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Investigation of parameters of transparent conducting periodic structures manufactured by laser ablation

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In this paper we report on results of an investigation of a transparent mesh electrode in a wide wavelength range 550–2000 nm with low electrical resistance manufactured by laser ablation with using an aluminum film with a thickness of 30 μm on a polyimide substrate with a thickness of 20 μm. The method is implemented using a fiber ytterbium nanosecond laser with a wavelength of radiation 1064 nm. The structure we have obtained has a transmittance $> 50\%$ in the designated spectrum range and the electrical resistance $< 1.5\,\Omega$/sq.

Keywords: transparent electrodes, conductive periodic structures, laser ablation, pulsed laser radiation, fiber laser, laser optics, laser technologies.

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Introduction

Metal periodic (mesh) structures can be used as transparent conductive electrodes in such optoelectronic devices as liquid crystal displays [1], displays based on organic light emitting diodes (OLEDs) [2], touch panels [3], solar modules, etc. Such structures with periodically repeating transmission „windows" must have both high transmission of optical radiation and low electrical resistance, as well as great flexibility and mechanical strength, since most modern devices require direct mechanical contact with a film mesh electrode, which must be stable to external influences. Therefore, at present, in addition to the traditional indium tin oxide (ITO), transparent in the visible region of the spectrum, but at the same time fragile, processed in production with a large amount of waste, expensive and rare material, transparent electrodes are being developed from alternative materials. These include carbon nanotubes [4], zinc oxide doped with boron (ZnO: B) [5] or aluminum (ZnO:Al) [6], tin oxide doped with fluorine (SnO_2:F), cadmium oxide doped with indium (CdO:In), pure metals: silver (Ag), aluminum (Al), copper (Cu) [7] and etc.

To date, various methods are used for manufacturing transparent conductive mesh structures [8]. Thus, researchers who used the technology of selective laser sintering of colloidal silver managed to establish that with a cell size of $> 300\,\mu$m, it is possible to obtain a transmittance of $\geq 85\%$ and a resistance of $\leq 30\,\Omega$/sq for a mech structure 50×50 mm with square transmission windows and conductors $11-12\,\mu$m [3] thick. The transparent copper structure, fabricated by electrospinning of polymer fibers followed by metal spraying, had the transmittance of 90% with the resistance of $2\,\Omega$/sq [9]. In the work [10], using jet printing, the two-layer transparent electrode was obtained, consisting of silver mesh and ITO layer; with square cell width of $300\,\mu$m and silver conductor thickness of $7.5\,\mu$m, the transmittance of 83.25% and the resistance of $1.4\,\Omega$/sq were achieved. Photolithography has also been used to produce a transparent metal structure. It was possible to achieve the transmittance of about 54.2% in the visible range of the spectrum and the resistance of $2.2\,\Omega$/sq for 200 nm thick silver mesh coated with 30 nm thick ITO layer, with distance between two adjacent conductors of about 200 *mu*m [11]. Structure with periodically repeating conductors, obtained by applying the flexible polydimethylsiloxane stamp with copper conductors to the substrate of polyethylenedioxythiophene : polystyrene sulfonate (PEDOT : PSS), had the transmittance of 80% and the resistance of $9.3\,\Omega$/sq [12]. Using laser ablation to produce the transparent electrode, specialists obtained the 36×36 mm copper structure with large number of holes $150\,\mu$m in diameter, the transmittance of which reached 67%, the resistance was about 20 $Omega$/sq [7].

Of all the above mentioned methods for manufacturing transparent conductive periodic structures, it is worth highlighting the electrospinning of the fiber followed by metal spraying, since using this technology it was possible to obtain the highest values of the transmittance and electrical conductivity [9]. However, in recent years, researchers have considered using the laser ablation method to create large mesh electrodes due to its relatively low cost and simplicity, as well as its high processing speed. In this work, the possibility of manufacturing a transparent conductive mesh structure from Al deposited on a polyimide (PI) substrate by laser ablation using a laser engraver operating in a nanopulse mode is considered.

