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γ-Radiation Dosimetry Using Screen Printed Nickel oxide Thick Films

K. Arshak, O. Korostynska, and J. Harris

Abstract - Thick films of Nickel oxide (NiO) were investigated for γ-radiation dosimetry purposes. Samples were fabricated using the thick film screen printing technique. Absorption spectra for NiO films were recorded and the values of the optical band gap for as-printed, irradiated and annealed films were calculated. It was found that the optical band gap value decreased as the radiation dose was increased. Samples with an Ag-NiO-Ag sandwich structure were exposed to a ^{60}Co γ-radiation source at a dose rate of 6 Gy/min. The relative change in current increased linearly with increased dosage up to 720 Gy. The I-V characteristics indicated a Poole-Frenkel conduction mechanism. It was found that annealing restored both the electrical and the optical properties of the samples.

I. INTRODUCTION

In recent years considerable interest has been shown in the study of various properties of films due to their importance in the manufacturing of semiconductor devices and integrated circuits in the electronic industry and related areas. Electrochromic materials, for example Nickel oxide, are used in applications such as energy efficient "smart windows", glare-free and variable reflectance mirrors, high-contrast non-emissive information displays, switchable displays, devices for thermal control, semiconductor-based sensors, etc. [1-4]. The aim of this paper is to document the use of NiO thick films in radiation dosimetry. The influence of ionizing radiation on Nickel oxide and its mixture with other oxides prepared by various techniques has been explored [5, 6]. Nickel oxide is a typical binary transition metal oxide with a rock salt structure and antiferromagnetic properties below a temperature of 523 K. Due to its low cost, thick film technology (using the screen printing technique) was chosen for the fabrication of resistive Ag-NiO-Ag thick film samples of a sandwich structure. The electrical and other properties of these films depend on the manner of their preparation.

It is believed that ionizing radiation causes structural defects (called colour centres or oxygen vacancies in oxides) leading to a density increase on exposure to γ-rays [7, 8]. Values of radiation damage were estimated from changes in both current-voltage characteristics and optical density. The threshold radiation damage dose depends on the thickness and structure of the device (sandwich, planar etc.). It was found that annealing for 12 hours at 388 K restored both the electrical and the optical properties of these samples.

II. EXPERIMENTAL PROCEDURE

A polymer paste was made of 92% NiO and 8% $C_8H_{18}O_3$ by weight with Diethylenglycolmonobutylether as a solvent. NiO polymer paste and commercial DuPont 4929 silver paste were used to fabricate the radiation sensitive material and contacts respectively. Pastes were printed on glass substrates using a DEK RS 1202 automatic screen printer to form a sandwich Metal-Semiconductor-Metal structure with an active area of 1cm^2 and a film thickness of 70μm. A ^{60}Co radiation source with a dose rate of 6 Gy/min was used for exposing the samples to γ-radiation. After each fixed exposure time, the current-voltage characteristics were recorded. A linear change in the current was observed as the total dose was increased from 0 to 720 Gy in steps of 180 Gy. The samples were damaged on further exposure. The exposed samples were annealed in a Thelco Model 6 laboratory oven for 12 hours at 388 K to restore their initial electrical properties. A higher annealing temperature was not recommended as a diffusion of material could occur. Irradiation of the samples and subsequent annealing was continuously repeated to ensure repeatability of the results. Thick films of NiO polymer paste having a thickness of 10 μm were chosen for the absorption spectra studies, as thicker layers of this paste were too absorbent. The absorption spectra for as-printed, irradiated and annealed NiO films of 10 μm in thickness were recorded using a CARY 1E UV-Visible Spectrophotometer. A reference sample was used to eliminate the polymer influence in the NiO paste when recording the absorption spectra.

III. RESULTS AND DISCUSSION

Current-voltage characteristics were measured after each exposure dose, which was increased in steps of 180 Gy. Fig.1 shows plots of the I-V characteristics that were recorded for as-printed and γ-irradiated Ag-NiO-Ag thick film sandwich type structures. Fig.2 shows a linear response of the normalized current $(I-I_0)/I_o$ with radiation dose up to 720 Gy under an applied voltage of 3V.

