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Optimization of Rheological Properties of Binders Used in Vacuum Assisted Resin Transfer Molding of Fiberglass

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Abstract—The effect of an active solvent such as diethylene glycol on the thermal stability and the rheological and strength properties of epoxy binder was studied. The composition of the binder was optimized according to the Pareto criterion and the optimum content of active solvent was found. Viscosity, ultimate bending strength, and ultimate tensile strength were the parameters used in the optimization process.

Keywords: polymer composite materials, epoxy binder, active solvent, optimization, Pareto criterion

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INTRODUCTION

Fiberglass is a polymer composite material that is gradually replacing metals and alloys and is widely used as a structural material in the aerospace industry, building construction, automotive industry, and other fields of industry and technology [1–6].

In the production of fiberglass products, both materials and products are manufactured in the same technological stage, whereas technologies for producing materials based on metals and those for producing structures made of metals are two separate processes. In recent years, the vacuum infusion technology known as Vacuum Assisted Resin Transfer Molding has become one of the most widespread methods for manufacturing fiberglass products.

The main difference between the infusion technology and the common technology for molding various products made of polymer composite materials via the hand lay-up method followed by the autoclave curing is the use of a fabric instead of prepreg, with the fabric being impregnated with a binder directly after lay-up [7–9]. This technology is widely used due to its economic efficiency because of simplicity, low cost of consumables, and the ability to perform the impregnation and curing without the use of expensive equipment and accessories.

In terms of the wetting process, any fibrous filler is a nonsmooth heterogeneous deformable surface. This circumstance provides wetting hysteresis during the impregnation of fiber with a binder and there are a number of various values of the contact angles of wetting.

The possibility of manufacturing a structure via infusion technology is determined primarily by the rate of impregnation, which depends, in turn, on the kinetics of wetting. The wetting process may be controlled by adjusting the viscosity of binder.

The viscosity of the majority of used binders is usually rather high [2]. Various additives are introduced into the binder composition to decrease the viscosity, with active solvents being the most widely used additives. However, the introduction of such materials leads to not only an improvement of wetting and other rheological properties of binders, but also a decrease in their thermal stability and mechanical strength [10].

The aim of the present study was to optimize the rheological properties of binders in terms of their strength criteria.

EXPERIMENTAL

Eight compositions of binders containing various amounts of active solvent were used as objects of the study (Table 1). The technology for preparing the binders included the following steps. 100 wt parts of ED-20 epoxy resin were initially weighed, and 10 wt parts of triethylenetetramine (TETA) were added. The latter is a common curing agent due to its chemical reactivity, low cost, and good processability, as it makes it possible to cure the binder at room temperature. The resulting mixture was stirred with a mechanical stirrer for 5 min, an active solvent such as diethylene glycol (DEG) was added, and the mixture was stirred for 5 min. The viscosity and the glass transition temperature of the obtained composition was then determined.

Viscosity was determined with the use of a CAP 2000 Brookfield viscometer and the glass transition temperature was evaluated via the differential scanning calorimetry on a DSC 204 F1 instrument [11].

Table 1. Compositions of the used epoxy binders

Binder number	Content of components of binder, wt parts		
	ED-20 epoxy resin	curing agent (TETA)	active solvent (DEG)
1	100	10	0
2			1
3			5
4			10
5			15
6			20
7			25
8			30

The obtained binders were poured into organosilicon forms and cured at room temperature for 24 h, followed by determination of the ultimate bending strength (according to *GOST* (State Standard) *4648*) and the ultimate tensile strength (according to *GOST* (State Standard) *11262*) of the cured binders.

RESULTS AND DISCUSSION

The results of experimental determination of the viscosity, the glass transition temperature, the ultimate bending strength, and the ultimate tensile strength are summarized in Table 2.

Analysis of the results given in Table 2 shows that an increase in the content of active solvent in the binder results in an improvement of its viscosity; however, all mechanical characteristics deteriorate (Fig. 1). Three parameters are considered as optimization criteria:

- h_1, Pa s, is the viscosity;
- h_2, MPa, is the ultimate bending strength; and
- h_3, MPa, is the ultimate tensile strength.

The content of active solvent (**x**) is taken to be a space of optimized parameters.

