LOW VELOCITY IMPACT RESPONSE OF POLYPROPYLENE BIOCOMPOSITES REINFORCED WITH MAN-MADE CELLULOSE AND SOFT WOOD FIBRES

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Abstract. The study presents an experimental investigation of the impact response of hybrid biocomposites that are based on polypropylene homopolymer HP400R reinforced with man-made cellulose fibres and cellulose microfiller. Man-made cellulose fibres, such as rayon tire cord yarn (Cordenka) and soft wood microfibres (Weho 500) were added to the polypropylene matrix with different weight contents. Composite specimens were manufactured by injection moulding. Absorbed energies and maximum impact forces of biocomposite were determined for each filling ratio of Cordenka and Weho 500 and have been selected as the parameters of the study. It was performed by the method of planning of the experiment and response surface method. Results showed that combination of two types of fillers improved impact properties. Response surface plots reveal that energy and maximum load of biocomposites increases with increasing content of reinforcements, however, the man-made cellulose Cordenka fibres have dominant influence on the obtained results. An addition of the Cordenka fibres increased the absorbed energy by ~372 % and ~243 % in comparison with the unreinforced polypropylene matrix and composite reinforced with 30 wt. % of Weho fibres. The substitution of Cordenka fibres with soft wood fibres gave increase only by ~72 % and ~27 % in comparison with the unreinforced polypropylene matrix and composite reinforced with 30 wt. % of Cordenka fibres.

Keywords: impact, hybrid, composite, softwood, cellulose, fibre.

Introduction

One of the main challenges that is faced nowadays is environmental protection. New material production strategies require creation manufacturing of new materials that cause minimal harm to the nature. Biocomposites belong to these materials [1-2].

They are usually a substitute for the standard composites reinforced with synthetic fibres (aramid, glass and carbon fibres). Composite materials reinforced with synthetic fibres have high strength, good wear resistance, reliability, high fatigue life, and a number of other desired properties. However, in comparison with natural composites, they have a number of serious disadvantages, such as high cost, and non-biodegradable properties [3-4].

The use of natural fibres or biobased fibres (plants used as a source for cellulose) with polymer matrices is characterized by such advantages as low density, renewability, and sometimes biodegrability, which are the reasons of their eco-friendliness. Currently, biocomposites are the subject of extensive research, especially in the field construction industry, by virtue of such advantages as low weight and low manufacturing costs. Products made of biocomposite not only lower the cost of the product, but also reduce the cost of its subsequent disposal [5-6].

In order to reduce the material cost of biocomposites their fibrous reinforcement can be combined with low-cost microfibres. This hybrid combination of reinforcement can still preserve some essential properties of fibrous composite and improve the processing. This hybridisation of reinforcement can reduce the cost of the composite and even improve its performance and mechanical and physical properties [7]. The development of new generation materials with the required properties promotes great interest, since the created materials have improved mechanical properties and ensure new possibilities for their applications [8].

Thus, the use of natural fibres as reinforcing materials in polymer composites for the production of new materials has promoted great interest in recent years. The use of natural fibre provides a new impetus for study of new materials and the development of new technological processes.

Polymer composites reinforced with wood fibres have been one of the key subjects of the research in recent years. Wood fibres are lignocellulosic natural fibres that are widely used for reinforcing various polymer matrices (PE, PP, and PVC). They are divided into two types – softwood and hardwood. Softwood fibres are obtained from soft conifers and hardwood - from hard deciduous trees, respectively [9-10].

The choice of polymer as a matrix material is important for the reinforcement of the cellulose fibres. The use of polypropylene as matrix material is favoured by virtue of its low density, flexibility

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and low price compared to other polymers. Reinforcing of polypropylene by improvement of its mechanical properties offers new possibilities for engineering applications [11].

Combination of man-made cellulose fibres and softwood flour in reinforcement can improve mechanical properties. Man-made cellulose fibres (which are mostly rayon viscose) are produced from natural resources (wood pulp) and constitute of biodegradable cellulose. In some applications they have potential to become a substitute of common glass fibre reinforcement. The man-made cellulose fibres are an important material in polymer industry owing to their stability of properties, biobased origin and better recyclability and disposal than glass fibres. They are also excellent raw material for the production of various materials in building industry [12-17].

The use of hybrid composites as structural materials due to their early stage of development requires extensive testing before their practical application. Some types of damages occurring during the end-product exploitation are very dangerous, because they cannot be detected visually and as result they can limit application of particular materials. Thus, in addition to tensile, bending or hardness testing, impact tests are being also carried out. Mostly for impact testing of hybrid composites for structural applications the Charpy and Izod tests are used, which are low-velocity impact tests. Additionally drop-weight (falling dart) can be used to characterise the damage behaviour on broader spectrum of velocities and energies in these composites, which gives more insight in their impact properties [18-20].

