\gamma)}$ in $\psi_{1}^{(\alpha\beta\gamma)}$ in izpeljemo niz enačb, ki ponazarjajo celostno obnašanje kompozita s kratkimi vlakni [4]. To naredimo z izrazi, ki povežejo mikrodeformacije v podcelicah z makrodeformacijami v kompozitu skozi ustrezni tenzor A [4]. Povezava med povprečnim raztezkom in napetostjo v vsaki podcelici ter celotnim raztezkom in napetostjo je sedaj:

A similar process is used on all the borders between the subcells β , $\gamma = 1,2$. By doing this the micro-variables $\phi_i^{(\alpha\beta\gamma)}, \chi_i^{(\alpha\beta\gamma)}$ and $\psi_i^{(\alpha\beta\gamma)}$ can be eliminated and a series of equations that represent the overall behaviour of the short-fibre composite can be obtained [4]. This is achieved by establishing the relationships that connect the micro-strains in the subcells to the macro-strains in the composite by an appropriate tensor A [4]. The average stress and strain in each subcell and can now be expressed by the total strain in the following way:

$$\varepsilon^{-(a\beta\gamma)} = A \varepsilon^{-(a\beta\gamma)} \tilde{\varepsilon} \text{ in/and } \sigma^{-(a\beta\gamma)} = C A \varepsilon^{-(a\beta\gamma)} \tilde{\varepsilon}$$
(9).

Tenzor A je sestavljen iz naslednjih komponent:

$$\vec{A} = \begin{pmatrix} \vec{A}_M \\ \vec{A}_\sigma \end{pmatrix}^{-1} \begin{pmatrix} 0 \\ \vec{J} \end{pmatrix}_{-1}$$

where B is:

kjer pomenijo: AM elastične lastnosti materialov podcelicah, Ac geometrijske izmere celice in J povprečne raztezke celotnega kompozita. Na koncu lahko vpeljemo dejanski elastični tenzor kompozita B :

kjer je B podan z naslednjo enačbo:

$$= \frac{1}{V} \sum_{\alpha=1}^{2} \sum_{\beta=1}^{2} \sum_{\gamma=1}^{2} v_{\alpha\beta\gamma} \overline{C}^{(\alpha\beta\gamma)} \overline{A}^{(\alpha\beta\gamma)}$$
(12).

Za ponazoritev odpovedi materiala je uporabljena Tsai-Hillova porušna hipoteza. Med obremenjevanjem materiala se za vsak vnaprej določen prirastek raztezka preveri, ali je v določeni podcelici prišlo do porušitve. V tem primeru Tsai-Hillova hipoteza prevzame obliko enačbe (13):

B

For the composite failure the Tsai-Hill failure criterion is used. While loading the material with force, every increment of strain defined in advance is checked for failure in any of the subcells. In this case the Tsai-Hill criterion assumes the form of Equation (13):

$$\left(G^{(\alpha\beta\gamma)} + H^{(\alpha\beta\gamma)}\right) \left(\sigma_{11}^{(\alpha\beta\gamma)}\right)^{2} + \left(F^{(\alpha\beta\gamma)} + H^{(\alpha\beta\gamma)}\right) \left(\sigma_{22}^{(\alpha\beta\gamma)}\right)^{2} + \left(F^{(\alpha\beta\gamma)} + G^{(\alpha\beta\gamma)}\right) \left(\sigma_{33}^{(\alpha\beta\gamma)}\right)^{2} - 2H^{(\alpha\beta\gamma)}\sigma_{11}^{(\alpha\beta\gamma)}\sigma_{22}^{(\alpha\beta\gamma)} - 2G^{(\alpha\beta\gamma)}\sigma_{11}^{(\alpha\beta\gamma)}\sigma_{33}^{(\alpha\beta\gamma)} - 2F^{(\alpha\beta\gamma)}\sigma_{22}^{(\alpha\beta\gamma)}\sigma_{23}^{(\alpha\beta\gamma)} + 2L^{(\alpha\beta\gamma)}\left(\sigma_{23}^{(\alpha\beta\gamma)}\right)^{2} + 2M^{(\alpha\beta\gamma)}\left(\sigma_{13}^{(\alpha\beta\gamma)}\right)^{2} + 2N^{(\alpha\beta\gamma)}\left(\sigma_{12}^{(\alpha\beta\gamma)}\right)^{2} = 1$$

$$(13)$$

 $\sigma = B\varepsilon$

 $G^{(\alpha\beta\gamma)}, H^{(\alpha\beta\gamma)}, F^{(\alpha\beta\gamma)}, L^{(\alpha\beta\gamma)}, M^{(\alpha\beta\gamma)}$ in $N^{(\alpha\beta\gamma)}$ porušne trdnosti materiala v podcelici αβγ. Zaradi velikega števila linearnih enačb je treba rešitev poiskati z računalnikom.