The following preference ratios occur according to the first, second, and third criteria, respectively:

$$h_1(\mathbf{x}') < h_1(\mathbf{x}'') = \mathbf{x}' \succ_X \mathbf{x}'';$$
$$h_2(\mathbf{x}') < h_2(\mathbf{x}'') = \mathbf{x}' \succ_X \mathbf{x}'';$$
$$h_3(\mathbf{x}') < h_3(\mathbf{x}'') = \mathbf{x}' \succ_X \mathbf{x}''.$$

In the above equations, the following designations are used: $\succ$ is the sign meaning the preference of **x**' decision in comparison to **x**" decision of the *X* set, that is, both decisions belong to the *X* set of decisions.

Thus, the larger the ultimate strength value, the higher the mechanical properties, whereas, in the case of viscosity, the opposite pattern is observed, namely, the lower the viscosity, the higher the mechanical properties of polymer composites. The mutual inconsistency of individual particular criteria suggests that the considered task is a multicriterion optimization problem [12].

In the multicriterion optimization, the Pareto axiom plays a crucial role [13]. If the evaluation of one of two decisions is not worse for all components than the evaluation of the second decision and, at the same time, distinctly better for at least one component, the first decision is preferable to the second, that is,

$$\mathbf{x}', \mathbf{x}'' \in X, h_i(\mathbf{x}') \le h_i(\mathbf{x}''), i = 1\ldots m;$$

$$\exists k \in \{1, 2, \ldots m\} : h_k(\mathbf{x}') < h_k(\mathbf{x}'') = \mathbf{x}' \succ_X \mathbf{x}''.$$

The following designations are used in the equations: $\in$ is the sign meaning that **x**' and **x**" belongs to the *X* set, and $\exists$ is the sign meaning that the {1, 2, ...*m*} set "exists," with 1, 2, ...*m* being its elements.

Analytical approaches for solving the problem of finding the Pareto-optimal decisions are reported in [14] and a review of numerical methods is given in [15].

Since the method for determining the Pareto optimum implies finding a compromise in terms of minimization, the strength criteria are converted into the relative strength reductions:

$$\bar{h}_2 = \Delta h_2 = h_{2\max} - h_2;$$

$$\bar{h}_3 = \Delta h_3 = h_{3\max} - h_3.$$

Table 2. Properties of the studied binders

Binder number (see Table 1)	Viscosity, Pa s	Glass transition temperature, °C	Ultimate bending strength, MPa	Ultimate tensile strength, MPa
1	17	61	139	109
2	12.5	61	143	105
3	3.6	58	145	90
4	1.15	58	148	90
5	0.86	58	115	85
6	0.54	56	107	83
7	0.27	56	110	85
8	0.18	56	107	85

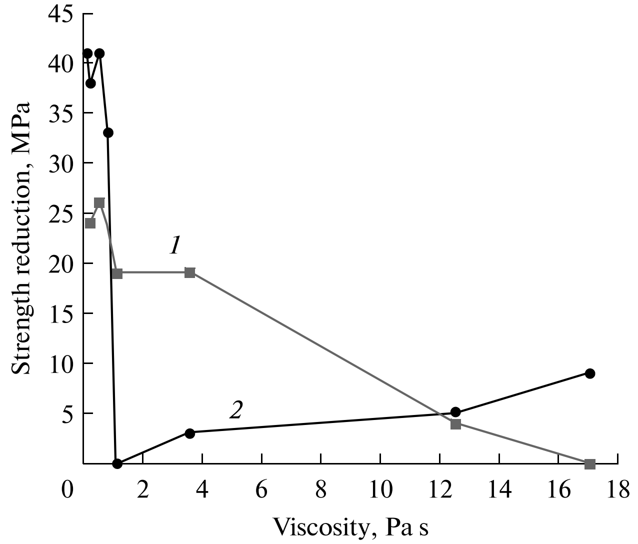

Fig. 1. Dependence of (*1*) the ultimate tensile strength and (*2*) the ultimate bending strength on the viscosity.

