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Photothermoplastic films as recorders in observation systems of zonal structures

*Vasile K. Rotaru[a], Igor V. Dementiev[a], Oleg Ya. Korshak[a], Sevastyan N. Neamtsu[a], Stephan V. Robu[b], Hossin A. Abdeldayem[c], Igor V. Ciapurin[d], Nickolai V. Kukhtarev[e]
[a]Dept. of Physics, Moldova State Univ., 60 A.Mateevici str., Chisinau, MD-2009, Moldova
[b]Dept. of Chemistry, Moldova State Univ., 60 A.Mateevici str., Chisinau, MD-2009, Moldova
[c]NASA Goddard Space Flight Center, Code-554, Bldg. 19, NASA-GSFC, Greenbelt, MD 20771
[d]School of Optics/CREOL, Univ. of Central Florida, P.O. Box 162700, Orlando, FL 32816-2700
Dept. of Physics, Alabama A&M Univ., 4900 Meridian str., Normal, AL 35762

ABSTRACT

Photo-thermo-plastic films (PTPF) with high-density recording (up to 1000 lines/mm) can be used repeatedly for structure zoned shooting. However, more work is underway to increase their light sensitivity and make it resistant to action of radioactive and powerful electromagnetic radiation. It was found that chalcogenide glass semiconductors (CGS) combine properties of both glasses and semiconductors that determine possibility of its using in different systems of optical data recording. Most bright representatives of glass semiconductors are being sulfide and selenide of arsenic. Results of complex investigations on photoelectric properties of their thin layers are presented.

The paper presents results of technological changing of the field of maximal photosensitivity of CGS layers in the range of optical spectral range from 400 to 800 nm, which satisfies requirements of optical data recording systems for spectrum-zoned-shooting. For increasing of CGS thin layers' photosensitivity the initial materials were doped by tin. Experiments have shown that doping by Sn on the level 1.2-1.4 at.% increases photosensitivity of layer by more than one order of magnitude. High photosensitivity of obtained PTPF determines possibility of their wide application in different optical image registration systems, which can be used as board memory devices.

Keywords: photothermoplastic films, remote sensing, chalcogenide glass, photosensitivity, resolving power

1 INTRODUCTION

Photo-thermo-plastic films (PTPF) are non-silver two-layered structures for optical data recording [1]. Semiconductor-thermoplastic configuration provides recording of photographic, holographic, and other types of optical data without a wet chemical development, giving ones the better radiation resistance. Optical data recorded on photothermoplastic (PTP) media is characterized by simplicity of development and erasing processes; unlike xerography it does not need any special developing material; unlike CCDs it does not need any complex electronic interface. The property of radiation resistance makes these systems particularly useful for remote sensing of different objects from Space.

This report sums up the experience in fabrication and practical use of PTP recording structures. Recording on PTPF has the resolving power that exceeding one in usual photographic systems. Up to date, the main disadvantage of PTPF is its relatively low photosensitivity, and number of PTPF applications is also restricted by phase nature of recorded images.

Usually PTPF contains several more layers for improving of its parameters. The first layer is an adhesive one which is deposited on polymer substrate. The second layer is semitransparent electrode. The third layer is being deposited upon the electrode and is in charge of electron injection. The fourth layer is a barrier one, often represents by a heterojunction, and then transport layer is followed. As previous investigations showed (see Ref. [2]), the semiconductor layer weakly affect photographic parameters of PTPF.

* E-mail address: rotaru_vk@yahoo.com

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Visualizing layer (thermoplastic) could also be modified. Alteration of the type of the used polymers, their additional doping, creation of visualizing multi-layers, and changing of the surface properties of thermoplastic layer allows varying of thermo-development regimes, i.e. temperature diapason and potential of charging while the optical data record [3].