The main purpose of this study was investigation of the man-made cellulose fibres (Cordenka) and softwood fibres (Weho 500), as well as their hybrid combination on the impact behaviour of the reinforced composites based on polypropylene matrix. Absorbed energies and maximum impact forces during puncture of composite were determined for each filling ratio of Cordenka and Weho 500 and have been selected as the parameters of this study. The overall research approach was performed by the method of planning of experiment and response surface method. This research shows that the relatively costly man-made cellulose fibres may be partially substituted with Weho fibres, which results in improved impact properties.

Materials and methods

Hybrid composites were prepared at the Department of Material Technology, West Pomeranian University of Technology in Szczecin, Poland. PP Moplen HP400R granulate produced by Basell Orlen Polyolefins (Płock, Poland) was used as matrix material. This material is used for injection moulding applications. The man-made cellulose fibres Cordenka with a cut length of ~1.7 mm and a fibre diameter ~18 μm were supplied by Cordenka (Gorzow, Poland). Soft wood fibres (Weho 500) were supplied by Jelu-werk Ludwigsmühle, Rosenberg, Germany. The cutting was done by Ekotex company, Namysłów, Poland.

Cordenka and Weho 500 fibres were dried before compounding at 103°C in an air circulating oven before mixing [16]. Then the fibres and polypropylene were compounded at temperature from 150 to 200°C and rotation speed of the screw of 40 RPM. Then extruded hot strands were cooled to room temperature and cut into granule. Finally, specimens for impact test were prepared from these granulate compounds by the injection moulding process at temperature 160 - 180°C and under injection flow of 20ccm/s.

The low velocity impact tests were performed according to ISO 6603 and using a testing machine INSTRON Dynatup 9250 HV Impact Tower. The dimensions of specimens are 60 mm × 60 mm with 2 mm thickness. The total mass of the impactor (including striker and drop weight framework) was 5.83 kg. Diameter of striker is 25.4 mm with a hemispherical type. The drop height of 0.39 m gave a kinetic energy of 22 J. The corresponding velocity of the striker at the moment of impact was 2.61 m/s. The impact tests were conducted at room temperature. Ten specimens were tested at each filling ratios of Cordenka man-made cellulose fibres, Weho 500 soft wood fibres and their combination. During the experiment the specimens were clamped between supporting and clamping rings.

Fig. 1 shows the impact response of the specimen. It includes energy (energy that the specimen absorbed during the impact test), displacement between the striker and specimen support (deflection) and the force exerted by the strike (load) (Fig. 1).

Full factorial design was used to study the influence of experimental variables on the responses: the absorbed energy and maximum impact force occurring during the test. 16 experimental runs for two factors and four levels have been selected. The full factorial design is a design strategy, where researchers measure responses at all combinations of the factor levels [21]. Experimental variables and their levels are reported in Table 1.

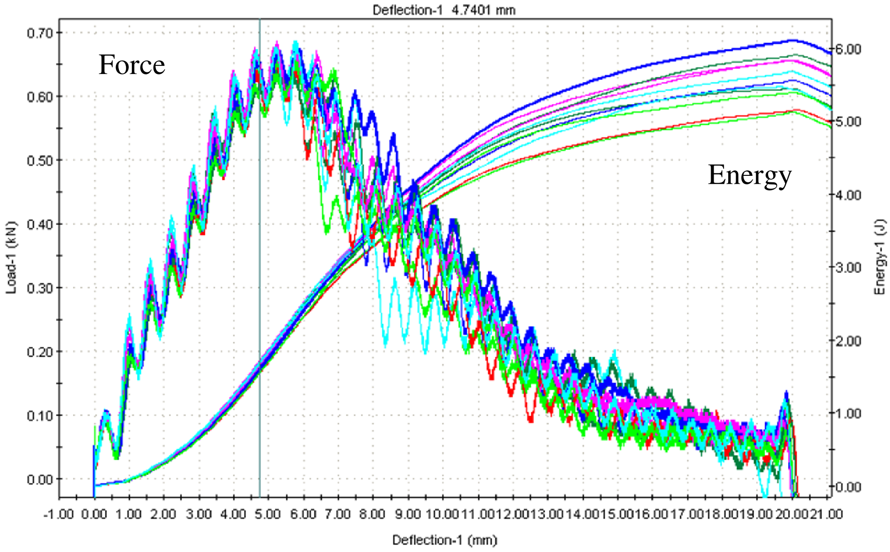

Fig. 1. **Typical low velocity impact response**

Table 1

Experimental variables and their levels

Experimental variables	**Levels**			
Cordenka fibres, wt. %	0	10	20	30
Weho 500 fibres, wt. %	0	10	20	30

