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POLYMER SCIENCE

Influence of hollow glass microspheres on the mechanical and physical properties and cost of particle reinforced polymer composites

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Abstract. The goal of the study was to find a cost-effective composition of a particle reinforced composite that is light in weight but has sufficient mechanical properties. The matrix of the particulate composite is unsaturated polyester resin that is reinforced with alumina trihydrate particles. Part of the alumina trihydrate proportion was replaced with hollow glass microspheres to reduce weight and save costs. In order to find out the influence of the light filler on the physical and mechanical properties of composites, materials with different percentages of the light filler were prepared. Test specimens were cut from moulded sheets that were fabricated with vacuum assisted extruder. Tensile strength, indentation hardness measured with a Barcol impressor, and density were determined. Based on the experimental data a multi-criteria optimization problem was formulated and solved to find the optimal design of the material. Artificial neural networks and a hybrid genetic algorithm were used. The optimal solution is given as a Pareto curve to represent the distinction between the density and selected mechanical properties of the composite material. The composite material filled with 6% hollow glass microspheres showed 3% loss in the tensile strength and 26% loss in the surface hardness compared to the composition without the filler. The weight decreased by 13% compared with the initial composition. The addition of hollow glass microspheres did not lower the net value of the material, it increased 7%.

Key words: materials engineering, particle reinforced composites, particulate composites, multi-criteria optimization, hollow glass microspheres, polymer matrix composites, light-weight composites.

INTRODUCTION

Particulate composite material consisting of unsaturated polyester resin (UP) and alumina trihydrate (ATH) shows good mechanical and physical properties. The material is used for fabricating counter tops, sanitary ware, and furniture. The use of the material is limited in some applications because of its weight and cost. Lighter weight would decrease transportation costs, enable to produce larger products, and make handling and installation easier. Several solutions have been used to overcome these problems. A variety of lightweight fillers have been added to particulate composites in order to achieve weight savings. For example polymethylmetacrylate (PMMA) powder [1], hollow glass microspheres [2], and silicate based lightweight particulate fillers [3] have been used.

Microscopic hollow soda–lime–borosilicate spheres are low-density particles that are used in a wide range of industries. The hollow microspheres are used to reduce warpage and shrinkage, to adjust the rheological properties, to reduce part weight, and to lower cost. On the other hand, the increased concentration of lightweight particulate reinforcement such as micro balloons can aggravate the mechanical properties of the particle filled composite material [4]. If the filler is less resistant to penetration than the thermoset matrix, the filler will lower the indentation hardness of the composite [5].

The goal of the current study was to find a cost-effective material composition that has low density but sufficient mechanical properties.

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The procedure developed for solving the posed problem includes:

- design of the experiment;
- an experimental study for determining mechanical and physical properties of the material prepared with different filler ratios;
- response modelling;
- multi-criteria optimization;
- cost calculation.

The evaluation of the objective functions from experimental data is time consuming, too expensive, and thus not reasonable. A common technique for reducing the need for time resources and minimizing cost in optimal design problems is to use surrogate models for the approximation of the objective and constraint functions.

A number of surrogate model types are available, with applications in different areas ranging from sociology to space [6,7]. The most widely used types of surrogate models include Artificial Neural Networks, Radial Basis Function models, Rational Functions, Support Vector Machines, Kriging models, etc. In the current study, artificial neural networks were used for the modelling of response surfaces, corresponding to different mechanical properties of the composite material. Multi-criteria optimization problem was formulated on the basis of analysis of the relationships between mechanical properties of the material. Finally, the solution of the multi-criteria optimization problem was obtained by applying the concept of Pareto optimality and the methodology introduced by the authors in [8,9].

MATERIALS AND METHODS

A polymeric composite is characterized by a number of physical and mechanical properties. The most demonstrative properties to observe the effect of fillers are tensile strength, indentation hardness, flexural strength, and flexural modulus.

