NEUTRON IRRADIATION INDUCED CHANGES IN THE ELECTRIC CONDUCTIVITY OF TGSe CRYSTALS

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ABSTRACT

The d-c. electric conductivity (σ) was measured along b and c - axes of TGSe crystals as a function of temperature and fast neutron irradiation dose. The obtained results revealed an increase in (σ) with the neutron dose accompanied by an increase in the activation energies both in the para-and ferroelectric phases. Also results are presented of the investigation of the influence of rejuvenation process on the conductivity and transition temperature of irradiated TGSe crystals. The total number of defects due to all possible reactions that occurred inside the TGSe crystal lattice was estimated.

INTRODUCTION

Triglycine selenate (NH_2 CH_2 COO H)$_3$. H_2 SeO_4 (TGSe) belongs to the well-known triglycine sulphate (TGS) group of isomorphous compounds and it exhibits a second order ferroelectric phase transition close to the tricritical point at 22 ^{o}c[1].

Elastic, thermal and dielectric properties of crystals of this compound have been investigated thoroughly [2-5] and main laws governing the critical behaviour of this uniaxial ferroelectric are understood [6].

The influence of structure defects and impurities on the properties of ferroelectric crystals near phase transition is currently attracting much attention.

Most the work has been carried out on TGSe crystals [7-11], while much less works have been devoted to studies of radiation effects (with γ-rays) in TGSe crystals [12-14].

The influence of γ-irradiation on the dielectric properties on TGSe crystals has been studied by Lagutina et al. [12]. The studies revealed particular strong dependence of the dielectric hysteresis loop on irradiation.

Amin et al. [13] measured the d-c electric conductivity (σ) of TGSe crystals along b-axis before and after irradiation by γ-rays (0.6 Mr) revealing an increase in (σ) due to irradiation.

Strukov et al. [14] have studied the influence of γ-irradiation on elastic and dielectric properties of TGSe crystals . The results obtained demonstrated a change in the nature of the phase transition in crystals as a result of irradiation.

In an earlier work, Hamed et al. [15] have reported on the anisotropy of electric conductivity (σ) of TGSe crystals. It has been pointed that the highest value of (σ) coincides with the direction along which the lattice of TGSe is the least strongly bound.

We investigated the influence of irradiation with fast neutrons on the d-c. electric conductivity of TGSe crystals, measured along b and c-axes.

EXPERIMENTAL

Crystals of TGSe were prepared from a stoichiometric aqueous solution of aminoacetic acid (NH_2 CH_2 COO H) and selenic acid (H_2 Se_4) and then purified by three-fold recrystallization from water. Single crystals of TGSe were obtained by slow evaporation. The crystals were grown above the Curie temperature (303 K) [15]. The samples were cleaved perpendicular to the ferroelectric axis "b". The samples were prepared in the form of rectangular plates about 1 mm thick. An area of about 3 mm^2 was covered with graphite to act as electrodes. Each sample was held between copper electrodes of a holder specially designed for this purpose. Pre-calibrated Cu-constantan thermocouple was used for the temperature measurements.

The d-c electric conductivity (σ) was measured as described previously [9].

In order to remove excess polarization from the sample's

surfaces the sample was heated to a constant temperature, 323 K, while applying the required external electric field along the considered direction (E=8 K V cm^{-1}).

Measurements of the temperature dependence of the steady state conductivity were performed in the single-domain state on cooling from 323 K to 288K with cooling rate of about 0.15K min^{-1}. The uncertainty in measuring σ was less than ± 2%.

The exposures of the samples were carried out by using D-T compact neutron generator producing 14 MeV fast neutrons with flux of 6 x 10^5 n. $cm^{-2}.S^{-1}$. The samples were irradiated for different times ranging from 2 hours up to 8 hours, receiving different doses from 0.25 Gray up to 1.0 Gray.

