

THE EFFECTS OF γ -IRRADIATION ON THE OPTICAL AND ELECTRICAL PROPERTIES OF BUFFER SOLUTION GROWN CdS THIN FILMS

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(Received 27 April 2000; Revision Accepted 14 Sept. 2000)

ABSTRACT

Pure and aluminium doped (0.001 wt %Al) CdS thin films were deposited on microscope glass slides using buffer solution growth technique based on CdSO_4 as the cadmium source, thiourea as the sulphur source, and $(\text{NH}_4)_2\text{SO}_4$ as a buffer. The effects of gamma radiation on the optical and electrical properties of the CdS films have been investigated by measuring their optical absorbance and electrical conductivities before and after irradiation by different doses of γ - radiation in the range 7.27×10^2 to 2.04×10^5 Gy.

The absorbance was found to be dependent on film's thickness (and aluminium dopant concentration) and γ - radiation doses. CdS films (pure and doped) γ - irradiated at high doses (2.6×10^6 Gy) have the lowest absorbance. Gamma - irradiation induced dark electrical conductivity in CdS films, which was dependent on absorbed γ - radiation dose rate. The conductivity was permanent at high γ - radiation dose (2.04×10^5 Gy) and transitory below this dose.

Generally, the optical band gap values of unirradiated and γ - irradiated CdS : Al films were lower than that of pure CdS films. The band gaps and electrical conductivities of pure and CdS : Al films, unirradiated and irradiated at low γ - radiation doses ($\leq 1.45 \times 10^4$ Gy) are the same i.e. 2.43 ± 0.02 eV and 0.068 ± 0.005 ($\Omega\text{-cm}$)⁻¹, and 2.31 ± 0.02 eV and 0.69 ± 0.05 ($\Omega\text{-cm}$)⁻¹ for pure and CdS : Al films respectively. At high γ - radiation doses, the band gaps decreased to 2.30 eV and below 2.30 eV for pure and CdS : Al films respectively, accompanied by increase in the dark electrical conductivities.

Possible mechanisms responsible for the observed changes are briefly discussed.

Keywords : Cadmium sulphide films, gamma - irradiation, Solar cells properties.

INTRODUCTION

Polycrystalline CdS thin films have a relatively wide energy band gap ($E_g = 2.42$ eV) and possesses n-type semiconducting characteristics. These properties make it a very desirable window layer, for photon absorption, for many heterojunction thin film solar cells such as CdS - CdTe, CdS - Cu_2S and CdS - CuInSe_2 . (Chu and Chu, 1995).

Low - cost techniques used to prepare CdS thin films include solution growth (Chu and Chu, 1995, and Dhare et al, 1995), spray, pyrolysis (Chavez et al, 1997) and vacuum deposition (Chu and Chu, 1995). Solution growth technique is a good method for obtaining very thin CdS film as a window layer for a thin film solar cell because it is inherently a low temperature process which does not damage the surface of the thin film as an absorber during deposition. Each technique produces films with different properties which should be optimized for the particular applications of the films.

A major problem associated with the use of some solid state devices in a radiation environment is the degradation due to radiation damage. Solar cells used in a space environment for terrestrial photovoltaic conversion of solar energy (e.g. CdS - CdTe solar cells) are continually exposed to bom-

bardment from space electromagnetic radiations, electrons, protons, e.t.c, which could cause damage in them and drastically reduce their useful lifetimes (Yamaguchi and Amano, 1983). For example, on the ATS - 6 satellite, n⁺ / p silicon solar cells, protected by cover glass and UV blocking filter, lost 13 - 17% of their original maximum power output after 2 years and four months in synchronous orbit (Goldhammer, 1979).

