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AMORPHOUS CHALCOGENIDE SEMICONDUCTORS FOR DOSIMETRY OF HIGH-ENERGY IONIZING RADIATION

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ABSTRACT

The possible application of amorphous chalogenide semiconductors as radiation-sensitive elements of high-energy (E>1 MeV) dosimetry systems are analyzed. It is shown that some of these materials are characterized by a broader region of absorbed doses and low-temperature thresholds of radiation induced bleaching than conventional colouring oxide glasses.

KEYWORDS

Dosimetry; dose; chalcogenide; glass; thin film; optical absorption; photoconductivity.

STATEMENT OF THE PROBLEM

Solid-state dosimetry systems based on colouring oxide glasses are widely used for registration of high-energy ionizing radiation (Frank et. al., 1973; Pikaev, 1975). They are sufficiently simple in exploitation and production, stable to the influence of external actinic factors, but they do not allow registration of absorbed doses of radiation more than about 0.1-1.0 MGy. Additional inconveniences of these materials are connected with the necessity of high-temperature annealing (800-1000) K for restoration of initial optical properties. Taking the above mentioned factors into account and using our experimental results obtained during the past 15 years (Shpotyuk, 1987; Shpotyuk, 1990a, b; Matkovskii et al., 1994), we shall analyze the possibilities of practical application of amorphous chalcogenide semiconductors (AChS), which are characterized principally by another complex of physical properties in comparison with oxide glasses (Borisova, 1983), for industrial dosimetry of high-energetic radiation.

RADIATION-STRUCTURAL PROCESSES IN AChS

Arsenic sulfur (AChS) types, which were studied for the first time by Kolomiets and Goryunova fourty years ago, are unique solid-state materials showing the complex of "traditional" semiconducting properties (Borisova, 1983). At the level of atomic structure, they are inorganic polymers characterized by short-range ordering in the displacement of the various structural groups and fragments. Thus, for example, the atomic structure of amorphous arsenic trisulphide As_2S_3 (AAT) can be presented as a

network of pyramidal AsS_3 units mutually connected by bridge As-S-As complexes (Felts, 1986). However, the limited concentration of homopolar covalent bonds As-As and S-S (to 10-20 %) exist in AAT in the framework of partially polymerized complexes. These "wrong" chemical bonds are specific structural defects showing an essentially modified energy spectrum of AChS near the band gap (Elliott, 1986). The second type of structural defects in AChS are the so-called D-centres appearing as a result of external influences by pairs of oppositely charged over-(positive charge) and under-coordinated (negative charge) arsenic and sulfur atoms (Street, 1977; Shpotyuk, 1990). The whole process of coordination defect formation in AChS is realized as specific acts of destruction and polymerization transformations due to the associated variants: 1) electron-hole pair excitation in accordance with the self-trapped exciton model (Street, 1977); 2) excitation of single electron and hole pairs autolocalized at structural fragments, called soft atomic configurations (Klinger, 1983); 3) chemical bond breaking or σ-electron excitation (Matkovskii et al., 1990). It has been shown previously that coordination defect formations induced by irradiation are considered to be interconnected processes of rebonding or chemical bond switching (short-range order changes) and subsequent relaxation transformations (intermediate-range order changes) comprising a large space of the AChS network from two to five coordinated spheres (Elliott, 1986). Thus, radiation-induced structural transformations in AChS are associated at the finishing stage by two-types of changes in the defect subsystem: changes of chemical bonds distribution in the framework of various structural groups, as well as changes of anomalous coordinated atoms with uncompensated electric charge. Both transformations can be studied experimentally using the differential Fourier-spectrometry technique of induced optical absorption or reflection in the region where the main vibrational bands of structural groups lie. This technique was first applied in 1988 for photostructural investigations of AAT (Kornelyuk et al., 1988) and is based on determination of rebonding reactions stimulated by external factors. It has been established (Shpotyuk, 1990b; 1993) that, in the case of gamma-irradiation of vitreous arsenic trisulphide, the structural conformational changes are stretched in the direction of chemical bond replacements due to such reactions as:

$$(\text{As-S}) \rightarrow (\text{As-As}), \quad (1)$$
$$(\text{S-S}) \rightarrow (\text{As-S}). \quad (2)$$

The present data show that the statistical weight of (1) reaction is greater than that of (2) reaction. The subsequent annealing process causes the opposite changes in the chemical bond distribution. Hence, these structural transformations are actually reversible.