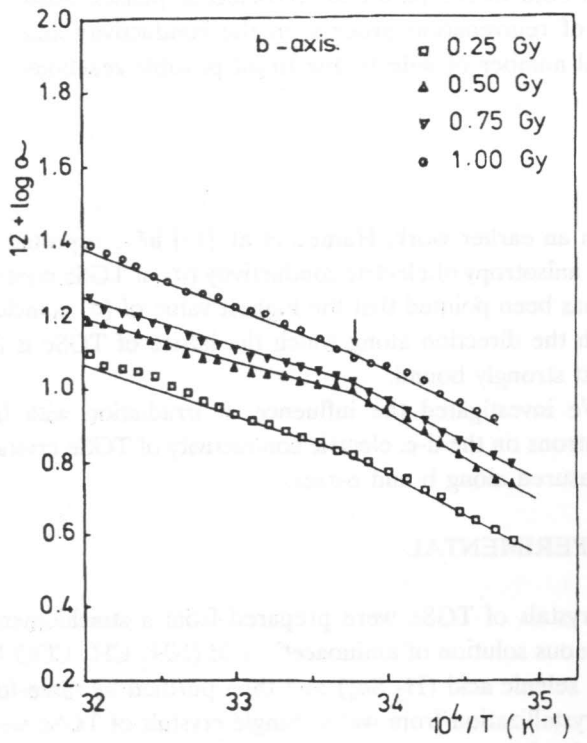

Figure 1. Reciprocal temperature dependence of electric conductivity of TGSe crystals irradiated with fast neutrons, measured along b-axis (at E = 8 kC/cm).

RESULTS AND DISCUSSION

Figure (1) displays the temperature dependence conductivity (σ) for four samples of TGSe, irradiated by fast neutrons with different doses along the ferroelectric, b-axis. The main feature of the electric conductivity (σ) curve is similar to the recent results obtained for TGSe crystals [15,16].

The changes in the activation energy (W) of the samples (or the change in the slop of the curves) occur at a temperature, which is coincident with the Curie temperature, T_c of the samples. The activation energy (W) is higher in the ferroelectric region ($T<T_c$) than that in the paraelectric region ($T>T_c$).

This is in a fair agreement with the previous reported data on ferroelectrics [17].

Also the transition temperature,t_C, is unaffected by neutron irradiation [11].

From Figure (1) it can be seen that the electric conductivity (σ) increases with irradiation dose in the para-and ferroelectric phases.

It is possible that lattice vacancies created by neutron irradiation are responsible for the observed change in the ionic type electric conductivity [18]. Also the diffuse of defects involves transport of charge and this may contribute to the conductivity (conduction mechanisms) [19].

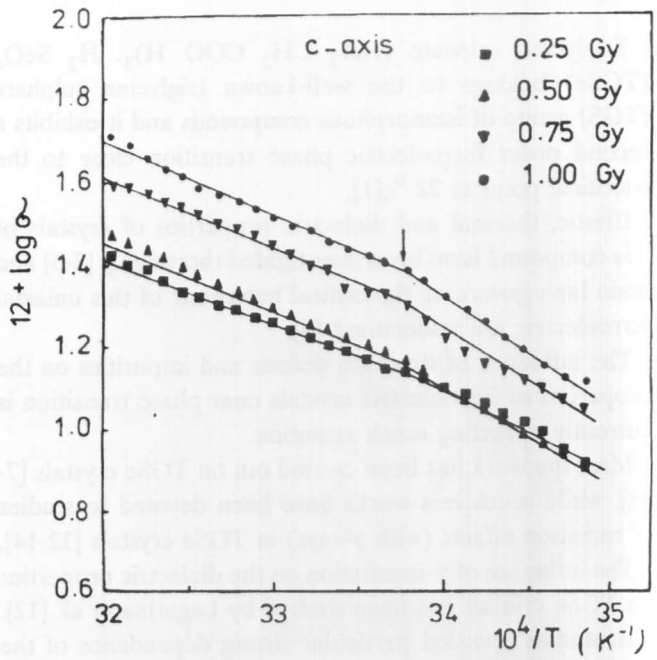

Figure 2. Reciprocal temperature dependence of electric conductivity of TGSe crystals irradiated with fast neutron, measured along c-axis (at E= 8 KV/cm).

The temperature dependence of (σ) measured along the c-axis for irradiated TGSe samples is depicted in Figure (2). The same effect is obtained as in the case of b-axis (Figure 1), however the values of σ, along c-axis are rather higher [15].

The activation energy (W) is estimated both in the para- and ferroelectric regions from the slope of the line.

Along c-axis the value of activation energy in both phases (W_p and W_F) was found to increase with the increase of the irradiation dose. While the situation along b-axis is more complicated. This complicated behaviour in the case of b-axis is mainly due to the contribution of the spontaneous electric polarization field which is effective along the b-axis (at $T<T_c$) [1].