In the current study the percentage of the light filler and the matrix to reinforcement ratio were considered as design variables. The design of experiment was performed by varying the values of these parameters. In order to find out the influence of the design variables on the mechanical properties of the composite different material compositions were prepared and tested.

Test specimens were cut from moulded sheets, which were produced with vacuum assisted extruder-type mixing equipment. The vacuum chamber of the machine removes air from the casting dispersion and helps to achieve non-porous material.

Preliminary cure of the composite was done at room temperature (23 ± 2 °C) for 12 h. This was followed by post cure in a conventional thermal oven.

Tensile tests of the composite plastic materials were performed according to standard ISO 527-1:2000. The test specimens were cut from the casted slabs with a waterjet.

The indentation hardness of the material was measured with a GYZJ 934-1 Barcol impressor according to ASTM D 2583 “Standard Test Method for Indentation Hardness of Rigid Plastics by Means of a Barcol Impressor”.

The density of the material was determined by weighing method. A kit of an analytical scale and a weighing jig was used for weighing a specimen of the material in air and fluid. The density was determined by the equation:

$$\rho = \frac{m_{\mathrm{Sa}} \times \rho_{\mathrm{w}}}{m_{\mathrm{Sa}} - m_{\mathrm{Sw}}}, \qquad (1)$$

where m_{Sa} is specimen weight in air, m_{Sw} is specimen weight in fluid, and ρ_{w} is density of the fluid.

MODELLING OF THE PARTICLE FILLED POLYMER COMPOSITE

Let us proceed from a predetermined set of designs obtained from an experimental study. In the following, the output data obtained from the tests are treated as response values. Artificial neural networks are used for surface fitting. The surface constructed by the use of neural networks does not normally contain the given response values (similarity with the least-squares method in this respect).

Based on test data, the relationship between the mechanical properties of the material (output data) and design variables (light filler and matrix to reinforcement ratio) was established.

Analysis of the behaviour of the mechanical properties considered leads to the following conclusions:

- The mechanical properties describing general strength/stiffness characteristics of the material (tensile strength, flexural modulus) have similar behaviour with respect to design variables and can be combined into one group of properties. A reduced order model can be obtained by selecting one of these properties to represent this group (e.g. tensile strength);
- The surface hardness has similar behaviour with mechanical properties responsible for general stiffness/strength characteristics, but its physical meaning is quite different (it is responsible for local characteristics);
- Contradictory behaviour can be perceived between the tensile strength and material density, also between the surface hardness and material density.

To procede form analysis done so far, the multi-criteria optimization problem is formulated on the following bases:

- The three different objectives considered are tensile strength, surface hardness, and material density;
- Two objectives, tensile strength and surface hardness, are combined into one objective employing the weighted summation (or compromise programming) technique.

The relationship between the two combined criteria and material density can be determined by using the concept of Pareto optimality. Such an approach allows obtaining the 2D Pareto front.

According to the weighted summation technique, all the criteria combined are scaled, multiplied by weights, and summed into the general objective f_1 as

$$f_1 = \sum_{i=1}^{m} w_i f_{1i}, \tag{2}$$

where m is the number of the optimality criteria used, w_i is the weight of the i-th criterion and

$$\sum_{i=1}^{m} w_i = 1, \quad 0 < w_i \le 1. \tag{3}$$

In this case $m = 2$ and the scaling of the objectives can be performed as

$$f_{11}(x) = \frac{\max T_S(\overline{x}) - T_S(\overline{x})}{\max T_S(\overline{x}) - \min T_S(\overline{x})},$$

$$f_{12}(x) = \frac{\max S_H(\overline{x}) - S_H(\overline{x})}{\max S_H(\overline{x}) - \min S_H(\overline{x})}, \tag{4}$$

where T_S and S_H stand for tensile strength and surface hardness, respectively, and $\overline{x}$ is vector of design variables. Scaling the third optimality criterion, material density M_T, is a little different:

$$f_2(x) = \frac{M_T(\overline{x}) - \min M_T(\overline{x})}{\max M_T(\overline{x}) - \min M_T(\overline{x})}, \tag{5}$$

because it is subjected to minimization (the first two objectives, tensile strength and surface hardness, are subjected to maximization). As a result, there are two objective functions f_1 and f_2 subjected to minimization and given by formulas (2)–(5).

