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CATASTROPHIC MODEL OF DESTRUCTION AND EXPERIMENTAL RESEARCH OF POLYMER MATERIALS IRRADIATED BY ELECTRONS

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The paper is devoted to research of physical and mechanical properties of metallized polymer composites based on polyimide and silver irradiated by electrons up to different integral doses and temperature. General principles of operation of upgraded computerized test setup have been described. Obtained experimental data have been explained within catastrophic exponential model of destruction.

Keywords: metallized polymer composites, model of destruction, electrical properties, irradiation dose.

Introduction

Metallized polymer materials are of great applied interest in space and military field as well as in FMCG sector lately. The most advanced metallized polymer materials are polyimides of raised temperature resistance and with dialectic characteristics and small coefficient of expansion. Advanced materials search is mainly aimed at manufacturing composite films with nanostructures with various phase distribution.

Test setup

Automated setup based on rupture-test machine RMU-0.05-1 has been installed for tension testing of samples of wire, metal ribbon, rubber and plastic. The setup consists of two modules. The first module is a transmitting module consisting of series of converters “voltage-frequency”, buffer and auxiliary elements. The module was made on base of sensitive converters ADVFC-32 by Analog Devices. Series of converters “voltage-frequency” has got 8 channels, which convert the signals into rectangular pulse signals of definite frequency. Number of channels may be changed depending on tasks of experiment. Operating range of output signal from series of converters is 50 KHz. Frequency of a signal may be increased up to 200 KHz if necessary. The converters may easily scale output signals by using input signal of a sensor. Modern displacement sensors have got pulse output as a rule. Signals of the sensors are transmitted immediately after buffering, which lets avoid of losing accuracy. Digital code may be read off byte-serial. The setup may perceive signals such as current, voltage and pulse signals as well.

The second transmitted module consists of series of functional blocks along with line receivers. Here demultiplexor functions as a switch, which calls over channels of the transmitting module in turn. Rectangular pulse signal from series of converters of demultiplexor is transmitted into eight-bit decimal counter. It reads frequency of the next channel in the series. The scheme may be re-commutated for measurement of period when frequency of a signal becomes too low if necessary.

Rupture-test machine RMU-0.05-1 has got the following characteristics:

- maximum permissible load 500 N;
- number of ranges of load measurement - 6;
- limit of permissible value of relative measurement error of weighting device ±1%.

Polyimide films have been exposed by temperature in the setup, the block-scheme of which is presented in fig.1.

The test setup consists of the following blocks:

1- Experiment chamber for thermal exposures;
2- Calibration weights, film fixtures, elongation control;
3- Control unit of experiment chamber;
4- System power supply unit;
5- Control unit of film temperature;
6- Research unit of dynamic properties of films;
7- Transmitting module;
8- Demultiplexor;
9- Data processing system.

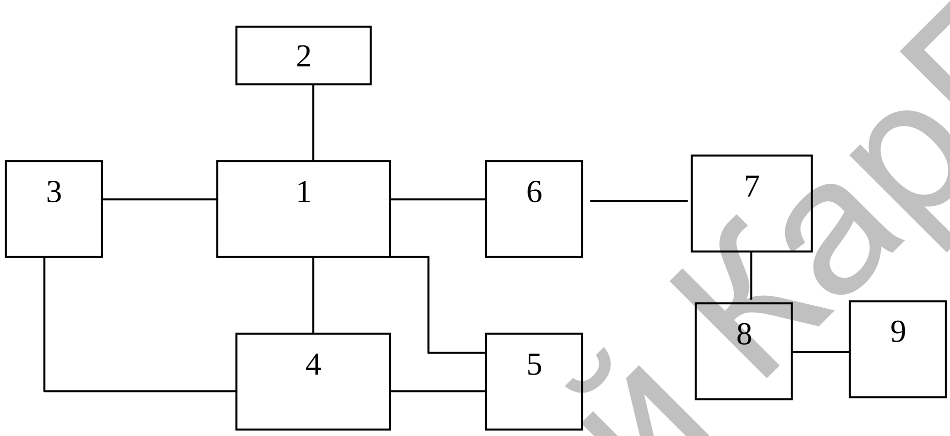

Fig. 1 Block-scheme of setup for temperature research of irradiated and nonirradiated polyimide films