 $G^{(\alpha\beta\gamma)}, H^{(\alpha\beta\gamma)}, F^{(\alpha\beta\gamma)}, L^{(\alpha\beta\gamma)}, M^{(\alpha\beta\gamma)}$ and $N^{(\alpha\beta\gamma)}$ are the failure strengths of the material in the subcell $\alpha\beta\gamma$. Because of the large number of linear equations involved, the solution has to be found by using a computer.

(11).

Tensor A consists of the following components:

$$\vec{A}_{M}$$
 represents the elastic properties of the materials, \vec{A}_{a} is the geometrical dimensions cell and \vec{J} is the average strain of the whole

the ons of the co ole composite. Now the effective elastic tensor B of the composite can be established:

$$= \begin{pmatrix} A_{M} \\ \bar{A}_{O} \end{pmatrix} \begin{pmatrix} \bar{J} \\ \bar{J} \end{pmatrix}$$
where \bar{A}_{M} represents the elast

$$\begin{pmatrix} \vec{A}_{\sigma} \\ \vec{J} \end{pmatrix} \begin{pmatrix} \vec{J} \\ \vec{J} \\ \vec{J} \end{pmatrix}$$
 where \vec{A}_{M} represents the elastic p subcell materials, \vec{A}_{σ} is the geometric

2 EKSPERIMENTALNO DELO

2.1 Brizganje

Da bi raziskali vpliv količine naravnih vlaken, je bilo treba pripraviti več mešanic iz različnih osnovnih polimerov in z različnimi deleži vlaken. Za osnovo smo uporabili PP K948 ter PS Empera 524N. Naravna vlakna so bila sestavljena iz surove celuloze ali mehkega borovega lesa. Njihova vhodna dolžina se je spreminjala od 0,035 do 0,7 mm, razmerje med dolžino in premerom vlakna a pa je bilo pred postopkom med 3 in 30. Vendar pa so se med sestavljanjem zrn in med brizganjem v polžu vlakna lomila, tako da je bila končna največja dolžina 0,18 mm in a okoli 7 mm. Ker je bilo treba zrna PS za potrebe brizganja pripraviti na višii temperaturi kakor za PP, to je na 205 °C namesto 180 °C, se je to poznalo na mehanskih lastnostih vlaken, saj je bil elastični modul za več ko polovico manjši. Sestava mešanic in parametri brizganja so podani v preglednicah 1 in 2.

Vsak kompozit je bil najprej sušen 4 ure pri temperaturi 100 °C in nato izbrizgan v običajni preizkušanec za natezni preizkus po standardu ISO 527-2.

2 EXPERIMENTAL WORK

2.1 Injection moulding

In order to research the influence of the quantity of natural fibres, several mixtures from various matrix polymers and with different fractions of fibres were prepared. PP K948 and PS Empera 524N were used as the base materials. The natural fibres were made of cellulose or pine soft wood. Their input lengths varied from 0.035 to 0.7 mm and the aspect ratio of the length and the diameter of a fibre a was between 3 and 30 before processing. But during the compounding of the granulate and during the injection moulding, the fibres broke down. Therefore, the maximum output length was 0.18 mm and the value of a was around 7. The composite that used PS as a matrix needed to be prepared for injection at a higher temperature than for PP, i.e., at 190°C instead of 180°C. This had a clear influence on the fibres' mechanical properties and the Young's modulus was less than half the size of the fibres mixed with PP. The injection-moulding parameters and the composition of mixtures are shown in Tables 1 and 2.

Each mixture was dried for 4 hours at 100°C and then injected into a standard dog-bone shape specimen for tensile testing according to the standard ISO 527-2.