2 PARAMETERS OF GLASSY CHALCOGENIDE LAYERS

The variety of the constructions of charging devices is dictated by the purposes of a particular recording device. A successful explanation of the optimal regimes for each type of the charging device has been found. Collected data is being used for PTP recording and also to predict the behavior of the surface potential $V_s(t)$ on PTPM using characteristics of the known charging devices.

Kinetics of the surface potential V_s at the stage of charging of the photosensitive layer of As_2S_3, As_2Se_3, and their solid solutions proves a complex nature of the processes inside the material. It was rigorously studied for the different corona producing devices of different efficiency. With the growth of the potential the maximum appears on the curves of $V_s(t)$. The subsequent decreasing of V_s we tend to connect with the growth of the space charge at the lower levels. The screening of the surface potential by this charge leads to apparent decreasing of the effective thickness of the layer. In this case, the surface potential V_s, determining the light sensitivity of PTPM, is formed by the charge, accumulated on the surface and inside the bulk of the layers as well as in between them. Using such simple model, the effective deepness **b** of the charge bedding and their amount **N** were calculated.

For the As_2Se_3 layers with thickness d=2.4 μm, the values **b** an **N** were: **b**=1.5 μm, **N**=$2\cdot10^{17}$ cm^{-3}. For the layers consisting of the solid solution $(As_2Se_3)_{0.7}(As_2S_3)_{0.3}$ and d=3.0 μm, these values were 1.9 μm and $3\cdot10^{17}$ cm^{-3} correspondingly, and for As_2S_3 layer with thickness d=2.2 μm the corresponding values were: **b**=1.6 μm and **N**=$7\cdot10^{17}$ cm^{-3}. These values of the accumulated charge concentrations are in good agreement with the values of densities of localized states in these materials. The growth of illumination is increasing the field of predominant of the bedding of compensating charge that leads to the more pronunciated maximum on the $V_s(t)$ curve. Such dependence is not observed at the temperature growth up to 80˚C that means that effective deepness of charge bedding is not changed in this case.

A number of requirements have to be met in the process of fabrication of PTPF based on chalcogenide glasses. Among them are high specific resistance (10^{13}-10^{14} Ohm·cm), dark conductivity is of less importance), mechanical durability, thermal stability of electro-physical parameters and recycling of the used materials. The basic requirements to the photosensitive semiconductor layers affiliated to the PTPF structure are their high resistance and large multiplication ratio of resistance change under the electromagnetic irradiation exposure. In spite of the successes in development of electrophotography and PTP recording on the base of the materials mentioned above, currently there are remained the actual tasks of photosensitivity increasing, widening of spectral range for recording, and realization of new properties of media for information recording. For its successful application in two-layer PTPF photosensitive semiconductor layer should have dark resistivity about ρ_d~10^{12}-10^{14} Ohm·cm at the multiplication ratio of photo-response at the level of 10 at the minimal illumination of 5-10 Lx. Modification of composition allows purposeful changing of the properties of thin films and obtaining the layers, satisfying the requirements of any practical needs. Also, one of the important tasks in the field of recording of optical information from space is the search of the materials of recording media, possessing high photosensitivity and transparency for polychromatic light including monochromic light of definite wavelength.

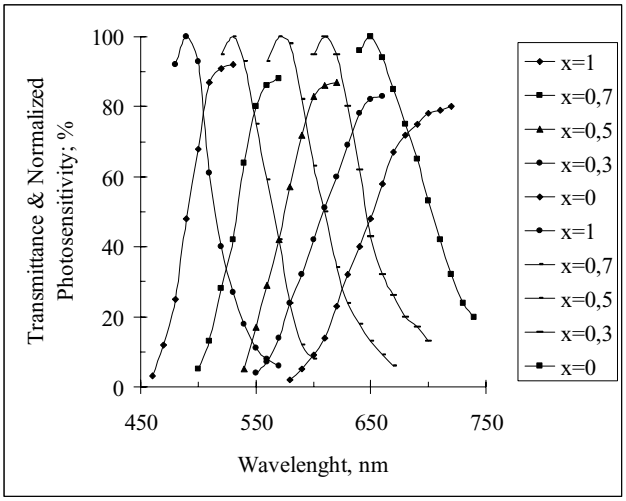

Fig.1. Interrelations between transmittance and normalized photosensitivity for $(As_2S_3)_x(As_2Se_3)_{1-x}$ layers.