Experimental part

The process of manufacturing the transparent conductive periodic structure from Al film consists of two stages. The

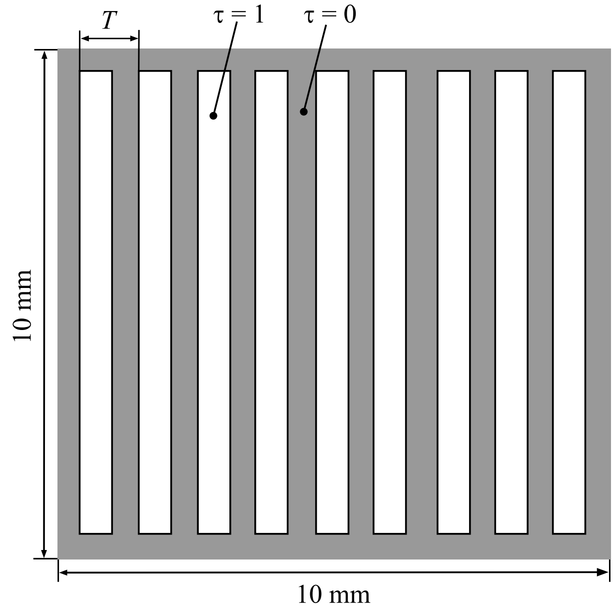

Figure 1. Model of the periodic structure. T is period.

first stage is to develop a drawing-layout of the future mesh structure in a graphic editor. In our study, the pattern layout of the structure is the square with a side of 10 mm, in which repeating lines with repetition period T are located (Fig. 1). In Fig. 1, the areas subjected to laser ablation are marked in white, and the region of residual metal is marked in gray. The second stage is the actual process of applying the developed pattern to the Al-film by means of laser ablation using the laser engraver PEDB-400B. As a radiation source, we will use the IR fiber laser, which is widely used in laser systems and technologies [13–17].

The PEDB-400B laser engraver, which is used in our study to produce a transparent Al structure on a PI substrate, includes the following main elements: the ytterbium (Yb) fiber laser with emission wavelength of $\lambda = 1064$ nm providing scanning of the laser beam over the treated surface using two mirrors mounted on galvanometers. Laser has the output power range of 10–30 W, pulse repetition frequency range of 20–80 kHz, and pulse duration of 100 ns. The scanning head makes it possible to scan the surface at speeds up to 7000 mm/s. The operation process of the laser engraver is carried out under the control of a personal computer, with the help of which it is possible to change the laser parameters and the scanning speed. To focus the laser beam on the processed surface, an SL-1064-112-163G F-Theta object lens with rear focal length of 163 mm is used, the field of view of which in the image space is characterized by maximum object area of 112×112 mm. The F-Theta-object lens, unlike a single spherical lens, does not introduce field curvature and allows focusing off-axis beams propagating at different angles to the optical axis of the object lens, on the processed surface. The object lens used by us makes it possible to obtain the waist diameter of the laser beam $\sim 40\,\mu$m.

In our study, experiments are carried out on the preparation of transparent conductive periodic structures with an area of 10×10 mm from an Al film with thickness of $30\,\mu$m, deposited on a PI substrate with a thickness of $20\,\mu$m, using the laser ablation with a laser pulse duration of 100 ns. To reduce the mechanical deformations of the Al film caused by uneven heating of the material under the influence of laser radiation, the steel plate is placed under the sample, and neodymium magnets are installed on top of the sample along the contour of the processed area 10×10 mm (Fig. 2). Magnets compress the Al-film, thereby improving the quality of the resulting mesh structure.

In our study, to measure the transmission of optical radiation in a wide spectral range of 350–2000 nm with a step of 2 nm, we use a Shimadzu UV-3600i Plus spectrophotometer with an integrating sphere with a resolution of 1 nm at normal light incidence on the sample. The integrating sphere makes it possible to estimate the transmission from the total flow through the sample. The transmission measurement is carried out in several local parts of the sample, and the transmission of the mesh structure is calculated as an average value. For detailed study of the topology of manufactured transparent mesh structures, we use a Leica s9i stereomicroscope. Surface resistance is determined by the four-probe method using Keithley 2000 multimeter and Mill-Max 854-22-004-10-001101 four-probe head.

With the pulse repetition rate of 50 kHz, laser output power of 27 W, 45 laser beam passages at a scanning rate of 4000 mm/s, it is possible to obtain mesh structures with $T \approx 70, 80, 90\,\mu$m (Fig. 3). Such structures represent microline arrays, where the thickness of the removed line is $\sim 40\,\mu$m, which corresponds to the technical characteristics of the F-Theta object lens used.