5 61				
	PP	PS		
Temperatura grelnika [°C] Heater temperature [°C]	180	190		
Temperatura orodja [°C] Mould temperature [°C]	30	30		
Dodatni tlak [MPa] Packaging pressure [MPa]	500	544		
Hitrost brizganja [mm/s] Injection speed [mm/s]	30	17		
Čas hlajenja [s] Cooling time [s]	20	35		

Preglednica 1. Parametri brizganja Table 1. Injection-moulding parameters

Pregledn	ica 2. Sest	ava testirani	ih kompozitov [7]
Table 2.	Compositio	on of tested	composites [7]

12.2	Masni delež	Volumski delež	Povprečna dolžina	
Osnova Matrix	vlaken Fibre mass ratio [%]	vlaken Fibre volume ratio [%]	vlaken Average fibre length [mm]	<i>a</i> _{<i>R</i>}
PP*	50	0,73	0,1	3
PP*	60	0,79	0,1	3
PP*	40	0,63	0,1	3
PP	50	0,73	0,1	3
PP	60	0,79	0,1	3
PS	40	0,63	0,035	3,5
PS	50	0,73	0,035	3,5
PS	30	0,55	0,14	5
PS	20	0,41	0,14	5
PS	20	0.41	0,18	6,5

Kompozitu so bila dodana mehka lesna vlakna namesto celuloznih Instead of cellulose, soft-wood fibres were added

2.2 Natezni preizkus ter določevanje mehanskih in geometrijskih lastnosti vlaken

Material smo enoosno obremenjevali v smeri x_i ter merili trenutno vzdolžno dejansko napetost v kompozitu $\sigma_{ef} = \overline{\sigma}_{11}$ in vzdolžni dejanski raztezek $\varepsilon_{ef} = \overline{\varepsilon}_{11}$. Natezni preizkus je bil opravljen pri hitrosti raztezanja 3 mm/min. Vzdolžni modul elastičnosti E_A smo izračunali iz strmine krivulje $\varepsilon_{ef} - \sigma_{ef}$ po standardu za polimerne materiale ISO 527-1.

Za izračun po PMC je treba poznati natančne podatke o vhodnih materialih, kar se v primeru naravnih vlaken izkaže za velik problem, saj je težko dobiti že tako navaden podatek kakor je gostota. Navedena gostota izdelovalca velja samo za vlakna, ki jih dobimo ob dostavi, vendar pa je na mikroravni okoli njih veliko praznega prostora, ki se seveda ob mešanju s polimerom zapolni. Zato je bilo treba meriti gostoto in prostornino kompozita. Ob poznavanju gostote osnove lahko potem s sklepnim računom izračunamo gostoto posameznega vlakna ter od tu po enačbi (14) prostorninski delež vlaken, ki ga potrebujemo za izračun po PMC:

2.2 Tensile testing and the definition of mechanical and geometrical fibre properties

The material was uniaxially loaded with stress in the x_i direction and then the axial effective stress $\sigma_{ef} = \overline{\sigma}_{11}$ and the effective strain $\varepsilon_{ef} = \overline{\varepsilon}_{11}$ in the composite were measured. Tensile testing was then performed at a strain speed of 3 mm/min. The axial Young's modulus E_A was calculated from the inclination of the $\varepsilon_{ef} - \sigma_{ef}$ curve according to the standard for polymer materials ISO 527-1.

To apply the GMCs, precise data about the input materials have to be known, which can be a problem with natural fibres. Even data as trivial as the density is hard to obtain. The density specified by the manufacturer only applies to received fibres in bulk packaging; it does not take into account the micro-voids of space around the fibres, which are then of course filled with matrix. Consequently, the density and the volume of the composite needed to be measured. From this data the density of an individual fibre can be derived. The expression for the fibre-volume ratio is then:

AC:

$$v_f = \frac{V_f}{V_f + V_m} = \frac{\frac{m_f}{1 - m_f} \cdot \frac{\rho_m}{\rho_{com}}}{1 + \frac{m_f}{1 - m_f} \cdot \frac{\rho_m}{\rho_{com}}}$$
(14),

 V_f in m_f pomenita prostornino in maso vlaken, V_m označuje prostornino osnovnega materiala, medtem ko ρ_m in ρ_{com} pomenita gostoto osnove in gostoto kompozita. Poleg prostorninskega deleža je treba

where V_{f} and m_{f} are the volume and the mass of the fibres, V_{m} denotes the matrix volume and ρ_{m} and ρ_{com} denote the matrix and the composite density. In addition, the volume-fraction aspect ratio between the length and