The increases of (W) due to neutron irradiation may be attributed to the fact that the isotropically introduced radiation defects act as scatterers and the new radioactive species act as a source of electrons (β^-).

Table (1) represents the total number of defects [20,21] due to all possible reactions that occurred inside the crystal lattice of TGSe. The values of cross-sections as a function of neutron energy are well known and published in many literatures [22]. The defects arising from secondary radiation (γ, β^-, β^+,...) are neglected throughout this work because the values of cross-sections are relatively small in comparing with the predominant elastic scattering reaction.

Table (1).

Element	No. of defects
H	22.02×10^{12}
O	13.77×10^{11}
C	10.60×10^{11}
N	58.60×10^{10}
Se	11.29×10^{10}

Table (2) presents the values of (σ), W_p and W_F, for TGSe crystals., along b and c-axes as a function of fast neutron irradiation dose.

The ageing process of ferroelectrics consists in the formation and growth of spheres with ordered moments about the dipolar centre, whereas rejuvenation (annealing above, T_c) consists in a melting of these spheres [23]

Table (2).

Irradiation dose (Gray)	b-axis			c-axis		
	$\sigma \times 10^{11}$ at T=313k	Activation energy (eV)		$\sigma \times 10^{11}$ at T=313 K ($\Omega^{-1}.cm^{-1}$)	Activation energy (eV)	
		W_p	W_f		W_p	W_F
0.25	1.23	0.28	0.45	2.75	0.32	0.40
0.50	1.62	0.24	0.48	3.02	0.34	0.46
0.75	1.86	0.27	0.44	4.26	0.34	0.50
1.00	2.51	0.32	0.40	5.25	0.36	0.58

The effect of rejuvenation process on the (σ) of TGSe crystals after irradiation by fast neutrons was studied by measuring the electric conductivity (σ) for the irradiated sample of TGSe after it has been annealed at constant temperature (323 K) for different times ranging from 1, 24 to 72 hours.

The obtained results showed an interesting dependence of σ on the rejuvenation time (Figure (3)).

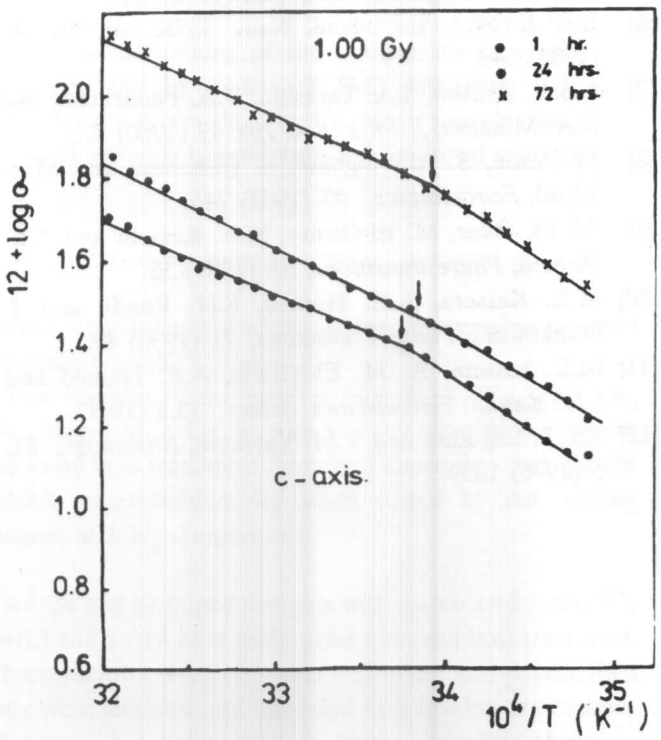

Figure 3. Reciprocal temperature dependence of electric conductivity of TGSe crystals irradiated with fast neurons by a dose of 1.0 gray, measured along c-axis (at E = 8 KV/cm), for different rejuvenation times.

It is clear from Figure (3) that σ increases with rejuvenation time (R.T) and the transition temperature, T_c, decreases from 23 to 22.6 and to 22 °C for 1, 24 and 72 hours respectively.

The changes observed with the rejuvenation process seem to be a phenomenon related to diffusion of space charges that occurred during the rejuvenation process.

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