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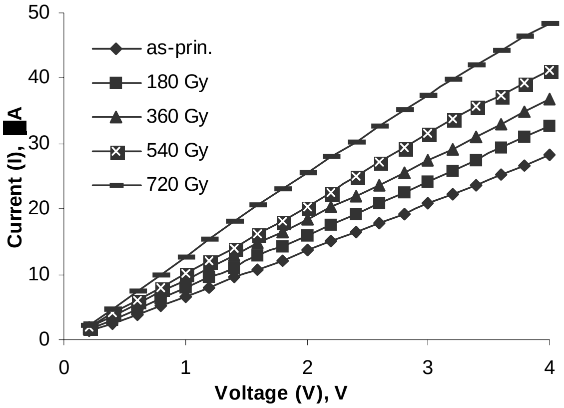

Fig. 1. Plots of current-voltage characteristics that were recorded for as-printed and irradiated Ag-NiO-Ag thick film sandwich type structures.

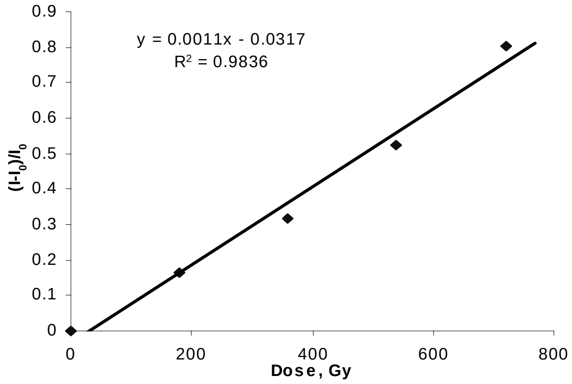

Fig.2. Dependence of normalized current $(I-I_0)/I_0$ with radiation dose under an applied voltage of 3V.

Fig.3 shows the plots of log (I) versus $V^{1/2}$ for as-printed and irradiated NiO thick films. This relation is indicative of either Schottky emission or the Poole-Frenkel effect. Both phenomena are described by the equation:

$$I_c \propto \exp\left[\frac{\beta E^{1/2}}{kT}\right] \tag{1}$$

where I_c is the circulating current, E is the electric field gradient ($E=V_b/d$, V_b being the applied voltage and d is the film thickness), k is Boltzmann's constant, T the absolute temperature in Kelvin and β is the field lowering coefficient given by Eq.2:

$$\beta = \left(\frac{e^3}{n\pi\varepsilon_0\varepsilon_r}\right)^{1/2} \tag{2}$$

where e is the electronic charge, ε_0 is the permittivity of free space and ε_r is the relative permittivity of the dielectric. The difference between these two effects is expressed by $n = 1$ for the Poole-Frenkel effect and $n = 4$ for Schottky emission. In the Schottky emission process, the electrons are emitted from the metal electrode into the conduction band of the insulator over the potential barriers between the electrons at Fermi levels of the electrode and of the conduction band of the insulator [9]. Poole-Frenkel conduction occurs as a field emission of electrons from localized donors located at an energy level below the insulator conduction band.

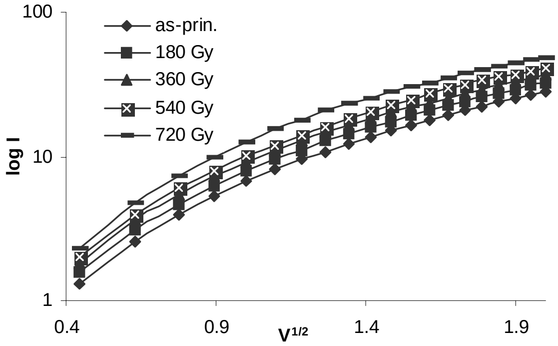

Fig.3. Plots of log (I) vs. $V^{1/2}$ for as-printed and irradiated NiO thick films.

To distinguish the predominant conduction mechanism, theoretical values of β coefficient had to be calculated and consequently, it was necessary to measure the capacitance. Related measurements were performed using an impedance analyzer (HP 4277A LCZ-meter) at frequency of 1 kHz. Theoretical permittivity was calculated using the Eq.3:

$$\varepsilon_r = Cd/\varepsilon_0 A \tag{3}$$

where C is the measured capacitance and A is the effective area. For comparison, the measured and calculated permittivity values of as-printed and γ-irradiated samples are given in Table I.