Sl. 2. Prikaz PS s 30 % celuloznih vlaken, povečava 200X, optični mikroskop Fig. 2. PS with 30 % of cellulose fibres, magnification 200X, optical microscope



 S1. 3. PS + 30 % celuloznih vlaken, posneto z elektronskim mikroskopom, povečava 4000X
 Fig 3. PS with 30 % of cellulose fibres, scanned with an electron microscope, magnification 4000X

za izračun poznati tudi dolžino vlaken in razmerje med dolžino in premerom vlaken. Ker se vlakna zaradi trenja ob vrtenju polža lomijo, smo njihovo dolžino ugotavljali z optičnim in elektronskim mikroskopom. Povečave z optičnim mikroskopom so bile 40X in 200X (sl. 2), z elektronskim pa 500X, 1250X, 2500X ter 4000X (sl. 3). PMC ne uporablja razsežnih enot, zato se mora dolžina vlaken izraziti z razmerjem $a_r = l_f / d_f$ med dolžino vlakna l_f in premerom d_r .

3 IZRAČUN PO PMC IN PRIMERJAVA S PREIZKUSOM

V tem poglavju so opisane elasto-plastične lastnosti kompozita, izračunane z GMC ter primerjane z eksperimentalnimi podatki. Primerjava bo pokazala, ali lahko metoda celic dovolj natančno simulira dejanski potek obremenitve biopolimernih kompozitov. Za preračun po PMC smo si pomagali s programsko kodo MAC/GMC 4.0, razvito v NAS-inem raziskovalnem centru Glenn (Glenn Research Center) in v vesoljskem inštitutu Ohaja (Ohio Aerospace Institute), ki uporabnika razbremeni reševanja velikega števila linearnih enačb, ki nastanejo ob analizi kompozita. Na sliki 4 je prikazana primerjava med potekom dejanskega in teoretično izračunanega nateznega preizkusa, dobljenega s PMC.

Najtežje je bilo določiti mehanske lastnosti celuloznih vlaken, saj ti podatki za posamezna vlakna ne obstajajo, prekratka so, da bi z njimi opravili npr. natezni preizkus za posamezno vlakno, kar lahko storimo pri daljših vlaknih ([8] in [9]). Kot izhodiščne vrednosti so bile uporabljene mehanske lastnosti mehkega borovega lesa ([10] in [11]), iz katerih je prečiščena surova celuloza. Vendar pa ne smemo pozabiti, da je bila naša celuloza še toplotno obdelana, kar se je še posebej poznalo pri kompozitu na podlagi PS, saj je tu prišlo do opaznih razlik med preizkusom in izračunom, ki je vedno podal večje vrednosti. Iz tega lahko sklepamo, da je bila toplotna obremenitev vlaken tako visoka, da so se spremenile mehanske lastnosti. Po zmanjšanju elastičnega modula s 6.000 MPa na 2.300 MPa je prišlo do zadovoljivega ujemanja med teoretičnimi izračuni in izmerjenimi podatki pri vseh različnih dolžinah in prostorninskih deležih, zato lahko upravičeno sklepamo, da je to dejanski elastični modul.

Obnašanje vhodnih materialov v plastičnem področju smo simulirali s klasičnim the diameter of an individual fibre needs to be known. Since fibres break down due to the friction induced by the rotation of the screw, their length needs to be assessed with an optical and an electron microscope. The magnifications for the optical microscope were 40X and 200X (Fig. 2), and with the electron microscope, 500X, 1250X, 2500X and 4000X (Fig. 3). The GMCs does not use dimensioned units, which is why the fibre length is expressed by a ratio $a_r = l_f/d_f$ between the fibre length l_r and its diameter d_r .

3 CALCULATION ACCORDING TO THE GMCS AND A COMPARISON WITH THE EXPERIMENT

In this section the elasto-plastic properties of composites are calculated with the GMCs and then compared to the experimental data. The comparison will show if the GMCs can simulate, accurately enough, the loading process of woodplastic composites. For the calculation according to the GMCs, the program code MAC/GMC 4.0 developed by the NASA I Glenn Research Center and the Ohio Aerospace Institute was used. This code can relieve the user of having to solve multiple linear equations, which result from analysing the composite. In Fig. 4 the comparison between the real and the simulated (obtained by the GMCs) tensile test is presented.