Thus, the multi-criteria optimization problem can be formulated as

$$f(\overline{x}) = \min(f_1(\overline{x}), f_2(\overline{x})), \tag{6}$$

subjected to linear constraints

$$x_i \le x_i^*, \quad -x_i \le x_{i*}, \quad i = 1, \ldots, n, \tag{7}$$

and non-linear constraints

$$\sigma(\overline{x}) \le \sigma^*. \tag{8}$$

In (7) x_i^* and x_{i*} stand for the upper and lower bounds of the design variables, respectively. The non-linear constraint (8) is imposed on stress σ and requires that it does not exceed the given limit value σ^*.

EXPERIMENTAL RESULTS

One of the most important characteristics of the composite material, its tensile strength, depends on the adhesion strength between the matrix and the reinforcement material [5,10]. The results of tensile tests are shown in Fig. 1. The increase of glass microspheres weight% in the material decreases its tensile strength.

Another important mechanical property, Barcol hardness, is largely influenced by the modulus of the matrix and the filler. The light filler is less resistant to penetration than the matrix and the Barcol hardness decreases as depicted in Fig. 2. The decline of hardness is much more drastic than of tensile strength. Loss of surface hardness means that the wear and scratch resistance of the material will decrease. These are important properties in service.

As expected, the low-density particles have a positive effect on the density of the material. The light filler reduces the density of the composite and thus lowers the mass of the product (Fig. 3).

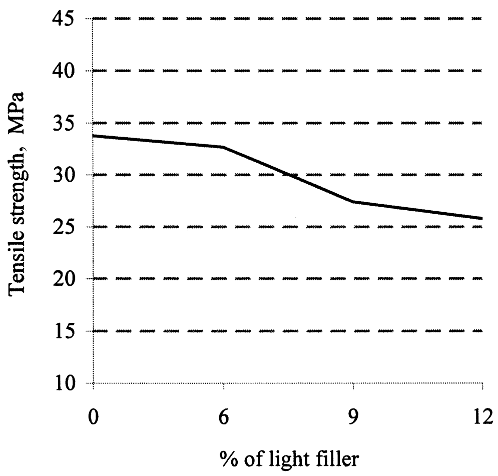

Fig. 1. Tensile strength vs. light filler content.

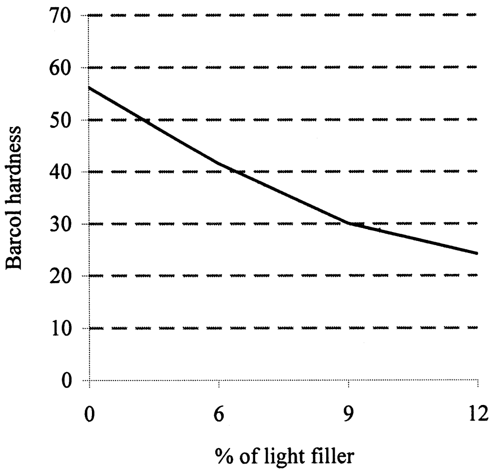

Fig. 2. Barcol hardness vs. light filler content.

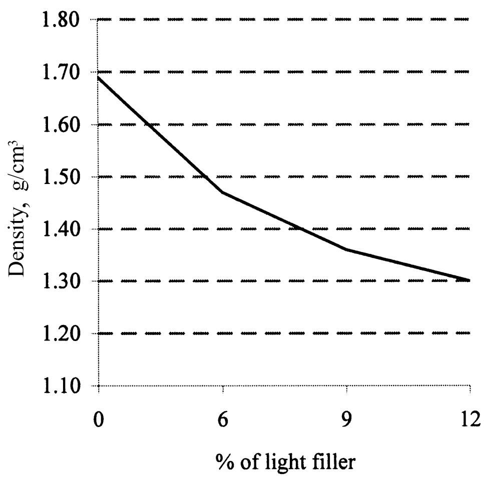

Fig. 3. Density vs. light filler content.