In our study, an experiment is also being carried out to create transparent mesh structures by changing the scanning speed and the number of passes of the laser beam over

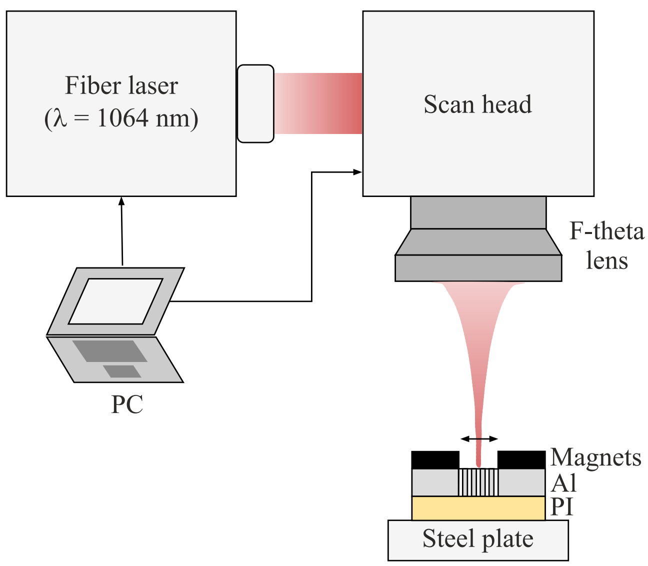

Figure 2. Experimental unit scheme.

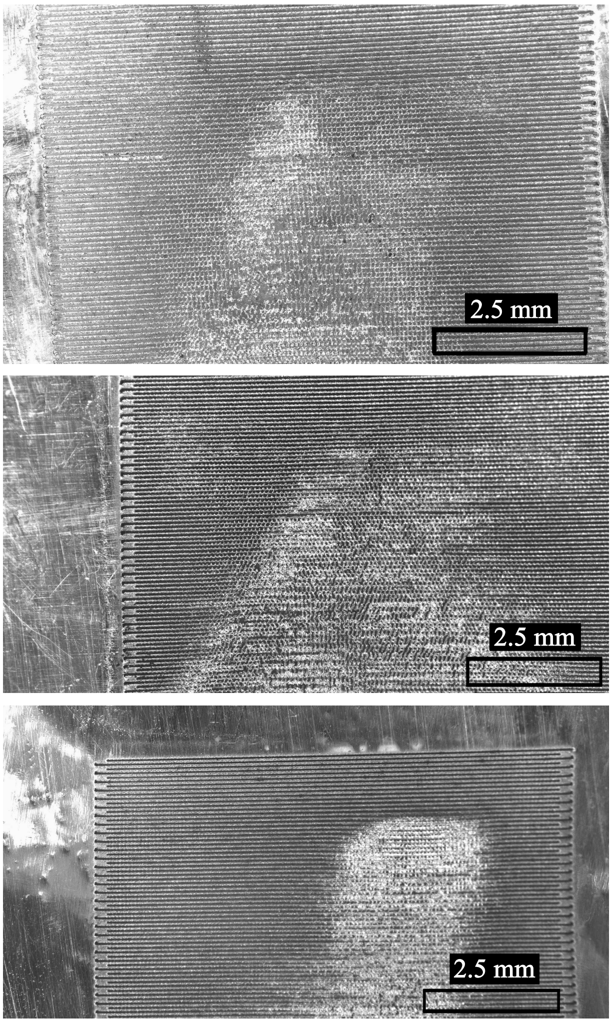

Figure 3. Structures with period T obtained in the I experiment $70\,\mu$m (a), $80\,\mu$m (b), $90\,\mu$m (c).

the treated surface, is conducted. First, laser processing is performed 15 times at a scanning speed of 4000 mm/s, then 25 times at a speed of 6000 mm/s. As a result of such transition to „easy“ processing mode, it is possible to obtain mesh structures without rough areas on the surface c $T \approx 70, 80, 90\,\mu$m (Fig. 4).