The hardest part was determining the mechanical properties of cellulose fibres, due to non-existent data for the individual fibres because they are too short to perform a single filament test that can be performed with longer fibres ([8] and [9]). The reference values were the mechanical properties of pine soft-wood ([10] and [11]), from which the cellulose was refined. But one must not forget that in the observed case the cellulose was also heat treated, which had a significant impact, especially on the fibres mixed with the PS. Here, the initial testing revealed a substantial difference between the simulation and the real tests. The results of the simulation were always higher, so the Young's modulus had to be reduced from 6,000 to 2,300 MPa. After doing this, the alignment of the curves was satisfactory for all the fibre length and volume fractions, so it can be concluded with reasonable confidence that this is a realistic Young's modulus.

The behaviour of the input materials in the plastic area was simulated by the classical



Preglednica 3.	Vstopni	parametri	testiranih	materialov
Table 3. Input	paramet	ers of teste	d materia	ls



prirastkovnim plastičnim modelom [6], v katerem krivuljo, ko napetost preseže mejo elastičnosti pa do porušitve, ponazorimo z nizom premic, kar močno poenostavi računanje, saj lahko na vsaki premici posebej računamo z običajno, linearno teorijo. V preglednici 3 so predstavljeni vstopni podatki za celulozna in lesna vlakna ter PP in PS. V obeh primerih krivulja, dobljena z PMC, zelo dobro sledi eksperimentalni krivulji (sl. 4), razlikujeta se le v točki porušitve, saj v resnici material še nekaj časa teče. To lahko pripišemo matematičnemu modelu PMC, ki določa, da je med osnovo in vlakni popolna vez in da do porušitve lahko pride samo znotraj določene podcelice $\alpha\beta\gamma$ glede na Tsai-Hillov kriterij po enačbi (13), ne pa tudi na meji med dvema materialoma. Na sliki 3 se lepo vidi praznina, ki ostane za izpuljenim vlaknom, kar pomeni da ni popustilo vlakno, ampak vez med vlaknom in osnovo. Primerjava

incremental plasticity model [6], where the curve from the yield stress to the breakage is represented by a series of linear lines. This simplifies the calculation, since the linear theory can now be used. In Table 3, the input data for cellulose and wood fibres, PP and PS, are presented. In both cases the curve obtained by the GMCs matches the experimental curve (Fig. 4) very well. The only difference is at the breaking point, where in reality the strain of the composite is much bigger. This can be attributed to the mathematical model of the GMCs, which anticipates perfect bonding between the matrix and the fibres. The failure can, in this fashion, occur only within a certain subcell $\alpha\beta\gamma$, according to the Tsai-Hill failure criterion, as described in Equation (13), and not at the interface of the two materials. In Fig. 3, the void that remains after the fibre has been pulled out is clearly shown. This means that the fibre did not break. Instead,

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Sl. 5. Prikaz poteka natezne trdnosti v odvisnosti od prostorninskega deleža celuloznih vlaken za PP (a) in PS (b), ----- PMC, ---- preizkus





Sl. 6. Prikaz velikosti modula elastičnosti v odvisnosti od prostorninskega deleža celuloznih vlaken za PP (a) in PS (b), ----- PMC, ----- preizkus

Fig. 6. Presentation of axial Young's modulus in relation to the fibre-volume ratio of PP (a) and PS (b), ----- GMCs, ----- experiment

rezultatov pri drugih kompozitih nas pripelje do enakih sklepov. Za potrditev uporabnosti modela PMC je bilo predvsem treba preveriti njeno odstopanje glede na preizkus pri različnih the bond between the matrix and the fibre failed to carry the stresses any longer. When the results for the other mixtures are compared, the same conclusion can be drawn. To confirm the applicability of the





(----- GMCs, ---- experiment)

prostorninskih deležih ter dolžinah vlaken. Na slikah 5 do 7 je prikazano spreminjanje bistvenih mehanskih lastnosti v odvisnosti od prostorninskega deleža celuloznih vlaken v kompozitu.