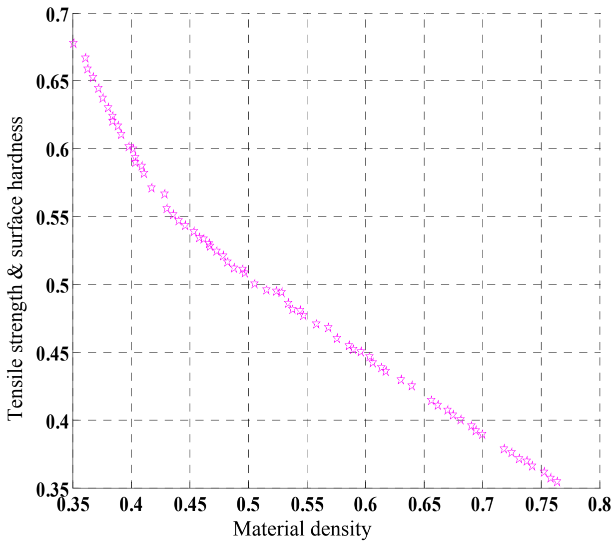

Fig. 4. The combined objective vs. material density.

NUMERICAL RESULTS

Analysis and simplifications done in the section on modelling allow obtaining a reduced order model that contains two optimality criteria. The Pareto front of the combined objectives (2) and material density (5) are given in Fig. 4.

Note that an increase of the values of the combined objective $f_1(\overline{x})$ means that the distance of the tensile strength and surface hardness from their maximal values will increase, thus the values of the tensile strength and surface hardness will decrease.

It can be seen from Fig. 4 that the relationship between these two objectives is described by a Pareto curve consisting of two linear parts (may be approximated as linear). Reducing the material density function $f_2(\overline{x})$ from 0.76 to 0.43 leads to the proportional decrease of the combined function (from tensile strength and surface hardness), further reduction of the material density function leads to much faster reduction of the combined function. The optimal solution for the posed optimization problem can be selected as the intersection point of two linear parts of the Pareto curve, but not necessarily. In general, the selection of an optimal solution is still complicated and depends on a number of factors such as the specific problem considered, additional information available, etc. [11,12].

The Pareto front of the objective functions does contain more information than the physical programming approaches. The shape of the Pareto front provides valuable information.

It should be mentioned that the simplifications made in the modelling section are based on analysis and can be confirmed by simulations with more complex models.

In order to achieve higher accuracy a real-coded genetic algorithm is employed. In a standard formulation the genetic algorithm may converge close to an optimal solution to a not optimal solution. A refined algorithm, hybrid GA has been proposed for design improvement. A global–local approach for the optimization has been employed [13,14]. The hybrid approach used provides a global search with the GA and was subsequently further refined with a gradient-based search. The function approximation and optimization modules are realized in the MATLAB and C++ programming environment.

COST CALCULATION

The cost of the composite material can be calculated based on mass or volume. The composite consists of materials with different densities. Because of that, the mass price and cubic price have a difference when the composition is changed (Fig. 5). Correct net value calculations are based on the cubic price. Hollow glass microspheres cost more than ATH but have much lower density. Therefore it was expected that the mass price would increase but the volume would compensate for it and the net value of the product would lessen.

Unfortunately, the low density and large volume of the light filler did not compensate for the higher price. Nevertheless, the decrease of the mass was larger than the increase of the price. With 6% of light filler the mass reduction was 13.1% but price increase 7.2%. As it was pointed out earlier, the mass of the product influences also other costs, like transportation cost and custom duties. This means that the overall savings from the weight reduction might compensate for the price increase.