Comparison of the properties (see Fig.1) was carried out in accordance with the following procedure. At the same graphs there were built the dependences of transmission T(%)=f(λ) and long-wave decay of the photocurrent

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$I_p(\%)=f(\lambda)$ for thin films of different composition. With account of peculiarities of the photo-effect in thin layers of chalcogenide glass semiconductors, mentioned in [4] the photocurrent was measured at the negative polarity of illuminated electrode. At the comparison of solid solutions the relative values of photocurrent were put in Y-direction. Values themselves had been determined in accordance with formula

$$I_p(\%) = \frac{I_p}{I_{p\max}} \cdot 100\%$$

Here I_p is the absolute value of photocurrent at the given wavelength of descending light, $I_{p\ max}$ – maximal value of photocurrent for given material. The projection of point intersection of T and I_p curves on the axis of wavelengths λ gives ones the optimal wavelength of the descending light λ^{max} at which material is satisfying to the requirement of largest sensitivity and transparency. Hence, the carried out comparison of the properties of thin films of solid solutions system $(As_2S_3)_x(As_2Se_3)_{1-x}$ has shown the opportunity to choose the thin film composition, which can be used in the different devices for information recording.

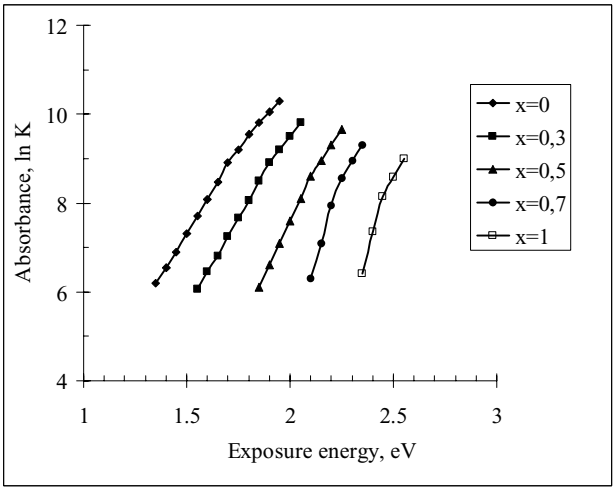

Fig.2. Dependence of the $(As_2S_3)_x(As_2Se_3)_{1-x}$ system absorption on photon energy of incident irradiation.

Measurement of the drift mobility of charge carriers was carried out through the time-flight method [5] on "sandwich" type samples with deposited chromic top electrode. The values of band gap width determined from the curves of adsorption in coordinates $(h\nu K)^{1/2}=f(h\nu)$ and from the spectral characteristics of photoconductivity (by the wavelength of monochromic light at which photocurrent decreases up to 50% of maximal value) are coincided. Width of the bandgap is linearly decreased with the composition alteration from E_g=2.4 eV (As_2S_3) to the value 1.75 eV (As_2Se_3). We determined that E_g values are practically congruent with values presented in Refs [6,7]. Dependence of the coefficients of adsorption of thin films $(As_2S_3)_x(As_2Se_3)_{1-x}$ on the photons energy of incident irradiation in the field of weak adsorption ($h\nu<E_g$), for x: 1 – 1.0; 2 – 0.7; 3 – 0.5; 4 – 0.3; 5 – 0 is presented in Fig.2.