To measure the transmittance of a transparent conductive mesh structure, it is necessary to understand that in our study we use an Al film deposited on a PI substrate with a thickness of $20\,\mu$m. In order to remove the Al layer and then measure the transmission spectrum of the PI substrate only (Fig. 5), it is necessary to use a laser engraver. By measuring the transmission spectra in the wavelength range $350-2000$ nm of the PI substrate and a two-layer sample, it is possible to construct the transmission spectrum of a transparent periodic Al structure using the formula for calculating the spectral transmittance of the Al structure:

$$\tau_{\lambda \mathrm{Al}}(\lambda) = \frac{\tau_\lambda(\lambda)}{\tau_{\lambda \mathrm{PI}}(\lambda)} \cdot 100\%,$$

where $\tau_\lambda(\lambda)$ is the spectral transmittance of the two-layer sample, $\tau_{\lambda \mathrm{PI}}(\lambda)$ is the spectral transmittance of the polyimide substrate.

Results and discussion

The result of the first experiment is to obtain transparent conductive periodic structures in the form of an array of microlines with $T = 70, 80, 90\,\mu$m (Fig. 3). It can be seen that the prepared structures have rather large rough areas in the center, which are the result of the release of molten Al from the ablation craters to the surface. If the height of the surface irregularities exceeds the wavelength of the incident radiation, then diffuse scattering occurs and, as the consequence, the decrease in the transmittance. Thus, the transmittance in the visible region of the spectrum of the mesh structure with $T = 90\,\mu$m, which was obtained from the results of the first experiment, is equal to 34.98% at the wavelength of $\lambda = 555$ nm (Fig. 5, a). It was also possible to prepare the mesh structure with $T = 70\,\mu$m, the transmittance of which at the wavelength of $\lambda = 555$ nm is equal to 33.26%, and the maximum transmittance in

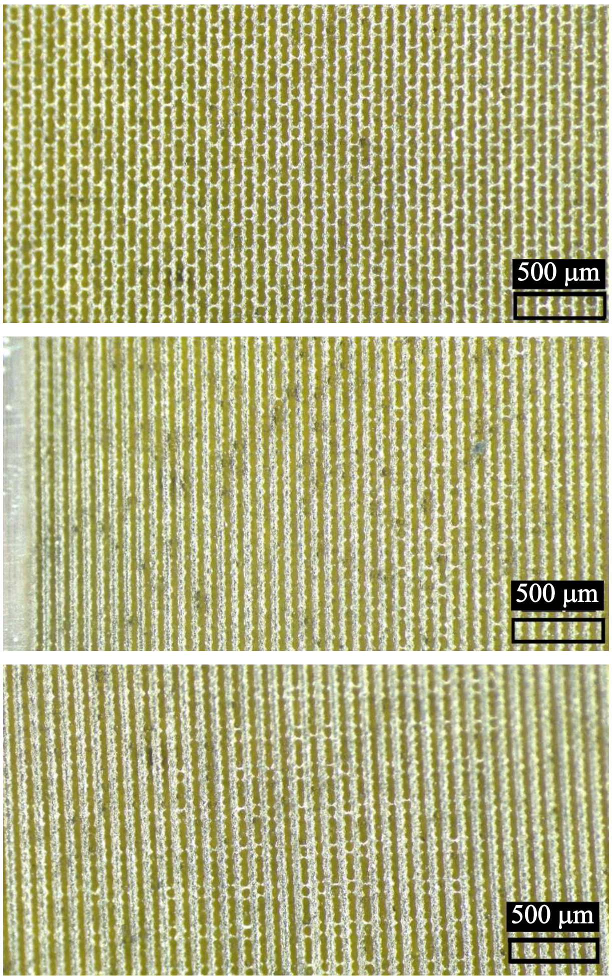

Figure 4. Structures with period obtained in the II experiment $70\,\mu$m (a), $80\,\mu$m (b), $90\,\mu$m (c).

the near infrared region of the spectrum is equal to 50% ($\lambda = 1950$ nm).

In order to reduce the surface roughness and obtain the mesh structure with high transmission, it was decided to reduce the duration of laser radiation exposure to Al by changing the number of passes and the scanning speed of the laser beam. As a result, it was possible to prepare high-quality transparent conductive mesh structures with high transmission of optical radiation compared to the transmission of mesh structures with rough areas. Thus, the transmittance of the mesh structure with $T = 70\,\mu$m exceeds 50% in the wavelength range 550–2000 nm (Fig. 5, *b*). It can be observed that the transmission of optical radiation decreases with the increase in T due to the decrease in the number of transparency windows and the increase in the area of the residual metal. In our study, the dependence of the electrical resistance of a transparent mesh structure on the repetition period of microlines T was also revealed. As T changes from 70 to 90 μm, the electrical resistance decreases from ~ 1.4 to 1.07 Ω/sq.

