Effect of the sol temperature on the structure and thickness of the SiO$_2$ coating of carbon fibers

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Abstract. This paper deals with the development of carbon fiber coatings that act as diffusion barrier, and the role of the weak boundaries in the metal matrix composites. The developed coating will be a cost effective alternative to CVD-coatings. The paper considers the temperature dependence of the structure of SiO$_2$-sol and the coating thickness on the surface of the carbon fibers formed by sol-gel method. Detected sol temperature range optimum for coating. Possible reasons for the observed phenomena are proposed.

1. Introduction

Active research of directionally reinforced composites with a metal matrix and carbon fiber dates back to the 60s of the last century. However, compared with carbon fiber reinforced polymers, similar composites with a metal matrix have not found wide application in industry. As a first approximation, this is due to several natural reasons. Firstly, the chemical interaction between the matrix and the fiber introduces rough defects on the fiber surface, which significantly reduces its effective strength, and hence the strength of the composite [1]. Secondly, this interaction leads to the formation of a “strong” matrix/fiber bond, which in most cases is not desirable due to the need for uniform distribution of fibers in the matrix and limitation of the fiber volume fraction (no more than 30%) [2].

A possible solution to these problems is to create a barrier layer at the matrix/fiber interface. The most common method of applying barrier coatings that plays the role of “weak” boundaries is the CVD method. For example, in [3] the authors using a CVD coating of pyrolytic carbon, managed to obtain a composite with carbon fiber and an aluminum matrix (Al/Cf composite), the strength of which reaches 1360 MPa - the highest strength of an Al/Cf composite in open print today. Another example is work [4], in which the authors obtained a composite with an effective fiber strength of 95% of the initial one using gradient CVD coatings C/SiC/Si.

However, despite the successful elimination of the natural causes mentioned earlier and the creation of composites with high strength, such composites are still not widely used. This is primarily due to the complexity and low cost-effectiveness of the CVD method for coating carbon fiber.

A more accessible method for obtaining coatings from a practical point of view is the sol-gel technology, which does not require extreme conditions. Sol-gel technology makes it possible to obtain oxide coatings practically in “room” conditions. Such coatings serve as a reliable diffusion barrier [5, 6], and can also act as “weak” boundaries [7].

Today, there are several dozen works devoted to the deposition of oxide sol-gel coatings of various compositions on carbon fiber [8-10]. However, systematic research in this area is currently lacking.
In this work, the dependence of the structure and thickness of SiO$_2$ coatings on the carbon fiber surface formed by the sol-gel method on the sol temperature is considered. The optimum temperature range for the sol is determined for coating.

2. Materials and methods

UMT40-3K carbon fiber (manufactured by UMATEX Group, Russia) was used as a substrate for the deposition of SiO$_2$ coatings.

An oxide coating was applied to the carbon fiber by the method based on the sol-gel technology, similar to the method described in detail in [11, 12]. Carbon fiber is preliminarily cleaned from a sizing agent in acetone. Further, to create carboxyl groups on the fiber surface participating in the formation of sol-gel particles, the fiber is treated in nitric acid. After that, the fiber is immersed in a sol-gel solution of the following composition: tetraethoxysilane, ethanol, water and hydrochloric acid. In this work was studied the effect of the sol temperature on the structure of the coating formed on the surface of the carbon fiber. For this, five series of experiments were carried out at temperatures from 20 to 60°C.

The obtained sol-gel coatings on the carbon fiber surface were studied using a VERSA 3D scanning electron microscope. The images were obtained in the mode of secondary electrons with an accelerating voltage of 10 kV at magnifications up to 50000 times. It is known from preliminary experiments that the coating on the filaments on the carbon fiber tow surface may differ from the coating on the inside. Since the majority of filaments are concentrated precisely in the inner part of the fiber, for the study of coatings by scanning electron microscopy was used a technique for preparing samples, which allows obtaining more representative data in comparison with direct research of the bundle outer layers. After coating, a fragment of 10 mm was cut from a continuous bundle. The fragment was divided into two parts, after which one of the parts was placed on the surface of a carbon conductive tape fixed on the scanning electron microscope standard stage. After that, using tweezers the filaments of the selected fragment were distributed evenly over the conductive tape surface.

3. Results and discussion

Figure 1 shows the microstructure of the obtained coatings. At a sol temperature of 20°C, a coating is formed on the carbon fiber surface from a multitude of sol particles with average size is 10 ± 4 nm. When the temperature rises to 40°C, the thickness of the coating increases, and its structure becomes looser. At a temperature of 50°C there is an abrupt change in the thickness and structure of the coating, which becomes much thinner and more homogeneous in thickness. A further increase in temperature leads to a further decrease in the coating thickness.

The coating has a grained structure. Probably, the grains forming the coating are sol particles, and the coating is formed by the mechanism of their aggregation. It should be noted that an increase in temperature leads not only to a decrease in the coating thickness, but also a decrease in the size of the coating particles.

The observed effect is probably associated with the influence of temperature on the coagulation processes and the characteristics of the electric double layer on the surface of the sol particles. In the works of the classic of sol-gel processes Ralph K. Iler, it was noted that an increase in the temperature of some aqueous SiO$_2$ sols to 80°C, contrary to expectations, leads to an increase in the gelation time or, in other words, inhibits the aggregation process. This fully explains the effect observed in this work.
Figure 1. Microstructure of SiO$_2$ coatings of carbon fiber obtained at various sol temperatures: (a) 20°C, (b) 30°C, (c) 40°C, (d) 50°C, (e) 60°C.

Figure 2 shows the dependence of the average coating thickness on the sol temperature.
The explanation of the above dependence undoubtedly requires further investigation of the process. Most likely, the sol temperature has such an effect on the structure and thickness of the coating due to the effect of temperature on the processes of polymerization, condensation, and dissolution in the colloidal system. Thus, it can be assumed that an increase in temperature up to 40°C promotes the condensation process. With a further increase in temperature, active dissolution of sol particles occurs, which leads to a significant decrease in the coating thickness.

4. Conclusion
The dependence on the temperature of the sol of the structure and thickness of SiO$_2$-coatings on the carbon fiber surface, formed by the sol-gel method, was determined. It is shown that the optimum temperature for coating is the temperature of the sol in the range from 20 to 40°C. The largest thickness of the obtained coatings was about 230 nm.

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