Fig.2 shows that in the field of weak adsorption ($h\nu<E_g$) all compositions there have exponential dependence of adsorption coefficient on the energy of descending phonons as an evidence of Urbach's law feasibility: $K=K_0\exp[(h\nu-E_g)/\Delta]$. Feasibility of this rule, generally speaking, points to the presence of "tail" of state's density in bandgap. Charge carriers possess the greatest mobility in the layers As_2Se_3 ($\mu\sim10^{-7}$ $cm^2/V\cdot c$). As the arsenic sulfide content growths the drift mobility of carriers is decreased and achieved the value of $\mu\sim10^{-10}$ $cm^2/V\cdot c$ in the case of $(As_2S_3)_{0.7}(As_2Se_3)_{0.3}$. Temperature dependence of the drift mobility has the exponent form: $\mu\sim\exp(-E_a/kT)$, where E_a is energy of activation of mobility. Dependence $\lg\mu=f(10^3/T)$ allows to find out the energy of trap's activation which are participating in the charge transfer. Determined value of the activation energy E_a for arsenic selenide is 0.1 eV; for solid solutions E_a is increased up to 0.18 eV for the $(As_2S_3)_{0.7}(As_2Se_3)_{0.3}$ composition. The sensitivity spectra of the PTPF on base chalcogenide glass semiconductors are presented in Fig 3.

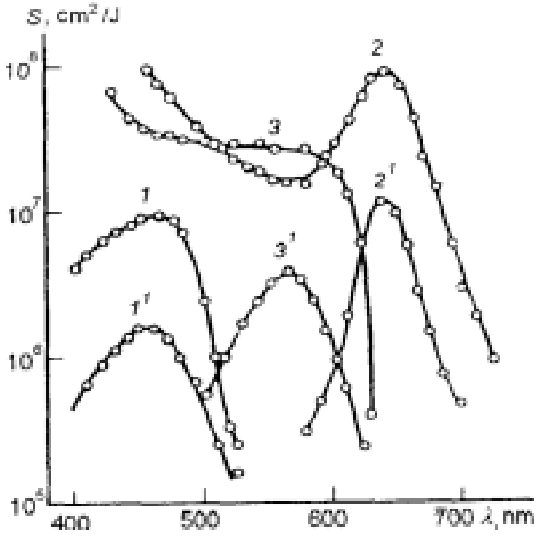

Fig.3. Spectral sensitivity of PTPF based on: As_2S_3 (1,1'); As_2Se_3 (2, 2');$(As_2S_3)_{0.3}\cdot(As_2Se_3)_{0.7}$ (3, 3').

These results could be successfully applied to systems of photothermoplastic recording for creation of PTPF from $(As_2S_3)_{0.5}(As_2Se_3)_{0.5}$ layers and heterojunctions on their base; however, the high speed of the relaxation of the dark surface potential limits their application in simultaneous recording

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procedure. The performed investigations allow determining the composition in doped As-S-Se systems with different percentage of containing component, possessing high exploiting parameters. Studying of optical, photoelectrical, and electrophotographical properties of As-S-Se-Sn compounds shows that these layers possess darkish specific resistance of $3\text{-}5 \cdot 10^{14}$ Ohm·cm at T=30°C, and the changing brevity of resistance is $K=10^2$ at E=10 lx. When the temperature rises up to the values used during PTP recording (~70°C), an insignificant deterioration of chalcogenide semiconductor parameters occurs. Maximum of spectral sensitivity of such layers is dependent on the layer composition and located within the range 460-480 nm in which multiplicity of the photo-response $(1.8\text{-}11)\text{-}10^3$ is ensured. When He-Ne laser (λ=630 nm) is used, the photo-response multiplicity constitutes from 20 to 70 while irradiation power is 10^{-5} W/cm^2. These compositions can be used for creation of PTPM with sensitivity from $1.4 \cdot 10^{-6}$ to 10^{-5} A/W. Thermal dependence of the conductivity of multi-component chalcogenide thin layers for temperature range 310-360 K and applied electric field is shown in Fig.4. For the first time there were obtained photosensitive layers with $\rho_d \sim 7 \cdot 10^{14}$ Ohm·cm and K=30 at the power of incident irradiation $8 \cdot 10^{-7}$ W. These layers, according to their sensibility, are close to traditional in electrophotographical selenium plates, thermally stable and can be applied for both simultaneous and consequent procedures of optical information recording; moreover, they possess high transparency (T=90% at λ=630 nm). These results allow to conclusion that these layers can be successfully used for preparation of two-layer PTPF capable to register optical information in diapason 400-700 nm. Studying of influence of electric field polarity on photocurrent and on light response showed that maximum values reach when negative potential applied to alight surface of thin CGS layers.