CdS thin films used as window materials in thin film solar cells for space applications would also be exposed to bombardment from space radiations, which could cause radiation damage or property modifications, leading to some changes in the performance of CdS as a window material. It is therefore very important to investigate and understand the physical aspects and fundamental limitations of CdS window materials for solar cell applications under various radiation conditions. Little or no attention has been paid to the study of radiation induced changes in polycrystalline CdS thin films (Singru, 1974). Hence the aim of the work reported in this paper is to study the effects of γ - radiation on the optical and electrical properties of buffer solution grown pure and aluminium doped CdS thin films on glass substrates.

EXPERIMENTAL PROCEDURE

CdS film deposition.

A schematic diagram of the experimental set

up for depositing CdS films using buffer solution growth technique is shown in Figure 1. It consists of a glass water bath, inside of which is centrally placed a deposition solution beaker at the bottom of which we have the magnetic "flea" stirrer. The temperature of the water bath and hence that of deposition solution were controlled automatically by the magnetic stirrer / hot plate unit (CLIFTON) which incorporates a thermostat. In this closed deposition system, a plastic cover on top of the deposition solution beaker was used to prevent evaporation of the solution. Four small holes were drilled in the plastic cover to accommodate (and hold tightly) two glass substrate holders made of teflon (one substrate per holder), a thermometer and the fourth hole ($\sim 4\text{mm}$) allows drops of thiourea, $(\text{NH}_2)_2\text{CS}$ to be added slowly using pipette to the deposition solution for the formation of CdS film. The glass substrates, fixed to the teflon substrate holders, and the thermometer were placed vertically in the deposition solution. The deposition solution was moderately stirred magnetically to enable Cd^{2+} and S^{2-} diffuse to the substrates for the heterogeneous growth of CdS film on the substrates surface.

Prior to the deposition of the CdS films, two microscope glass slides ($25\text{mm} \times 25\text{mm} \times 1\text{mm}$) used as substrates were each marked on one face into $5\text{mm} \times 10\text{mm}$ unit areas using a diamond cutter. These substrates were cleaned by soaking them in a solution made up of 10g of potassium dichromate in 110ml dilute sulphuric acid ($\text{H}_2\text{O}:\text{H}_2\text{SO}_4 \approx 10:1$). The glass substrates were washed in distilled water, rinsed in acetone/methanol mixture, washed again in distilled water, etched with dilute nitric acid, HNO_3 for one minute and finally rinsed in distilled water.

Cadmium sulphate, CdSO_4 was used as the cadmium source in the investigation. The following stock solutions were prepared for CdS films deposition: 1 M CdSO_4 , 1 M ammonium sulphate, $(\text{NH}_4)_2\text{SO}_4$, 1 M $(\text{NH}_2)_2\text{CS}$, and 1 M aluminium sulphate, $\text{Al}_2(\text{SO}_4)_3$. The deposition solution was prepared to give a final composition of 0.0025M CdSO_4 , 0.009M $(\text{NH}_2)_2\text{CS}$, 0.025M $(\text{NH}_4)_2\text{SO}_4$ at a final pH of 9.5 and deposition temperature of 85°C . See table 1. The deposition solution beaker initially contained 100ml of deionized water which was heated up to the desired deposition temperature. The calculated volumes of cadmium and ammonium stock solutions (Lambert et al 1968) which when mixed with the

100ml deionized water will give the desired final compositions of the deposition solutions as indicated above and in Table 1, were added at the desired deposition temperature. The pH of the deposition solution, under continuous stirring, was adjusted to the final value of 9.5 by slowly adding NH_4OH . When the deposition temperature and the pH of the solution stabilized at the desired values of 85°C and 9.5 respectively, substrates were immersed in the solution, with the plastic cover in place. Calculated volumes of thiourea from the stock solution was slowly added to the deposition solution to start the growth of CdS films on the substrates surface. CdS films were deposited on the