Iz grafov (sl. 5) je razvidno, da povečevanju deleža vlaken PMC sledijo eksperimentalno pridobljeni rezultati, saj sta si oba poteka krivulj zelo podobna, razlikujeta pa se v nekaterih končnih vrednostih, pri natezni trdnosti je največji odstopek okoli 15 %. Do večjih razlik lahko pride le pri čistem polimeru brez vlaken, simuliranje katerega pa ni namen metode celic. Še do večjega ujemanja pride pri merjenju vzdolžnega modula elastičnosti, kjer se krivulji skoraj prekrivata (sl. 6). Iz tega in grafov na sliki 4 lahko sklepamo, da PMC zelo dobro simulira obnašanje biopolimernih kompozitov v elastičnem področju, medtem ko v plastičnem področju, predvsem ko se material bliža porušitvi, daje manj natančne rezultate. To se še potrdi v primerjanju dejanskega in izračunanega največjega raztezka (sl. 7). Tu se zopet potrdi ugotovitev, da so usmerjenosti krivulj različnih mešanic podobne, vendar pride med PMC in preizkusom do prevelikih razlik v absolutnih vrednostih (do 37 %). Do teh razlik pride, ker PMC določa popolno vez med vlaknom in osnovo, medtem ko v resnici pride do popustitve vezi med površinama, kar zmanjša strižne sile v materialu,

GMCs, its deviation from the experiment at various fibre-volume ratios and fibre lengths had to be tested. The charts in Fig. 5 to 7 represent the changing of the main mechanical properties in reference to the volume ratio of cellulose fibres in the composite.

From Fig. 5 it is evident that by increasing the fibre data the trends of both curves are very similar. The difference exists only in the end values, where the ultimate stress can differ by up to 15%. Greater errors appear only for a pure polymer without fibres, but these conditions are not meant for the GMCs to simulate. Even greater matching appears when the axial Young's modulus is measured, where the curves are almost interlacing (Fig. 6). From this and from the charts in Fig. 4 it can be inferred that the GMCs simulates the behaviour of the woodplastic composites very well in the elastic area, while in the plastic area, especially near the failure, the results are less accurate. This is also confirmed when comparing the simulated and the real ultimate strain (Fig. 7). Here, the tendencies of the curves of different compounds are similar, but there are great differences between the GMCs and the experiment in terms of the absolute values (up to 37 %). These differences occur because the GMCs anticipates perfect bonding between the fibre and the matrix, while in reality failure occurs at the interface of two surfaces. This reduces the shear forces in the material, which enables further deformation of the composite. Apart





to omogoči, da se kompozit še naprej deformira. Poleg prostorninskega deleža vlaken v kompozitu ima na njegove mehanske lastnost velik vpliv tudi dolžina vlaken oz. razmerje med dolžino in premerom a_i . Tudi tu se jasno pokaže učinkovitost PMC, saj lepo napove izboljšanje mehanskih lastnosti z daljšanjem vlaken (sl. 8 in 9).

4 SKLEP IN RAZPRAVA

Prispevek je osredotočen na analizo zmožnosti posplošene metode celic določiti elasto-plastični odziv biopolimernih kompozitov. Analizirali smo kompozite z osnovo iz PP ali PS, dodali pa smo jim celulozna ali lesna vlakna različnih dolžin. Izkazalo se je, da po pridobitvi ustreznih vrednosti za lastnosti vlaken (njihovo neposredno merjenje je nemogoče), lahko PMC zelo dobro napove obnašanje kompozita. Izračunane vrednosti vzdolžnega modula elastičnosti (enoosna obremenitev) ter natezne trdnosti so bile v področju merjenja, to je pri masnem deležu vlaken med 20 % in 60 %, zelo blizu dejanskim. Večje razlike pa so se pojavile pri primerjanju raztezkov, saj je v resnici prišlo pri vseh mešanicah do bistveno večjih raztezkov, kot jih je pokazal izračun. To lahko pripišemo že prej omenjeni popolni vezavi med vlaknom in osnovo, ki jo predvideva pri svojem računanju PMC.



 Sl. 9. Vpliv dolžine vlakna na modul elastičnosti kompozita za PS (----- PMC, ---- preizkus)
 Fig. 9. Influence of fibre length on axial Young's modulus (----- GMCs, ---- experiment)

from the fibre-volume ratio, the length, i.e., the fibre aspect ratio between the length and the diameter a_r , also has a great impact on the mechanical properties. The effectiveness of GMCs is demonstrated here because it clearly predicts the improvement in mechanical properties with the lengthening of the fibres (Figs. 8 and 9).