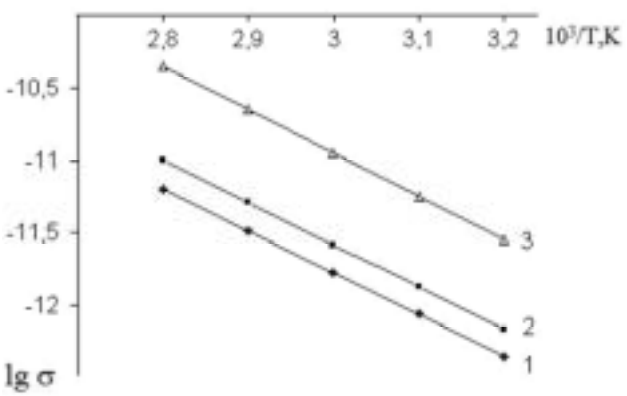

Fig.4. Thermal dependence of the conductivity of As-S-Se-Sn thin layers: $1\text{-}10^4$; $2\text{-}4\text{:}10^4$; $3\text{-}\ 8 \cdot 10^4$ V/cm.

Using PTPF production on a base of CGS thin films, we ensure that their spectral sensitivity covers X-rays, visible, and near-IR ranges. Also, one can select a wavelength range with a width of about 0.1 μm in which some of PTPFs are able to ensure photographing of objects in these spectral zones. Technologically, these CGS layers were created on flexible 40-m-long and 190-mm-wide films with thickness accuracy of better than 0.2 nm and electric parameters deviations not more than ±5%.

3 PARAMETERS OF THERMOPLASTIC LAYERS

During the search of new thermoplastic materials with improved deformation and resolution capabilities for the creation of PTPF, we have synthesized and studied a number of copolymers of carbazolyl-N-alkylmethacrylates (CAM) with octylmethacrylates (OMA) and with cetylmethacrylates (CMA) with composition of CAM parts from 50 to 80 mole%. Two types of thermoplastic layers: copolymer CAM:OMA 60:40 mole% - type **I**, and copolymer CAM:CMA 60:40 mole % - type **II**, were studied. It was established that replacement of OMA-parts with CMA-parts in copolymers significantly increases the deformation characteristics of the layers due to better plasticization of polymer by cetylmethacrylate parts. It was found that for attainment of maximal deformation and resolution capability the optimal temperature for type I copolymer makes up 85-87˚C but for copolymer of type II it amounts 70-73˚C. The dark specific resistance ρ_{dark} is equal to $2 \cdot 10^{15}$ Ohm·cm for copolymer I and $5 \cdot 10^{14}$ Ohm·cm for copolymer II at 21˚C temperature. With the temperature growing up to ~71˚C which is the recording temperature of PTP process, the ρ_{dark} value for is decreased both copolymers by one order of magnitude and became within the same order of magnitude as studied photosensitive semiconductor layers as it was described above. The last is being the optimal combination for creation of two-layer PTPF with improved exploitation characteristics.