CONCLUSIONS

Test specimens of the material with varied composition were fabricated and their properties were determined. A new composite was modelled on the basis of testing results.

Analysis of the candidates for objective functions was performed and a reduced order model was developed. The mechanical properties responsible for global stiffness/strength of the material were grouped and replaced by one representative objective function.

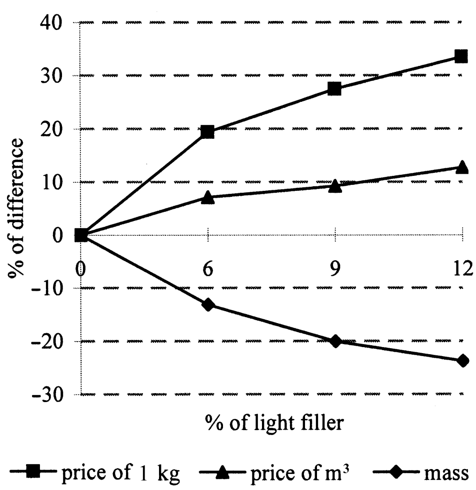

Fig. 5. Influence of the light filler content on the cost and mass.

A numerical procedure was developed for designing the new composite. Artificial neural networks and a real-coded genetic algorithm (GA) were used for modelling the response between objectives and design variables and solving the optimization problem, respectively.

The composite material filled with 6% of hollow glass microspheres showed 3% loss in the tensile strength and 26% loss in the Barcol hardness compared to the composition without the filler. The weight decrease was estimated at 13% of the initial composition. The addition of the light filler increased the net value of the material by 7%.

In conclusion, we can say that it is possible to find a compromise between lower density and loss in mechanical properties when using hollow glass microspheres as an additional filler in a particulate reinforced composite material. Nevertheless, thorough cost calculations should be made. It has to be considered whether the possible savings from the weight reduction compensate for the increase in the net cost of the material.

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Õõnsate klaassfääride mõju osake-armeeritud polümeerkomposiitmaterjali füüsikalis-mehaanilistele omadustele ja hinnale

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Uurimistöö eesmärgiks oli leida kuluefektiivne komposiitmaterjali kooslus, mis oleks väiksema massiga, kuid samas piisavate mehaaniliste omadustega. Polümeerkomposiitmaterjali maatriksmaterjalina kasutati polüestervaiku ja armeeringuks alumiiniumtrihüdraadi osakesi. Hinna alandamise ja massi vähendamise eesmärgil asendati osa alumiiniumtrihüdraadist õõnsate klaassfääridega. Uuriti klaassfääridest täiteaine mõju komposiitmaterjali füüsikalis-mehaanilistele omadustele. Ekstruuder-tüüpi seadmega valmistati vaakumvalumeetodil erineva klaassfääride sisaldusega katsekehad. Valmistatud katsekehadel testiti tõmbetugevust, Barcoli kõvadust ja tihedust. Leidmaks materjali optimaalset koostist, koostati ja lahendati katseandmete põhjal multikriteriaalne optimeerimisülesanne, milleks kasutati närvivõrke ning hübriidgeneetilist algoritmi. Koostatud arvutusmudelis pakuti komposiitmaterjali tiheduse ja mehaaniliste omaduste vastuolu likvideerimiseks välja optimaalne lahendus Pareto kõverana. Ilma kerge täiteaineta materjaliga võrreldes vähenes optimaalsete omadustega kuueprotsendilise klaassfääride sisaldusega komposiitmaterjali tõmbetugevus 3% ja Barcoli kõvadus 26%. Uurimistöö tulemusena väljatöötatud materjali puhul saavutati 13% massi vähenemine, kuid kergema täiteaine kasutamise tõttu suurenes omahind ligi 7%. Samas mõjutab massi vähenemine transpordikulu ja tollimaksu suurust, mille mõju lõpphinnale tuleb enne otsuste tegemist arvesse võtta.