Heating chamber is connected to regulator and temperature meter ITR 2526, which provides adjusting and temperature maintaining within the operating area of length l = 150 mm and range within of 293 - 673 K accurate to $\Delta T = \pm 2$ K. It allows to expose polyimide film homogeneously within the limits of relative lengths of the sample $\varepsilon = l/l_o$ from 0 to 200%. Temperature distribution along the chamber within 70- 180 mm is uniform. The conditions allowed to research static load and complex effect of static load Φ and temperature T upon irradiated and nonirradiated polymer materials.

Measurement procedure

Metallized polyimide films 35 mm of thickness, d = 35 micron manufactured in Chemical Institute at RoK National Academy of Science named after Bekturov have been tested.

The first manufacturing stage included obtaining of polyimide films of different thickness based on lacquer of polyimide **ABimide.** The manufacturing scheme includes lacquer pouring though nozzles into cohesive layer of belt-type conveyor and then thermal drying at . 90°C and thermal cyclization at 180°C during 1 hour and 2 hours respectively and then film rolling.

The second manufacturing stage for obtaining metallized films was as follows: polyimide film was treated in organic solvent, water-alcohol alkali solvent, washed, chelated, washed by dialysis. Termal and chemical metal reduction was made at 220°C. Metallized polyimide films are made as complete (one or two-) or discrete (as a picture, as a microcircuit) metallized layer impregnated into structure of polyimide base. Metal phase may be made of silver, copper, cobalt, nickel or Co-Ni alloy of thickness of 1-5 micron. Total thickness was 25-100 micron depending on thickness of original film.

Optical characteristics: there is 80 - 97% relative silver mirror at surface in visual and infrared zones. Electrical conductivity: surface resistance is 0.1-10 Ohm depending on thickness of metallized layer and metal nature. Dielectric properties: two-sided metallized films are capacitors with the following parameters: Dielectric constant -3.9; loss tangent – 0.0019; volume dielectric resistance – 1x1014 ohmxm; dielectric strength – 210 кV/mm; elastance 70-100 Pico farad per cm^2 depending on film thickness.

Delamination of metallized surface layer is not observed until destruction of integrity of whole polyimide layer at loads not less than 160 MPa (20^0C) and 100 MPa (more than 200°C). Then samples were stretched and deformed at room temperature and unaxial tension by using computerized test setup. Obtained results of the experiment were exported into Excel tables and processed (system Mathematical 5).

Thermal and mechanical research of polyimide films was made as follows: some samples were studied to find out their maxim static load (in percentage) up to their rupture at normal conditions. Temperature limits of polyimide materials destruction as criteria of thermal exposure were found out. Strength characteristics of samples were determined by means of calibration weights. Rupture stress was determined $\sigma = \sigma_{max}$, which is taken as 100% of maximum static load $\Phi = \Phi_{max}$. Static loads were fixed by using variations of calibration weights.

Then loads were chosen from 50% to higher for polyimide films. Upper and lower part of films were fixated by fixtures, film was suspended by using upper fixture from the stand and was lowered into vertical heating chamber. Weights were fixated to the lower part of film by using fixtures.

Task at the first stage was to investigate mechanical properties of polyimide materials. Samples were made of polyimide metallized film, 35 micron thick, 5 mm wide and 70 mm long (working length is 50 mm). Special device was used for preparing samples. The device is a massive frame, where clamping plates are placed and used for film fixing.

Some samples were tested by using rupture-test machine RMU-0.05-1 with purpose to obtain load versus elongation; mean load was also determined, where sample destruction was observed. Some samples were exposed by electron beam, temperature and mechanical load. Weights were chosen to get 80% of tension from rupture stress in samples. Source of electrons of high energy was accelerator ELU-4 installed in Abai Kazakh National Pedagogical University. Beam parameters as follows: electron energy ~ 2 MeV; total beam current ~ 1200 mkA. Elongation of the samples was observed under these conditions.