Also, thermoplastic materials BMA-50 (copolymer of styrene with butylmethacrylate, 50:50 mole%) and CEM-CMA (copolymer of the carbazolylethylmethacrylate with cetylmetacrylate) in conjunction with photosensitive CGS materials were studied. For estimation of quality of an image recorded on PTPF, procedure and experimental setup developed by Laboratory for Photothermoplastic Recording of Moldova State University were used. One can precisely determine all

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photographic characteristics for different conditions and modes of the PTP recording within the single test. This procedure consists in several steps. Firstly, standard transmission gray scale test (TGST) was projected on PTPF and recorded on it. Then TGST was read-out by means of visualizing optical system and its image was characterized by the corresponding ratio of the photodetector currents I_b/I_d measured in bright and dark fringes of reproduced test. Value $lg(I_b/I_d)$ was accepted as a criterion for estimation of the reproduced image quality in accordance with the Weber-Fechner law. Therefore, $lg(I_b/I_d)=f[lg(Et)]$ dependence serves as a characteristic curve for PTP recording. It allows calculate contrast coefficient as a characteristic curve slope $\gamma=\tan(\alpha)$,photographic width $L=lg(Et)_2-lg(Et)_1$, and photographic sensitivity $S=1/Et$. Coefficient of transformation for this read-out setup was assumed as unity. The $(Et)_{min}$ value was determined at the 0.2 level above the background value. A set of curves for surface potential relaxation for elaborated PTPF is shown in Fig.5.

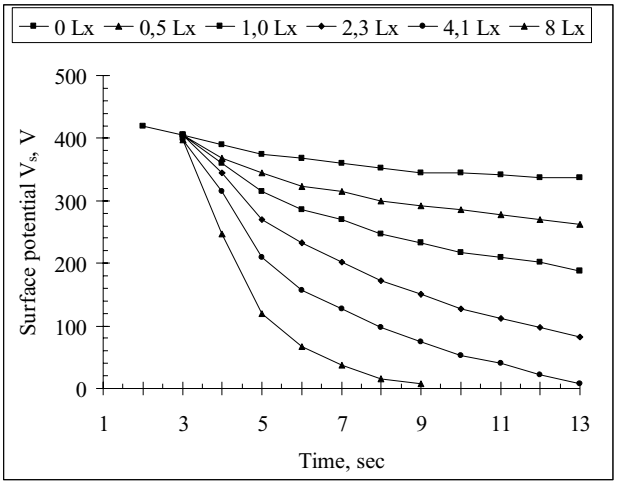

Fig.5. Surface potential relaxation for different levels of PTPF illumination.

Superficial dimples and craters appear nearly of the same size onto PTPF. It is known that resolution capability especially influences on the quality of recorded PTP image. Maximal resolution during recording of GOI test pattern on PTPF samples was about 350 mm^{-1} with "Industar-70" objective. However, the last one does not limit the PTPF resolution because resolving power is more than 3,000 mm^{-1} for holographic recording. Obtained results on determining of basic photographic characteristics of the investigated media were adequately described by means of mathematical model on kinetic processes in PTPF.

PTPF seems to be smart material for large area of technical applications [8]. However, due to recording of half-tone images by difference in deformation densities, the strong scattering circle appears around the central maximum formed by the spectrum of chaotic deformations [9]. The central part of the scattering circle has the maximum intensity and determines the resonant frequency, at which the maximum occurs in the dependence of the diffraction efficiency on the spatial frequency. As a matter of fact, the resonant frequency determines the resolving power. The influence of PTPF thickness on the resonant frequency has been well studied both experimentally and theoretically [9,10]. Let us note that the correlation between the thermoplastic layer thickness and resonant frequency is considered constant and not depended on the recording conditions.