DOSIMETRIC CHARACTERISTICS OF AChS

The most sensitive of the observed radiation-induced defect formation processes in AChS are their optical, electrical and photoelectrical properties. For example, the influence of ^{60}Co gamma-ray quanta (E=1.25 MeV) in the range of absorbed doses (D=1-10 MGy) leads to the longwave shift of the AAT fundamental optical absorption edge, achieving 0.04-0.06 eV for sample thickness d=1-2 mm. When all measurements are carried out with helium-neon laser wavelength (λ=633 nm) corresponding to the middle part of AAT transmission edge and optical density (OD) is used as the controlled parameter, then the dose dependence function of the gamma-stimulated changes ΔOD/OD is linear relatively to D in the range of 0.5-10 MGy and can be established as

$$\Delta OD/OD = S.\lg D + A = 0.30 \lg D - 1.6 \quad (3)$$

Parameter S in (3) characterizes the sensitivity of the dosimetric system to the specific radiation; A is some constant.

Optical properties of AChS are stable after irradiation for no less than 10 years, if the temperature is smaller than the thermal bleaching threshold T_m. The T_m values are essentially dependent on composition of the samples. After thermo-annealing with temperatures higher than Tm, the investigated materials can be used repeatedly. The influence of fabrication conditions of AChS is considerable at the first stage of gamma-irradiation when, besides above-mentioned process of coordinated-defect formation, the homogenization of glass network and relaxation of structural macrodefects (pores, cracks, internal stresses etc.) also take place. Therefore, for dose registration with high precision, it is advisable to carry out an "idle" cycle of AChS gamma-irradiation and thermo-annealing. Dosimetric characteristics of AChS do not depend appreciably on dose power, P, when the temperature in the source cavity is less than 330-

340 K. Fulfillment of this condition is possible when dose rate, D<15 Gy/s or the cycling integrated dose collects at D=3000-5000 Gy while keeping the temperature at a level of 310-320 K.

Constructive features and the main functioning principles of the AChS-based dosimetry systems are the same as those of other dosimeters based on radiation-stimulated changes of optical properties (Frank et al., 1973).

Table 1. Dosimetric characteristics of AChS

AChS	Controlled parameter	Range of measured doses (Gy)	Sensitivity (S)
AAT glass (fresh-prepared)	$\Delta OD/OD>0$	5.10^5-10^7	0.30
AAT glass (gamma-irradiated and annealed)	$\Delta OD/OD>0$	5.10^5-10^7	0.25
As_2Se_3 glass	$\Delta\sigma/\sigma<0$	5.10^5-5.10^6	0.06
(fresh-prepared	$\Delta\sigma_{h\nu}/\sigma_{h\nu}<0$	3.10^5-2.10^6	0.65
AAT film (fresh-prepared)	$\Delta OD/OD>0$	5.10^3-5.10^6	0.13
AAT film (photoexposed)	$\Delta OD/OD>0$	5.10^3-3.10^5	0.10

AChS in the form of thin layers (d=1-2 μm), obtained by the well-known "traditional" technique of thermovacuum deposition (Borisova, 1983), can be used as radiation-sensitive elements in the same way as similar glass systems. However, they are characterized by irreversible changes of optical properties (Shvarts et al., 1990) and sufficiently low sensitivities (S=0.05-0.15). Therefore, thin layers previously photoexposed by absorbed light are more relevant for practical applications (see Table 1). Such dosimeters operate due to combined photo-radiation effects in AChS associated with defect recharging processes (Kornelyuk et al., 1989). It must be noted that high values of sensitivity S>0.6 are reached in AChS-based dosimeters using steady photocurrent degradation, $\Delta\sigma_{h\nu}/\sigma_{h\nu}$ (see Table 1). However, these devices are complicated in practice and have a more limited range of useful doses. When dark electroconductivity, σ, is used as the control parameter, then the S value is not greater than 0.06. This means that such elements are unfit for device application in industrial dosimetry of high-energy ionizing radiation.

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