A second order polynomial regression equation, as shown in Eq. (1), has been developed to predict the absorbed energy and maximum load during the impact test of composites by correlating the input parameters of the design experiment.

$$\hat{F}(x) = b_0 + \sum_{i=1}^{m} b_i x_i + \sum_{i=1}^{m}\sum_{j=i}^{m} b_{ij} x_i^2 + \sum_{i=1}^{m}\sum_{j=i}^{m} b_{ij} x_i x_j \,, \tag{1}$$

where $F(x)$ – response;
x_i, x_j – the values of factors;
b_0 – constant;
b_i, b_j, b_{ij} – linear, quadratic end interaction coefficients, respectively;
m – number of the factors.

Results and discussion

Absorbed energy and maximum impact force has been measured using 10 specimens of biocomposites and average result value is reported in Table 2. Values given with standard deviation. The experimental data obtained by the low-velocity impact testing in the points of design experiments have been used to build the approximating functions by using the program VariReg software [22].

Analysis of variance (ANOVA) was applied for estimation of significance of the second – order models. Using a 5 % significance level, a model was considered significant, if the $p < 0.05$. The model of energy and maximum load less than 0.05 that indicated that model is significant. The results of model summary statistics showed the R^2 value more than 0.9 (0.995 and 0.980 for absorbed energy and maximum impact force, respectively) for both response models, that indicated a good correlation between the experimental and predicted responses. The predicted R^2 value (0.987 and 0.980 for energy and maximum force) in good agreement with the adjusted R^2 value (0.993 and 0.971 for energy and maximum force) indicated reliability of models. Relationship between the experimental variables x and the corresponding behaviour functions Y is given as follows.

Absorbed energy:

$$Y_E = 1.24245\text{-}1.293\cdot 10^{-2}\cdot C + 4.7395\cdot 10^{-2}\cdot W + 4.95\cdot 10^{-3}\cdot C^2\text{-}7.875\cdot 10^{-4}\cdot W^2 + 8.22\cdot 10^{-4}\cdot W\cdot C, \quad (2)$$

Maximum force:

$$Y_F = 0.3987 + 2.033\cdot 10^{-2}\cdot C + 9.5075\cdot 10^{-3}\cdot W\text{-}9.375\cdot 10^{-5}\cdot C^2\text{-}8.125\cdot 10^{-5}\cdot W^2\text{-}2.53\cdot 10^{-4}\cdot W\cdot C, \quad (3)$$

where C is the weight contents of reinforcement Cordenka fibres;
W is the weight contents of reinforcement Weho 500 fibres.

Table 2

Experimental variables with experimental and predicted responses

Runs	Experimental variables		Responses			
	Cordenka fibres, wt. %	Weho fibres, wt. %	Absorbed energy, J		Maximum impact force, kN	
			Experimental	Predicted	Experimental	Predicted
1	0	0	1.14 ± 0.09	1.24	0.39 ± 0.04	0.40
2	0	10	1.54 ± 0.18	1.64	0.46 ± 0.09	0.49
3	0	20	1.88 ± 0.31	1.88	0.53 ± 0.04	0.56
4	0	30	1.97 ± 0.24	1.96	0.66 ± 0.08	0.61
5	10	0	1.77 ± 0.21	1.61	0.63 ± 0.03	0.59
6	10	10	2.30 ± 0.25	2.09	0.68 ± 0.03	0.65
7	10	20	2.56 ± 0.11	2.41	0.70 ± 0.01	0.70
8	10	30	2.58 ± 0.09	2.57	0.70 ± 0.02	0.73
9	20	0	2.88 ± 0.36	2.96	0.76 ± 0.05	0.77
10	20	10	3.37 ± 0.32	3.52	0.81 ± 0.03	0.80
11	20	20	3.84 ± 0.63	3.93	0.82 ± 0.03	0.82
12	20	30	3.95 ± 0.42	4.17	0.80 ± 0.04	0.83
13	30	0	5.38 ± 0.39	5.31	0.90 ± 0.04	0.92
14	30	10	5.85 ± 0.59	5.95	0.94 ± 0.04	0.94
15	30	20	6.50 ± 0.35	6.44	0.95 ± 0.03	0.93
16	30	30	6.91 ± 1.15	6.76	0.92 ± 0.04	0.91

After selection of equation of regression the parametric studies were carried out additionally to scrutinize the influence of the experimental variables on behaviour functions. This was done by displaying 3D graphs of approximating functions. The results obtained for the absorbed energy and maximum force with different filling ratio of the manufactured composites are presented in Fig. 2 and Fig. 3, respectively.