4 PTPF-BASED RECORDING DEVICE FOR REMOTE SENSING

A tendency to wide the dynamical diapason of recorded images towards the minimal luminous flux from the space objects requires creation of photo-recording devices used special fast lenses. We used one of such lenses with the following parameters: focal length f=99.3 mm; numerical aperture D/f=1:1.12; resolving power R=960 mm^{-1}, the field of view angle 8°. All PTPF-based devices imply some critical elements for providing both PTPF sensitization by means of the corona

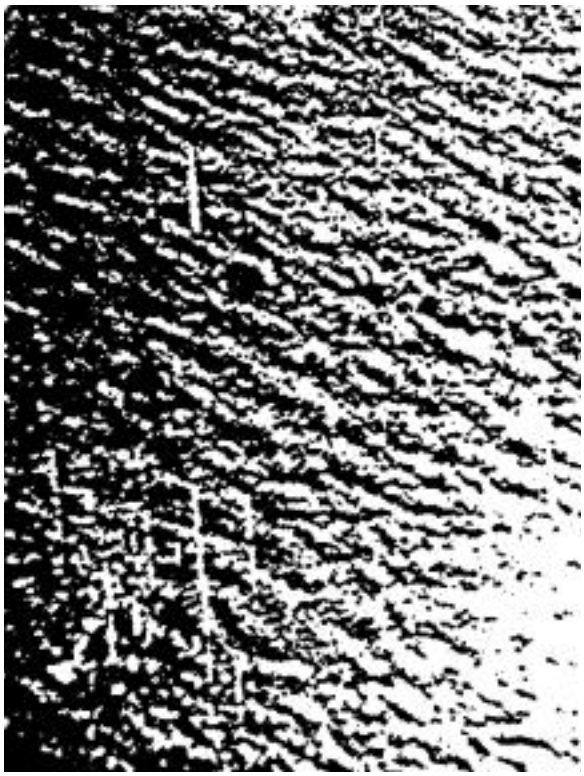

Fig.6. The Black Sea surface picture made from the 9000-m-altitude.

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discharge as well as thermal development of the latent image to the form of superficial relief on the PTPF. PTPF-based device for airborne and satellite imaging, PARUS, was described previously in Ref. [11]. Let us briefly repeat the main operation features of the PARUS. The lens provides the exposure of the PTPF photosensitive layer through the slit and the PTPF base. The microscope determines the synchronization of the PTPF movement and the Earth surface image; this also allows controlling both the quality of the image on the thermoplastic surface and its erasure during rewinding. The weight of the PARUS device is about 16 kg comprising the 7-kg-weight of the lens with the diaphragm and the shutter without the restoring or television transmission systems. To avoid overheating under the zero-gravity condition the suitable radiators at the energy-consuming components were taken off the closed cassette space to cool them outside by the air fan. The film-pulling device for these practical PARUS modifications was produced in two modifications providing the synchronization of the PTPF movement from 1.5 to 2.5 mm/s or from 2.6 to 4.3 mm/s providing optimal exposure time about 0.15 s.

Fig.7. Fragment of the panorama image of Chisinau town.

Fig.6 shows the Black Sea surface picture made from the 9000-m-altitude. The photographing with the moving device was modeled in the ground conditions by means of the special revolving table. The speed of revolution was synchronized with the PTPF movement provided by the film-pulling device. As a result under this condition from the upper floors of buildings, the panorama images of the town district were obtained. Fragment of such image created with the suitable reproducing systems is shown in Fig.7. This device could use both fresh-prepared and PTPF which had been stored up to 15 years under rather ordinal room conditions without any light shielding.

5 CONCLUSIONS

- PTPF-based devices allow optical data recording with minimal sensitivity from 0.13 to 4 $lx^{-1} \cdot s^{-1}$, average photographic width equals to 0.2, and average contrast value equals to 2.

- Current level of computers and optical image processing allow inputting the information to computer memory through the microcrater-by-microcrater mode, archiving, and transmitting it through communication channels for further processing.

- Increasing of PTPF sensitivity through the utilization of multi-component CGSs and novel polymeric compositions allows increasing the rate of information accumulation and decreasing the altitude of flight of aircraft equipped with such camera.

- Such slit camera was tested for remote sensing applications from the plane board and for in-ground panoramic photography. In the last case the slit camera was mounted on the top of adjustable table. Resolving power was about 220 mm^{-1} for the OSL-100 objective.

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