Experimental results and discussion

Fig.2 shows dependence of relative elongation on tension for nonirradiated metallized film.

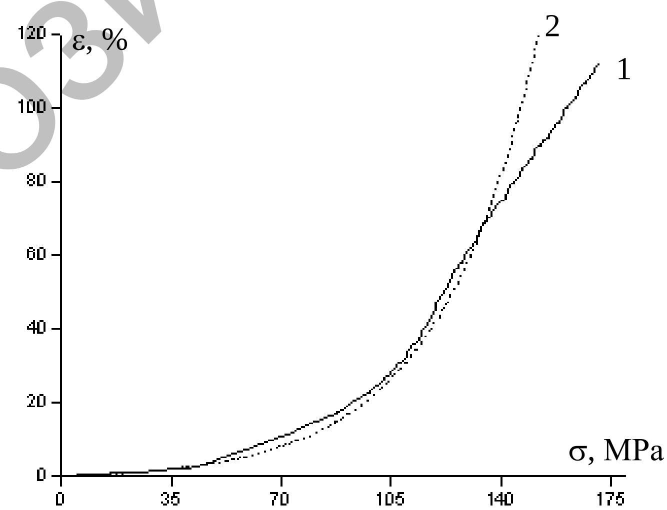

Fig.2 Dependence of relative elongation on tension for nonirradiated metallized films; 1- experiment; 2 – calculation

Metallization of polyimide films increases relative elongation up to 120% and applied stain may reach ~ 175 Mpa. It is related to the method of films manufacturing – chemical etching, silver in this case. Morphology of the upper layer of metallized samples is nanostructured but not homogeneous by volume of the upper layer. External appearance look likes close-packed metal grains of 50 nm in size. In the upper layer grains size is 8 – 10 nm. There is specific gradient of metal distribution over film volume with maximum metal content over the upper layer, which decreases with material depth. For calculation the following expression was used for dependence of ε on σ [3]:

$\varepsilon = \exp\left(\frac{\sigma}{E}\right) - 1$; or by expanding into series: $\varepsilon = \left(\frac{\sigma}{E}\right) + \frac{\sigma^2}{2!E^2} + \frac{\sigma^3}{3!E^3} + \dots$

where E - generalized strength modulus of polyimide film.

Generalized strength modulus for nonirradiated metallized film. E = 31 MPa.

Fig.3 (a, b, c, d) shows dependences of relative elongation on tension for irradiated metallized polyimide films. Generalized strength modulus for irradiated metallized film is varied depending on irradiation doze and it is equal:

at D = 10 MGy E = 31.1 MPa;
D = 20 MGy E = 31 MPa;
D = 30 MGy E = 30.1 MPa;
D = 40 MGy E = 27 MPa

Effect of electron irradiation upon metallized polyimide films shows as follows: relative elongation is decreased when irradiation doze is increased. F.e. relative elongation for 10 MGy is 100% and for 40 MGy is 18%. Respectively material strength is decreased from 160 to 80 MPa. As you may see from the fig. 3 the suggested mathematical model describes well dependence of relative elongation on tension for nonirradiated and irradiated by different dozes metallized films based on polyimide.

Complex exposure to some materials was the next stage of the experiment. Fig.4 shows dependence of relative elongation on rupture time for metallized polyimide films at different dozes of election irradiation. Complex exposure of temperature, static load (80% of rupture stress) and irradiation shows that relative elongation for nonirradiated metallized film was ~ 90 %, rupture time - 24 minutes; relative elongation for irradiated metallized film (40MGy) was ε = 30% и τ = 8 min, i.e. increased 3 times.

The nonirradiated film was ruptured at temperature T = 290 ^{0}C; The irradiated film was ruptured at D= 20 MGy and T= 230 ^{0}C; at D= 40 MGy and T = 95 ^{0}C.