4 CONCLUSION AND DISCUSSION

This paper focuses on an analysis of the applicability of the generalised method of cells to predict the elasto-plastical response of wood-plastic composites. Composites with PP and PS matrixes and cellulose or wood fibre inclusions of various lengths were analysed. It was shown that after obtaining appropriate values for the fibre properties (their direct measurement is impossible), the GMCs can predict with great accuracy the trends of composite behaviour. The calculated values of the axial Young's modulus (unidirectional load) and the tensile strength were, in the measured range, very close to real values (for fibre volume ratios from 20 to 60%). Greater differences occurred when measuring the strain. In reality failure occurred at much larger strains than presented in the simulation. This can be attributed to the perfect bonding between the matrix and the fibre that is anticipated by the GMCs.

Ob vsem tem se postavlja vprašanje, ali lahko v praksi uporabimo PMC za napoved mehanskega odziva biopolimernih materialov. Primerjava rezultatov preizkusov in izračuna pokaže, da lahko, vendar z omejitvami. Problemi se pojavijo predvsem pri nepoznavanju lastnosti celuloznih vlaken, saj ti izgubijo del svoje trdnosti pri toplotnem obremenjevanju med postopkom, zaradi vrtenja polža v valju brizgalnega stroja pa se vlakna tudi skrajšajo. Daljša so vstopna vlakna, večje je zmanjšanje dolžine. Da bi se izognili vsakokratnemu mikroskopiranju izbrizganega materiala, lahko najdemo približno skrajšanje vlaken v nekaterih virih [12]. PMC tudi ne upošteva naključne usmerjenosti ter predvsem zvitosti ter neenakomernega prereza vlaken, kar tudi zmanjšuje njeno učinkovitost pri napovedovanju odziva biopolimernih kompozitov. V nadaljnjih testiranjih bi bilo treba tudi upoštevati nepopolno vez med osnovo in vlakni, kakor je to naredil Bednarcyk [13] za kovinska vlakna ter vpliv dodatkov za izboljšanje vezi med osnovo in vlakni [1]. Vzpostaviti bi bilo treba tudi nabor podatkov z mehanskimi lastnostmi različnih vrst naravnih vlaken, saj povprečni uporabnik v industriji zelo težko pridobi ustrezne vhodne podatke. Prav tako v prispevku niso bile upoštevane časovno odvisne mehanske lastnosti materiala zaradi zapletenosti in naključnih lastnosti vlaken, kar pa bo predmet prihodnjih raziskav. Razvoj PMC tudi še ni končan, prav tako pa se biopolimerni kompoziti šele uveljavljajo, tako da lahko v prihodnosti pričakujemo dodatke k PMC, ki bodo upoštevali specifične lastnosti naravnih vlaken.

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All this raises the question of whether the GMCs can be used to predict the mechanical response of woodplastic composites. The comparison of the experimental and calculated results shows that the answer is yes, but with limitations. Problems arise because the properties of the cellulose fibres are not known. They lose a part of their strength while they are heat treated during the processing. A reduction in the length also occurs due to the screw rotation in the cylinder of the injectionmoulding machine. The longer the input fibres are, the greater the reduction is. To avoid repetitive microscopy of the injected materials, an approximate fibre-length reduction can be found in some papers [12]. The GMCs does not take into account the random fibre orientation and especially their curved shapes and non-uniform cross-section, which also reduces its capability to predict the response of wood-plastic composites. In further testing the imperfect bonding between the fibre and the matrix should be taken into account, as was done by Bednarcyk [13] for metal fibres. The impact of coupling additives should also be taken into account[#] here [1]. A database with the mechanical properties of different natural fibres should also be established. because presently an average industrial user obtains the correct input data with great difficulty. Also, in the paper, the time-dependent mechanical behaviour of the composite was not yet considered due to complex and random properties of the fibre, and this will be the subject of our future research. Moreover, the development of the GMCs is not yet finished, and woodplastic composites have just only begun to take their place among other composites. This means that in the future, improvements of the GMCs can be expected, which will take into account the specific properties of the natural fibres.

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