two substrates for 40 minutes. To prepare doped CdS films, different weight percentages of aluminium, i.e. 0.0005 & 0.001wt % Al obtained from corresponding calculated proportion (Lambert et al 1968) from $\text{Al}_2(\text{SO}_4)_3$ stock solution were added to calculated volumes of CdSO_4 and $(\text{NH}_4)_2\text{SO}_4$ stock solutions in a small evaporating dish. This mixture was then added to the deionized water at the required deposition temperature. After the deposition of CdS film, the substrates were removed and cleaned by flushing with distilled water to remove any surface-adsorbed particles, and then dried in air. Cd film on one substrate surface was removed with a cotton swab moistened with dilute HCl, before measurements were carried out on it. Film thickness was calculated gravimetrically from the mass of the deposit using S. 2000 (BOSCH) balance, CdS molar density of 4.82 g cm^{-3} and film dimensions. Each glass substrate on which CdS film was deposited was easily cleaved into $10\text{mm} \times 5\text{mm}$ samples without damaging the thin film, before measurements were carried out on it.

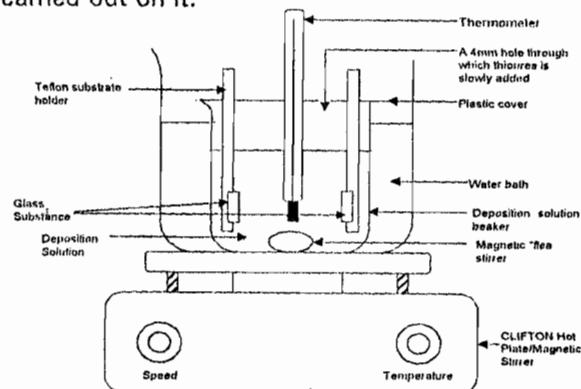


Figure 1 Experimental Set up for preparation of buffer solution grown CdS thin films.

γ - irradiation of CdS films

Pure and aluminium doped CdS thin films with thicknesses 122nm (pure), 136nm and 147nm were γ - irradiated at room temperature in air using a ^{60}Co gamma cell 220 manufactured by the Atomic Energy of Canada Ltd. A plain microscope glass slide used as a reference sample was γ - irradiated along with the CdS film deposited on glass slide (i.e. sample). The dose rate was 7.27×10^2 Gray/hr, and sample and reference sample were γ - irradiated for 1, 2, 5, 10, 20, and 281 hours, so that the integrated doses used ranged from 7.27×10^2 to 2.04×10^5 Gray. (See Table 1).

Optical characterization

The band structures of the CdS thin films were investigated by optical absorption measurements at room temperature using a CAMSPEC M201 visible spectrophotometer in the wavelength range 400-800nm. Incident photon flux was normal to the CdS film and a plain microscope glass slide (a substrate) was used as a reference sample during measurements. The absorbance of the CdS film was obtained by subtracting the absorbance of the reference sample from that for the sample. All measurements were carried

out a day after γ - irradiation. Digital data from the spectrophotometer was fed into a personal computer to obtain absorption spectra, which was used to calculate the energy band gap (E_g) using the relationship $[\alpha \text{ hv}]^2$ versus hv .

Electrical characterization

A room temperature, dark sheet resistances of the pure and doped CdS films before and after γ - irradiation were measured with a Keithley model 2000 multimeter, using two coplanar silver paste electrodes (1.0 x 10mm), 3mm apart. The electrical resistivity of each CdS film was then calculated from the thickness, and the film electrical conductivity, an inverse of the electrical resistivity, was obtained.

RESULTS AND DISCUSSION

The spectral distribution of absorbance for pure and aluminium doped CdS films before and after γ - irradiation in the range 7.27×10^2 to 2.04×10^5 Gy shows that the intensity of the absorbance is remarkably affected by both aluminium dopant concentration and γ - ray doses. Changes induced in CdS film characteristics by γ - irradiation are more pronounced in aluminium doped CdS films than in pure films, probably due to changes in band structure brought about by the incorporated aluminium dopants.