Effect of electron irradiation upon electrical properties of metallized polyimide films at different metal concentrations was investigated ξ (1N – low level, 2N – middle level, 3N – high level). Resistance was measured before and after irradiation at different dozes and different sample lengths: 10, 20, 30, 40, 50 и 60 mm.

The results show that electrical resistance is increased essentially for all three types of metallization of samples when irradiation dose D is increased. Metallization degree ξ affects upon electrical resistance increase. So for ξ = 1 N when irradiation dose is increased it becomes constant quicker than for the film with more metallization degree. This can be explained particularly by silver oxide formation in metallized layer during irradiation by electrons (dielectric). Research shows that specific electrical resistance of irradiated surface of metallized film ρ 10 – 20% more in comparison with other side of the sample. When doze increases the sharp increase of electrical conductivity of polyimide happens due to radiation defects formation in cross-linked structures or benzene nucleus matrices as well as accumulation of radiolysis products at irradiation. Total low level of dielectric loss is related to the fact that major mass of polar groups, which are carboxides, is located within cyclic nanotubes [1,2]. Sometimes at some irradiation doses local rupture of metallized layer may happen and electrical resistance reaches its maximum value.

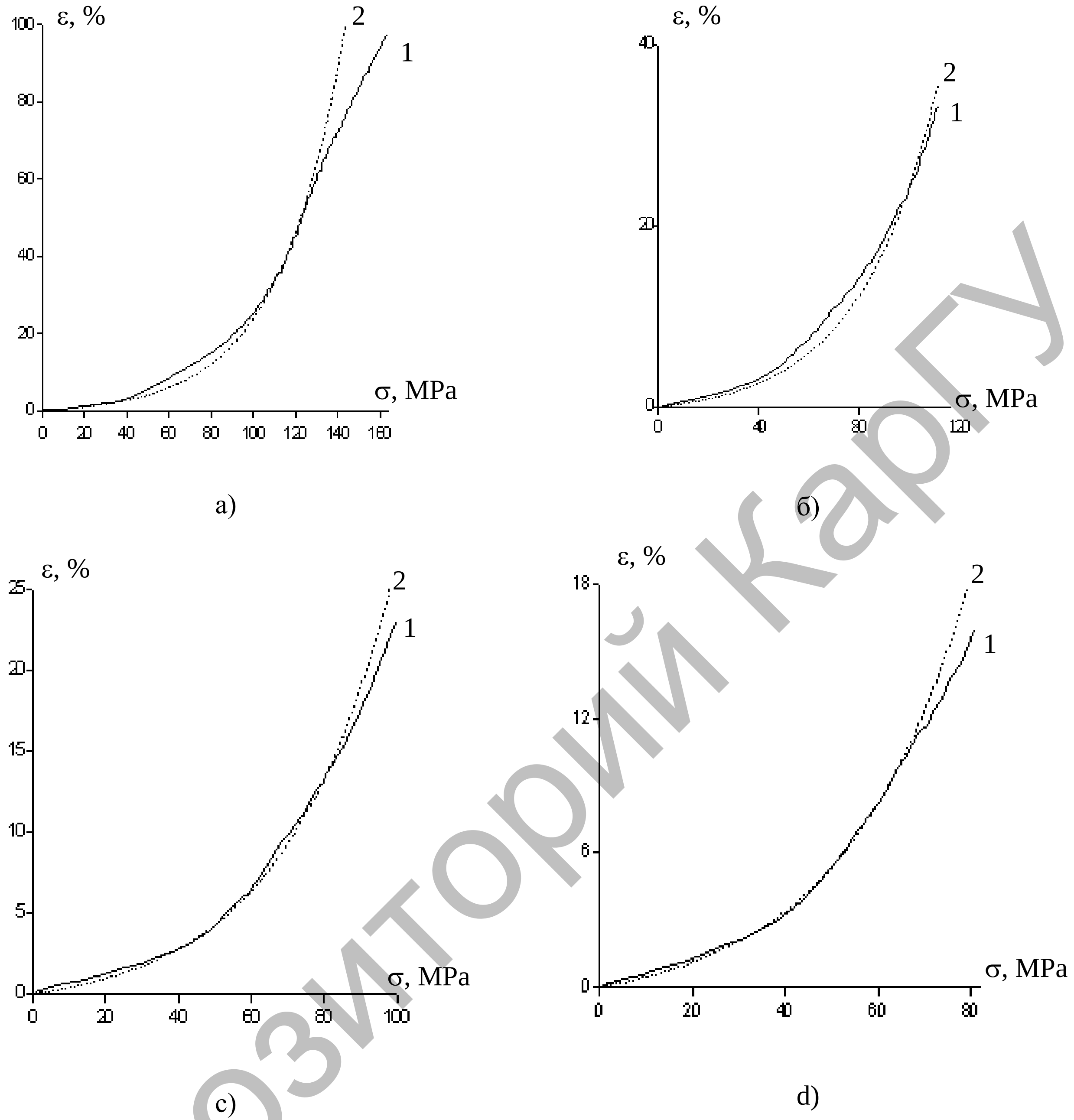

Fig. 3 Dependence of relative elongation on tension for metallized polyimide film irradiated by electron dozes D = 10 MGy(a), 20(b), 30(c), 40(d); 1 – experiment; 2 – calculation

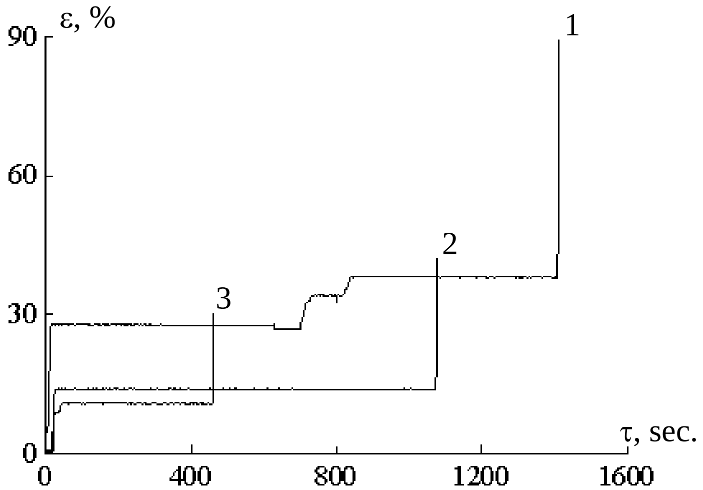

