Tensile strength of carbon composite materials: Influence of binding matrix

A A Antanovich1, S A Kolesnikov2

1 Vereshchagin Institute for High Pressure Physics of the Russian Academy of Sciences, 14 Kaluzhskoe shosse, 108840, Troitsk, Moscow region, Russia

2 Research Institute for Graphite-Based Structural Materials, 2 Elektrodnaya st., 111524 Moscow, Russia

E-mail: antanov@hppi.troitsk.ru

Abstract. The calculated estimates of the strength and deformation characteristics of carbon composite materials based on carbon fibers with polymer, carbon and carbon-ceramic matrices obtained by various technologies are presented. The calculation method is based on the structural model of the strength of the composite material as a “bound” beam of reinforcing fibers. Good agreement of the calculated data with the experimental results indicates the applicability of the calculation model used for the design of new materials and products. It is also shown that the isostatic technology of obtaining carbon matrices based on coal pitch allows to obtain carbon composites with properties closest to carbon plastics, which are characterized by the best ability to realize the strength of reinforcing fillers.

1. Introduction

Carbon composite materials based on high-modulus carbon fibers are widely used in various fields of modern technology. Carbon fibers as reinforcing elements provide high strength characteristics of the material, and the binding matrix ensures the joint operation of these reinforcing elements and their protection from mechanical damage and the environment. Depending on the purpose of the composite, polymer or carbon matrices can be used, as well as hybrid matrices containing a carbon and ceramic component.

First of all, we will be interested in carbon-carbon composite material (CCCM), the carbon matrix of which is obtained from thermoplastic coal pitches by isostatic technology, when carbonizing pitches at high pressure [1]. The physical and chemical basis for the formation of an effective matrix in isostatic carbonation are the features of chemical and diffusion processes, as well as the technological ability to fill the entire porosity of the solid carbon matrix. This technological possibility of filling the entire porosity with a solid carbon matrix is provided by the formation of only an open form of porosity at the exit of hydrogen at the stage of completion of carbonization. The open porosity form is available for multiple repeated processes of impregnation with liquid pitch and its carbonation up to almost complete exhaustion of porosity [2].

As a result, a carbon matrix is formed in the volume of CCCM, which differs in its physical properties from the more traditional polymer and carbon ones obtained by pyrolytic technology. Pyrolytic matrices include a carbon mass containing coke of phenol-formaldehyde resins and deposits of pyrolytic carbon in the pores.
In addition to the issues of formation of the binder matrix in carbon composites of interest is the study of the process of forming physical-mechanical characteristics of composites with the aim to determine the extent of the realization of the potential properties of the reinforcing component. For this purpose, in this paper, we have calculated the tensile strength of one-dimensional reinforced composite materials (rods) on the basis of one type of carbon fiber and different types of matrices. The calculated results were compared with experimental ones.

2. Methodical part

In the manufacture of carbon composites, carbon fiber was used in the form of a carbon structural woven thread UKN-5000 containing 5000 filaments. The diameter of the filaments ~7 microns. The strength distribution of filaments with a working length of ~10 mm was determined when they were stretched in a paper frame. As a result, the following parameters of strength distribution are obtained: the total number of tests is 128, the minimum value is 1.8 GPa, the maximum one is 6.0 GPa, the average — 3.78 GPa, coefficient of variation of the distribution is 0.227, standard deviation is 0.86 GPa and the Weibull parameter is 5.3. For the distribution of the modulus of elasticity total number of tests is 128, the minimum score is 220 GPa, a maximum one is 320 GPa, an average — 273 GPa, coefficient of variation of the distribution is 0.094, standard deviation is 25.6 GPa, and the parameter of Weibull is 12.8.