TABLE I
COMPARISON OF THE MEASURED AND CALCULATED PERMITTIVITY VALUES TO REVEAL THE CONDUCTION MECHANISM

Radiation dose, Gy	Capacitance C, x 10^{-11} F	Permittivity ε_r		
		Measured by LCZ-meter	Poole-Frenkel effect	Schottky emission
0	14.0	9.49	9.5	23.9
120	11.8	8.00	7.4	18.6
360	9.7	6.57	6.1	15.4
540	8.0	5.42	4.6	11.6
720	6.2	4.20	3.9	9.9

The experimental values of ε_r for the samples under investigation lay close to the calculated values for $n = 1$. One may therefore regard the high-field conduction mechanism as being predominantly of the Poole-Frenkel type. The decrease in the measured dielectric constant values with an increase in radiation dose is shown in Fig.4.

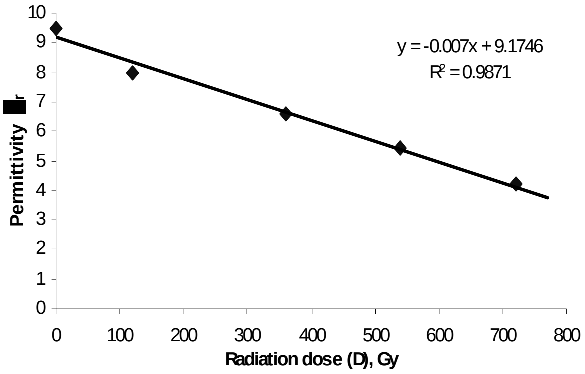

Fig.4. Linear decrease in the measured permittivity values ε_r with the increase in radiation dose.

The values of ε_r for as-printed samples were found to be 9.5. In contrast, the dielectric constant for Nickel oxide has been found to decrease with frequency and to reach a constant value of 11.9 at a frequency of 10^5 Hz and at a temperature of 298 K [10]. This discrepancy may be attributed to the different film fabrication techniques used.

The NiO band structure and electronic properties were the subjects of numerous theoretical and experimental studies [11, 12]. The optical properties of these films are important, as they provide information on the electronic band structures, localized states and types of optical transitions. Fig.5 shows the linear increase that was measured for the optical density (OD) of samples, with an increase in the dose at constant wavelength of $\lambda = 860$ nm.

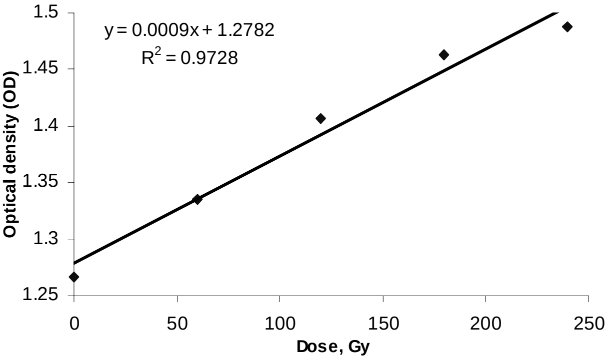

Fig.5. Increase in the optical density (OD) with increasing exposure dose at a wavelength of $\lambda = 860$ nm, for 10 μm NiO thick films.

The values of the optical band gap for as-printed and γ-irradiated NiO polymer films were estimated using the Mott and Davis' model [13] for the direct allowed transition using Eq.4:

$$\alpha(\nu)h\nu = B\left(h\nu - E_{opt}\right)^{1/2} \tag{4}$$

where α is the absorption coefficient, E_{opt} is the optical energy band gap, $h\nu$ is the energy of the incident photons and B is a constant.

The plots of $(\alpha h\nu)^2$ versus photon energy $h\nu$ for as-printed and irradiated NiO thick films are presented in Fig.6. In this study, the calculated optical band gap value for as-printed films was found to be 2.05 eV in contrast to the values found by others, depending on the various film deposition techniques used [14, 15].