Fig.4 Dependence of relative elongation on heating time for metallized polyimide film irradiated by electrons; 1 - D = 0, 2 - D = 20, 3 - D = 40 MGy

Technology of polyimide manufacturing influences a lot upon electrical properties of metallized films, which leads towards differentiation of surface structure of the two sides: gloss and matt. And it is proved that gloss side has got better optical properties that matt one and matt side has got less electrical resistance. Effect of electron irradiation upon electrical properties of metallized polyimide films at different metal concentrations was investigated ξ (1N – low level, 2N – middle level, 3N – high level). Resistance was measured before and after irradiation at different dozes and different sample lengths: 10, 20, 30, 40, 50 и 60 mm.

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Conclusions

Series of experiments of complex effect of irradiation dose and metallization degree on mechanical properties of the films have been conducted.

It is found out that metallization of polyimide films causes increase of relative elongation up to 120%; applied tension reaches ~ 175 MPa. It is related to peculiarities of nanostructures formation based on benzene ring (1-3 nm) and conditions of chemical etching. It proves by means of IR and ESR spectroscopy [4].

When irradiation dose D increases, relative elongation decreases and material strength becomes worse respectively. When irradiation dose D increases the essential increase of electrical resistance is observed for all types of metallization. As figures show the suggested model describes well dependence of relative elongation of metallized film based on polyimide on tension for nonirradiated and irradiated by different doses films.

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