Study of the influence of filler on the mechanical properties of composites based on polyimide

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Abstract. Physical and mechanical properties of polymer composite films on the basis of polyimide filled with high-temperature superconductors (yttrium ceramics) were studied. Calculations under the catastrophic models were carried out. Exponential and parabolic models show the best agreement with the experiment.

1. Introduction
Creation of polymer ceramic and composite materials with improved or novel properties and characteristics is an essential prerequisite and condition for further development of scientific and technological progress. The constant growth of requirements for modern technology leads to the appointment of the polymer, its nature, operation conditions and other factors [1, 2]. Advances in the field of superconductivity are key to energy, electronics, high-energy physics, air, land and sea transport, space exploration, medicine and many other areas [3]. Increased applications of superconducting materials and increase of the efficiency of their work require further improvement of performance, in particular mechanical properties [4]. Products of flexible HTS (high temperature superconductors) are of practical interest. Since the ceramic HTS materials have a high brittleness and low strength, important materials to be studied are film systems of polymeric composite materials (PCM) on the basis of their compounds having structural perfection polycrystallites and plastic properties of polymers [5]. Preparation of YBCO-films with desired properties can be carried out when understanding features of the formation of the cationic composition of the films during their growth [6]. Establishing a connection between the structure and properties of defects makes it possible to obtain information about the microscopic parameters that are important for understanding the mechanism of the processes in these materials.

The synthesis of new materials is based on the variation, integration and combination of objects of different nature, in particular, organized molecular structures, as well as on the optimization of their spatial organization [7]. Directional surface modification of HTS materials lies in the change of the chemical composition of sample surface by modifying the electrolyte composition and properties, adsorption of various surfactant particles, etc. [8-10].

To date, a huge amount of experimental data requiring theoretical interpretation has been collected, but existing models, including those of mechanical properties can not explain some of these data.

2. Methodology
Physical and mechanical properties of polymer composite films of polyimide base filled with HTSn of two concentrations were studied: C = 0.1 % wt., 0.5 % wt. Fine-grained powder of YBa\textsubscript{2}Cu\textsubscript{3}O\textsubscript{6,7}
was used as filler. Test samples were prepared by mechanical mixing of the polyimide-based varnish and fine crystalline powder of YBa$_2$Cu$_3$O$_{6+x}$, when $x = 0.7$. Mechanical tests were conducted on the tensile machine of C-TAP 2.0 type with uniaxial tension of special reverser at the constant load. The load had been applied until the film was disrupted at the unclamping rate of 1.53 mm/min and with increasing of load for 0.06 dynes/min. The work area was 40-mm long and 5-mm wide. The experimental results were processed using a special computer software that is installed as a standard Windows application [11].

3. Discussion

Figure 1 shows the dependence of the relative elongation $\varepsilon$ on the applied stress $\sigma$ unirradiated composite material for two values of the concentration of the second component. Introduction of the filler in the form of fine crystalline yttrium ceramic powder in polyimide causes a significant change in the plastic and strength properties of the polymer. Thus, when changing the filler content from $C = 0.1$ wt.% to $C = 0.5$ wt.%, a decrease of tensile strength ($\sigma$) from ~ 20 MPa to about 2 MPa in comparison with pure polyimide film is observed, and deformations of these materials are significantly reduced from $\Delta \varepsilon = 41\%$ to $\Delta \varepsilon = 43\%$, respectively. In the region of 40 - 47 MPa, for samples of PCM there is a sharp jump in the relative elongation, shifted to lower stresses with increasing filler concentration (YBCO). Increasing the concentration leads to a stabilization of the plastic properties of PCM and deterioration of its elastic properties ($\sigma$ reduced from 90 MPa to 72 MPa). This effect of filler concentration on the mechanical properties of PCM is due to a change of the porosity of material and structural activity of the filler related to polymer matrix. Change in the plastic properties of PCM samples is due to a high content of O$_2$ in YBCO crystals [11].

![Figure 1](image)

**Figure 1.** Dependence of elongation $\varepsilon$ on the applied load $\sigma$ for polyimide composite materials for two concentrations of the second component.

In the stress interval of $0 - \sigma_1$ (curve 3) and $0 - \sigma_2$ (curve 4) dependences of $\varepsilon$ from $\sigma$ are satisfactorily described by a linear model.

Establishment of the dependence of $\varepsilon$ from $\sigma$ (instead of $\sigma$ from $\varepsilon$, as is generally accepted) is the most correct from the physical point of view [11].

The formula for the reduced strain $\varepsilon_p$ on $\sigma$ was used to describe the dependence of $\varepsilon$ from $\sigma$.

In this case

$$
\varepsilon_p = \begin{cases}
    f(\sigma) - \varepsilon_0, & \text{when } 0 \leq \sigma \leq \sigma_f, \\
    f(\sigma) - \varepsilon_0 - \Delta \varepsilon_f, & \text{when } \sigma > \sigma_f.
\end{cases}
$$
here $\sigma_i$ is equal to $\sigma_{01}$, or $\sigma_{02}$ depending on the curve (1 or 2), $\varepsilon_0$ is the shifting value, $\Delta \varepsilon_i$ is changing of deformation during the phase transition.

Let’s consider three types of catastrophic models: exponential, quadratic exponential and parabolic [12]:

$$f(\varepsilon) = \exp\left(\frac{\sigma}{\sigma_0}\right) - 1,$$

(2)

$$f(\varepsilon) = \exp\left(\frac{\sigma^2}{E_1^2}\right) - 1,$$

(3)

$$f(\varepsilon) = \frac{\sigma^2}{E_2^2}.$$ 

(4)

In (2) $\sigma_0$ is the value of stress when $\varepsilon_p$ increases $e$ time.

The results of calculations of $\varepsilon_p$ from $\sigma$ for different models are shown in Figure 2 (a, b). Analysis of dependencies of $\varepsilon_p$ from $\sigma$ shows that the experimental data are described best by parabolic and exponential models. It should be noted that for the concentration of 0.1 wt% $\sigma_{01}$ is 47 MPa, $\varepsilon_0 = 2.78$ and $\Delta \varepsilon_{01} = 0.82\%$ and for 0.5 wt.% $\sigma_{02}$ is 39.8 MPa, $\varepsilon_0 = 3$ and $\Delta \varepsilon_{02} = 0.4\%$, respectively.

![Graph](image)

**Figure 2.** Dependence of $\varepsilon_p$ from $\sigma$ for composite material based on polyimide for various models.

At the same time the largest difference between the calculations and the experimental data is observed in the areas of stress $\sigma_{01}$ and $\sigma_{02}$. For a quantitative explanation of this effect it is necessary to use the appropriate models of phase transitions.

4. Conclusion

1. Introduction of yttrium ceramic filler into the polyimide causes a significant change in plastic and strength properties of polymer matrix. In the region of small deformations, dependence of $\varepsilon$ from $\sigma$ is
satisfactorily described by Hooke’s law. When $\sigma = \sigma_f$ there is a sharp jump. When $\sigma > \sigma_f$ the dependence is more complex.

2. The appropriate calculations were made using the exponential, exponential-quadratic and parabolic models. Parabolic and exponential models have the best agreement with the experimental data (curves shown). The largest difference is observed in the region of $\sigma_f$ that is due to the influence of phase transition on $\varepsilon(\sigma)$ dependence.

Acknowledgments
The study was supported by the Grant of the Ministry of Education and Science of the Republic of Kazakhstan (a.№ 274, 229).

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