Figure 1, consisting of four parts, shows the structure of carbon plastic based on epoxy resin in the initial state and in the process of development of destruction during mechanical tests. The diameter of the reinforcing rods was ~1.2 mm. The image on the first part of the figure obtained by means of optical microscopy shows an uneven distribution of filaments inside the rod. Numerous cavities formed between the contacting filaments are visible. Such a real dispersion of the bulk fiber content in the composite can lead to the statistical nature of the filler strength realization and the appearance of non-equilibrium internal stresses in the areas of convergence and contact of the filaments with each other.

Reconstructed images of sections of materials obtained with the help of a computer microtomograph are presented in parts 2 and 3 of the figure. Sections of the state of materials are obtained at distances of several mm from the immediate fracture crack. Part 2 of the figure shows the
stage of destruction, accompanied by a rupture of the matrix in the direction of the transverse direction of the reinforcing structures. The third part of the figure shows the stage of destruction as a result of stresses and cracks in the transversal direction of reinforcement. The fourth part of the figure shows the destruction of reinforcing rods with the formation of a discrete structure of the composite. As a result, the structural material loses all its performance properties.

The manifestation of these stages of destruction is a continuous process of destruction with increasing external load until the exhaustion of the bearing capacity of the material.

3. Estimates

The method of calculating the tensile strength of one-dimensional reinforced composite materials is based on the structural model of the strength of the composite material as a "bound" bundle of reinforcing fibers [3,4].

Estimates carried out for CCCM with the formation of the matrix using isostatic carbonization of coal tar pitch in the amount of structure of constructional carbon yarn. For comparison, the dependence of the realized potential strength for the material on the basis of pyrolytic technologies of material matrix formation is estimated. The results are compared with the corresponding estimates for carbon composites with epoxy resin-based matrices. Properties of composite materials based on carbon fibers and epoxy matrices are currently the standard for modern materials science.

In calculations, we used the following data bases for the properties of binding matrices: [3] for polymer matrices, [5] for matrices prepared by isostatic carbonization of coal tar pitch, and [6,7] for pyrolytically produced matrices. According to [3], the composite strength ($\sigma_{\text{comp}}$) is calculated in relation to the strength of the “bound” bundle of reinforcing fibers ($\bar{\sigma}$) using the expression

$$\sigma_{\text{comp}} = k_\sigma k_E k_p V_{f'\sigma},$$

where $V_f$ is the volume share of fiber, and coefficients $k_\sigma$, $k_E$, $k_p$, and $k_{yf}$ take into account a decrease in $\bar{\sigma}$ caused by dispersion in the strength of filaments, Young modulus, porosity, and volume content of filaments in the fiber respectively the variance of the volume.

Here it is assumed that the heterogeneity of the mechanical properties of the filaments weakens the strength of the bundle. The “weakest” in strength filaments are destroyed primarily as the external load increases and drop out of effective work of the composite as a whole, turning, ultimately, into discrete structures.

The “weakening” of the composite due to the dispersion of the elastic modulus is associated with non-uniformity and non-uniformity of stress in all filaments of the bundle. In this case, the most high-modulus filaments from the General ensemble in advance perceive the mechanical load, are destroyed and also drop out of effective work.

The coefficients in (1) are normally calculated using the bulky formulas [3] based on the statistical data base for the strength of carbon fibers and carbon matrices. The calculated values of strength realization coefficients as a function of $V_f$ are collected in Table 1 along with calculated values of $\sigma_{\text{comp}}$.

Table 1. Calculated coefficients of strength realization for carbon fiber UKN-5000 in a matrix of coal tar pitch.

| $V_f$ (share) | $\bar{\sigma}$ (GPa) | $k_\sigma$ | $k_E$ | $k_p$ | $k_{yf}$ | $\sigma_{\text{comp}}$ (MPa) |
|--------------|----------------------|-----------|-------|-------|---------|------------------------|
| 0.30         | 3.87                 | 0.660     | 0.790 | 0.757 | 0.769   | 360                    |
| 0.40         | 4.06                 | 0.660     | 0.790 | 0.757 | 0.726   | 460                    |
| 0.50         | 4.26                 | 0.660     | 0.790 | 0.757 | 0.675   | 570                    |
| 0.58         | 4.42                 | 0.660     | 0.790 | 0.757 | 0.622   | 630                    |
| 0.60         | 4.47                 | 0.660     | 0.790 | 0.757 | 0.606   | 650                    |
| 0.70         | 4.74                 | 0.660     | 0.790 | 0.757 | 0.483   | 640                    |
| 0.75         | 4.88                 | 0.660     | 0.790 | 0.757 | 0.354   | 510                    |
Figure 2 shows the final calculated dependences of the tensile strength of one-dimensional reinforced composite materials on the volume content of carbon fiber UKN-5000.