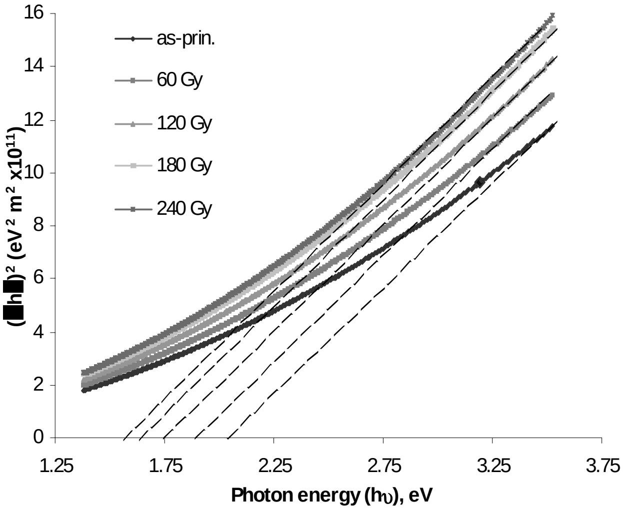

Fig.6. The plots of $(\alpha h\nu)^2$ vs. photon energy $h\nu$ for as-printed and irradiated 10 μm NiO thick films.

It has been reported that the optical band gap values E_{opt} for many compound semiconductor materials change under the influence of radiation exposure [16, 17]. Fig.7 shows a decrease in the value of the optical energy gap E_{opt} with an increase in the dose applied to the explored material.

Fig.8 shows the increase in the normalized energy of the localized states $({}_{\Delta}E-{}_{\Delta}E_0)/{}_{\Delta}E_0$ with the increase in radiation dose. The variation of the optical energy gap with electron irradiation energies and doses can be explained as the change in the degree of disorder. From the density-of-state model, it is known that E_{opt} decreases with an increasing degree of disorder of the amorphous phase [18].

Annealing of the irradiated samples for 12 hours at 388 K was found to restore both the electrical and optical properties of the samples.

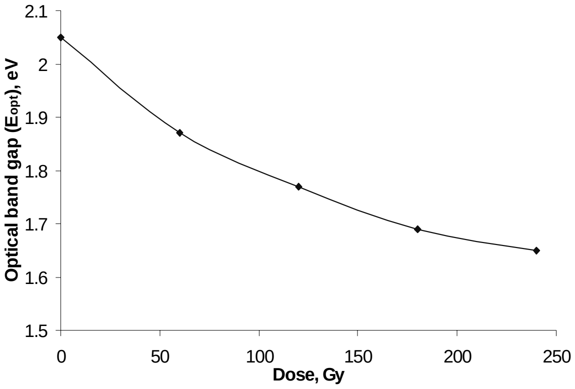

Fig.7. Change in the optical band gap E_{opt} with dose.

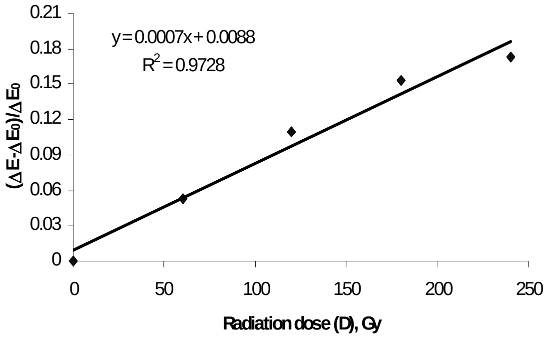

Fig.8. Increase in the normalized energy of the localized states $(_{\Delta}E-_{\Delta}E_0)/_{\Delta}E_0$ with the increase in radiation dose.

IV. CONCLUSION

The possibility of using screen-printed Nickel oxide (NiO) thick films as a sensitive material for γ-radiation was explored. Samples were exposed to a ^{60}Co γ-radiation source at a dose rate of 6 Gy/min. The characteristics were recorded after a fixed exposure time. The Poole-Frenkel conduction mechanism was dominant for these samples. Absorption spectra for 10 μm NiO thick films were recorded and the values of the optical band gap were calculated. It was found that the optical band gap was highly affected on exposure to γ-radiation and decreased with an increase in the dose. It was found that annealing restored both the electrical and the optical properties of the samples.

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