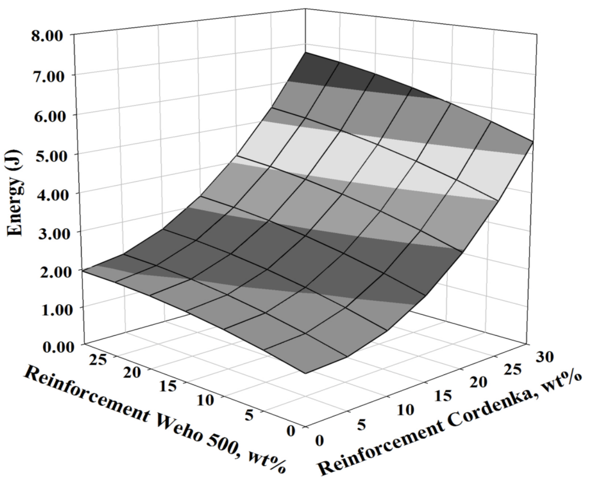

Fig. 2. **Dependency on the absorbed energy**

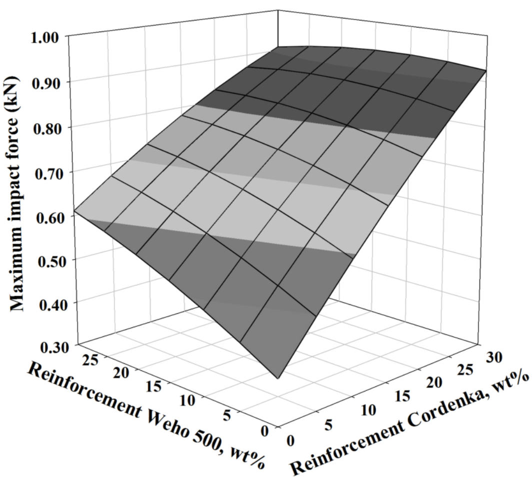

Fig. 3. **Dependency on the maximum force**

Fig. 2 shows the dependency of the absorbed energy on filling ratios of Cordenka man-made cellulose fibres, Weho 500 softwood fibres and their combination. It is observed that absorbed energy of composites increases with an increase of the reinforcement filling ratio. The greatest improvement was obtained with Cordenka fibres. When soft wood fibres were absent in the polymer matrix, the maximum energy reached 5.38 J. An addition of the Cordenka fibres in the composite increased the absorbed energy by ~372 % in comparison with the unreinforced polypropylene matrix. On the other hand, the reinforcing only with soft wood fibres had the smallest influence on the energy – 2 J. The substitution of Cordenka fibres with soft wood fibres gave increase only by ~72 %.

In hybrid composite reinforced with man-made cellulose fibres and soft wood fibres at 30 wt. %, the absorbed energy achieved the maximum of 6.76 J. Values of absorbed energy were increased by ~243 % in composite reinforced with 30 wt. % of softwood flour and by ~27 % in composite with 30 wt. % of man-made cellulose fibres.

Additional investigations were carried out to evaluate the influence of reinforcements on maximum load. The results obtained for maximum load for different filling ratios of the manufactured composites are presented in Fig. 3.

It can be seen that the values of maximum load significantly increase with an increase of the filing ratio of Cordenka man-made cellulose fibres. In the polymer matrix the addition of 30 wt. % of sofwood flour gave further improvement of the maximum impact force by 53 %. For the composite reinforced with 30 wt. % of Cordenka fibres the addition of 30 wt. % of sofwood flour gave insignificant improvement of the maximum impact force by 5 %.

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Conclusions

1. The effect of Cordenka man-made cellulose fibres, softwood fibres Weho 500 and their combination on the impact responses of the composites based on polypropylene matrix was investigated. The study was performed by the method of planning of the experiment and response surface method.
2. The maximum absorbed energy was obtained for the composite reinforced with highest filling ratio of Cordenka man-made cellulose fibres and softwood fibres Weho – 6.76 J. An addition of Cordenka fibres and Weho fibres in the composite increased the absorbed energy by ~493 % in comparison with the unreinforced polypropylene matrix
3. Addition of Cordenka fibres in the composite reinforced by softwood fibres Weho 500 decreases the influence on the impact force. In the hybrid composite reinforced with Cordenka fibres at 30 wt. %, the maximum impact force increases insignificantly with the increase of softwood fibres Weho 500 – 5 %.

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