It is assumed that the type of structure topology affects its parameters. Therefore, in order to obtain transparent conductive mesh structures with the higher transmittance, it is necessary to develop a different layout pattern, as well as use other materials as a conductive layer.

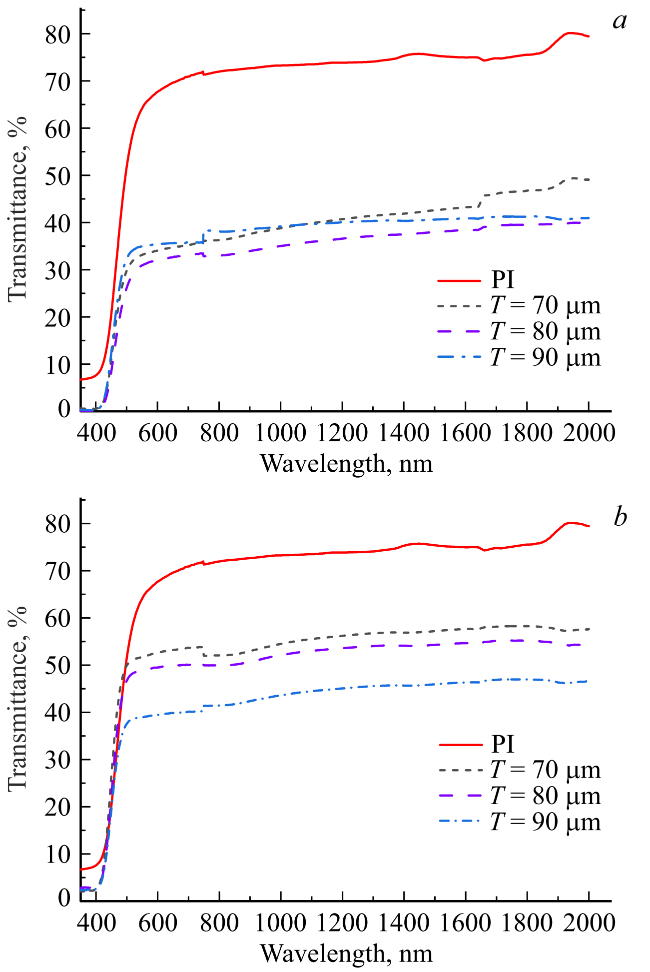

Figure 5. Transmission spectra of transparent conductive periodic structures obtained in I (*a*) and II experiments (*b*).

It is worth noting that the laser ablation method used in the presented experiments and the PEDB-400B laser engraver are suitable for making large mesh structures. The sample area is determined solely by the operating field of the F-Theta object lens, which can be replaced if necessary. In our case, the object lens with operating field of 112×112 mm is used, respectively, the maximum area of a possible mesh structure made using this lens is 12544 mm^2. In addition to the fact that the laser ablation method makes it possible to obtain structures of large sizes, it provides the high speed of their preparation. Unlike expensive and multi-stage technologies [3,9–12], the technological process of preparation transparent conductive periodic structures by laser ablation consists of only 3 stages: development of the topology of the future structure, laser processing and control of the parameters of the resulting structure. It is evidence that the process of preparation mesh structures by laser ablation is carried out much faster in time than by other methods.

Conclusion

In the course of this work, it was possible to prepare high-quality transparent current-conducting periodic 10×10 mm structures from a 30 μm thick Al-film deposited on 20 μm thick PI substrate by laser ablation. . From the structures obtained, it is necessary to separate out a mesh structure with microline repetition period $T = 70\,\mu$m, in which the optical radiation transmittance is $> 50\%$ in the wavelength range $\lambda = 550$–2000 nm, and electrical resistance $\sim 1.4\,\Omega$/sq. Such a structure can be used as a transparent conductor in optoelectronic devices operating in the visible and near infrared spectral ranges.

On the basis of the obtained results and the data of works [3,9–12] it was concluded that laser ablation is a high-performance method for preparation transparent conductive periodic structures from various materials. Also, this laser technology makes it possible to create structures of large sizes. The periodic structure prepared in our work by laser ablation has the transmittance comparable to that of periodic structures obtained by other methods using more expensive auxiliary materials and equipment [11].

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Conflict of interest

The authors declare that they have no conflict of interest.

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