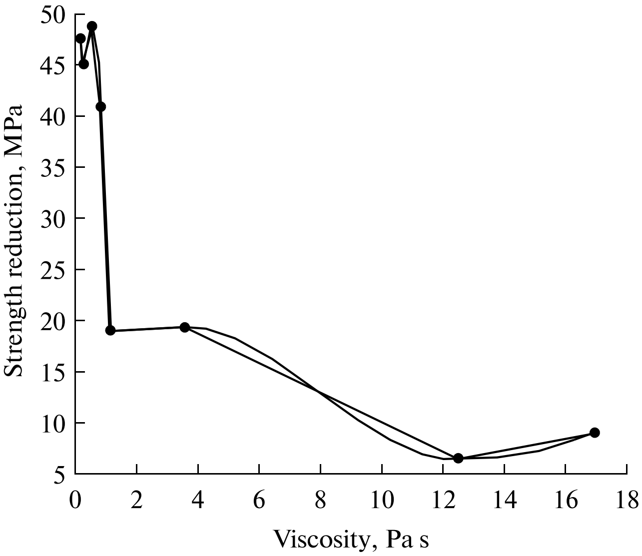

Fig. 2. Dependence of the quadratic estimate of strength on the viscosity.

Thus, the preference ratios are as follows:

$$\bar{h}_2(\mathbf{x}') < \bar{h}_2(\mathbf{x}'') = \mathbf{x}' >_X \mathbf{x}'';$$

$$\bar{h}_3(\mathbf{x}') < \bar{h}_3(\mathbf{x}'') = \mathbf{x}' >_X \mathbf{x}''.$$

The initial dependences are shown in Fig. 1.

Since the trends of particular criteria 2 and 3 are unidirectional, a quadratic convolution of the criteria is performed via the following equation:

$$\tilde{h}_2 = \sqrt{\bar{h}_2^2 + \bar{h}_3^2}.$$

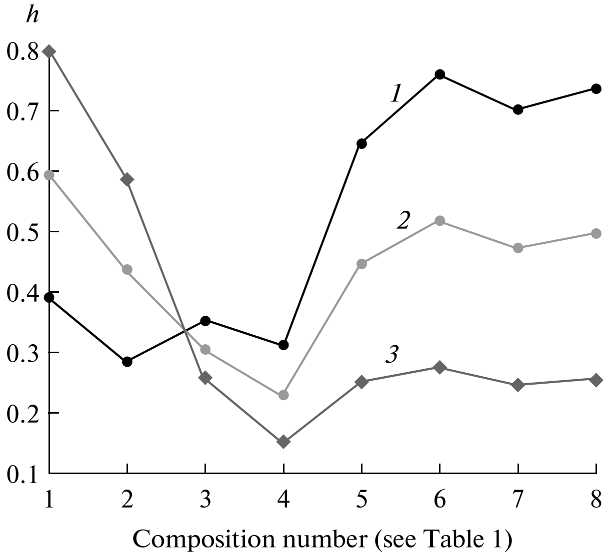

Fig. 3. Dependence of the *h* convolution of viscosity and strength criteria on the content of active solvent in the binder at (*1*) $\alpha = \{0.25–0.75\}$, (*2*) $\alpha = \{0.5–0.5\}$, and (*3*) $\alpha = \{0.75–0.25\}$.

The result is shown in Fig. 2.

As particular criteria are inconsistent, to find the Pareto-optimal decisions does not mean reaching a final decision. The number of found decisions is only suggested to the designer.

The curve approximating the Pareto frontier was obtained from the experimental data. To obtain the only solution, a randomized strategy based on the information on the relative importance of criteria was used. The generalized criterion was considered via the equation for the linear convolution of the $h = (h_1, \tilde{h}_2)$ vector criterion with $\{\alpha_i\}$ weights:

$$J(\mathbf{x}, \alpha) = \langle h(\mathbf{x}), \alpha \rangle = \sum_{i=1}^{m} \alpha_i h_i,$$

where $\langle h(\mathbf{x}), \alpha \rangle$ is the scalar product of $h(\mathbf{x})$, α sets (vectors) and $\alpha = (\alpha_1 ... \alpha_m)$ is the vector of nonnegative weights satisfying the condition:

$$\sum_{i=1}^{m} \alpha_i = 1.$$

The optimal decision was found in the case of weights $\alpha = 0.25–0.75$ and scaling of the maximum values of criteria.

Thus, the performed calculations (Fig. 3) resulted in the optimal binder composition containing 10 wt parts of active solvent (Table 1, number 4).

CONCLUSIONS

The effect of active solvent on the change in the rheological and mechanical properties of epoxy binder was examined. It was found that an increase in the content of active solvent in the binder leads to a considerable decrease in its viscosity, thus, providing a favorable influence on the whole technological process for manufacturing a product. However, the thermal stability and the mechanical strength deteriorate along with the viscosity reduction.

The optimal composition was shown with the use of criteria for the Pareto optimization to be composition number 4 containing 10 wt parts of active solvent.

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