![Figure 2](image)

**Figure 2.** The dependence of the tensile strength of one-dimensional reinforced composite materials on the volume content of carbon fiber: 1 — thermoplastic on the basis of epoxy resin; 2 — CCCM with a matrix of coal tar pitch (isostatic technology); 3, 4 — pyrolytically prepared carbon composites.

The main characteristic for the qualification of the structural material is its deformation curve. To obtain the calculated shape of the deformation curve, structural expressions of the monolithic conditions of composite materials were used [3,4].

At the same time, the following possibilities of manifestation of the mechanisms of composites destruction were consistently considered:

- The beginning of crushing the polymer matrix. This happens when the ultimate strain of matrix is smaller than that of fiber. A disintegrated matrix self-eliminates itself from resisting to an applied load.
- The beginning of the exfoliation of the matrix when Poisson coefficient of matrix is higher than that of binder.
- Development of transversal fracture of the matrix with a high content of carbon fiber. This event takes place when the rigidity of matrix is higher than the transversal elasticity modulus of fiber.
- The beginning of the crushing of fibers by frictional contact with the matrix.

Upon damage, composite converts into a system of unbound filaments. Based on the model for the integrity of composite materials [3,4], we calculated the domains of boundary conditions for manifestation of that or other fracture mechanism. Using an additivity rule, we estimated elasticity moduli for each stage of material fracture and then obtained deformation curves for each carbon material under consideration. Figure 3 shows the calculated deformation curves for reinforced carbon composites with $V_f \approx 0.58$. 
Figure 3. Calculated deformation curves for reinforced carbon composites with $V_f \approx 0.58$: 1 — thermoplastic on the basis of epoxy resin; 2 — CCCM with a matrix of coal tar pitch (isostatic technology); 3, 4 — pyrolytically prepared carbon composites.

4. Discussion
Our estimates well agree with experiment. For instance, the measured value of $\sigma_{\text{comp}} = 687$ MPa for the CCCM with $V_f = 0.58$ prepared by isostatic technology is consistent with a calculated value of 630 MPa (see Table 1).

Our analysis has shown that the main reason for a 20-30% decrease in the material strength is the dispersion in the properties of reinforcing filler. Next by its importance is the dispersion in the volume share of filaments. Variation in $V_f$ with in the range 0.5-0.6 in a given kind of material can lead to a 10-12% change in material strength. Local closer approaching of the fibers up to $V_f \approx 0.7$ may cause a drastic drop in material strength.

Elevated rigidity of matrix as well as its porosity also favor earlier damage of material under the action of an external load. The manifestation of these factors can be reduced to the greatest extent for composites based on isostatic technology, since the modulus of elasticity of the carbon coke matrix from coal pitches is closest to the modulus of the polymer matrix, and the pore volume in this case theoretically and practically can be minimal.

5. Conclusions
Modeling of fracture and deformation processes of different nature on the basis of general approaches allows to obtain reliable estimates of the properties of carbon composite materials for subsequent optimization of their production technology.

Isostatic technology for producing carbon composite materials makes it possible to obtain materials with properties closest to carbon plastics, which are characterized by the best ability to realize the strength of reinforcing fillers.

Our model for deformation and fracture of carbon materials gives reasonable results and can be recommended for optimization of process conditions. Isostatic technology ensures the fabrication of carbon composite materials with the properties close to those of carboplastics.

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