Optical absorption in unirradiated and γ - irradiated CdS films

From figures 2 and 3 (inset), the absorbance of unirradiated and γ - irradiated CdS :Al films is higher than that of unirradiated and γ - irradiated pure CdS films in the visible region. This shows that the measured absorption is dependent on CdS film thickness which was observed in this work to increase with aluminium dopant concentration in agreement with the observations of Lokhande and Pawar, 1982.

Generally for pure and aluminium - doped CdS films, the intensities of absorption change irregularly with increasing gamma ray doses. However, CdS films gamma irradiated for more than 20 hours (i.e. integrated dose of 2.04×10^5 Gy) have the lowest absorbance. Absorbance dropped by about 45% in the visible region. These observed changes in the intensities of absorption are believed to be mainly associated with gamma ray induced microstructural changes in the CdS films, because of the fact that the optical properties of thin films generally are affected by their structure (See next section below).

The absorbance in γ - irradiated pure CdS films is lower than that of unirradiated films for all the γ - radiation doses used. However, the absorbance changes irregularly with γ - ray dose.

Optical band gaps and dark electrical conductivities of CdS films

Figures 2 and 3 show that γ - irradiation has a pronounced effect on the energy gaps of the CdS films, especially at very high doses (2.04×10^5 Gy). γ irradiation induced dark electrical conductivity in CdS film which was dependent on absorbed γ - ray dose. The conductivity is believed to be due to ionization in CdS films by γ - rays.

The band gaps and electrical conductivities of pure and CdS:Al films, unirradiated and irradiated at low γ - ray doses ($\leq 1.45 \times 10^4$ Gy) are the same within the limits of experimental errors i.e. 2.43 ± 0.02 eV and 0.068 ± 0.005 ($\Omega \cdot \text{cm}$)⁻¹, and 2.31 ± 0.02 eV and 0.69 ± 0.05 ($\Omega \cdot \text{cm}$)⁻¹ for pure and CdS :Al films (0.001 wt% Al) respectively. It is pertinent to note that when a lower concentration of aluminium i.e. 0.0005 wt% Al was incorporated into CdS film, the estimated E_g values and dark electrical conductivities obtained (not included) were intermediate between that for pure CdS and CdS : Al films (0.001 wt% Al). Because of the fact that the optical properties of thin films are affected by their structure, it is believed that unirradiated CdS films (pure and doped) and those irradiated at low γ - ray doses have similar structure. Thus low γ - ray doses have not produced measurable structural changes in the CdS films. Lack of appreciable changes in the electrical conductivities of the CdS films in the low γ - ray dose range is probably because the energy imparted to the valence electrons by the ionizing radiation (γ - rays) may not be enough to raise them to the conduction band, but only to the exciton band where the electrons remain electrostatically bound to the corresponding holes in the valence band. This electron - hole pair (an exciton) produced by excitation process has no net charge and therefore do not contribute to the electrical conductivity of the CdS films.

For high γ - ray doses (2.04×10^5 Gy), the band gaps of pure CdS films decreased by about 5% to 2.30 eV, while the band gaps of CdS:Al films decreased to (2.20 - 2.27 eV), at γ - ray doses above 3.63×10^3 Gy accompanied by increase in the electrical conductivity.

In pure CdS films, induced electrical conductivity is believed to be caused by the ionization effect of the γ - rays in the CdS films. High dose γ - rays have sufficient energy to cause appreciable ionization in the CdS films, such that a large number of electrons are raised from the valence band to the conduction band (i.e. pair production). Both the electrons and "positive holes" are free to move independently through the CdS lattice and the free electrons are believed to contribute to the enhanced n-type electrical conductivity in the CdS films. The observed enhanced electrical conductivity is a manifestation of the availability of excess charge carriers (electrons)

Table 1: Summary of the effects of γ -irradiation on the optical energy gaps and electrical conductivities of pure and aluminium doped CdS films. $[CdSO_4] = 0.0025M$; $[(NH_4)_2CS] = 0.009M$; $[(NH_4)_2SO_4] = 0.025M$, pH = 9.5; T = 85°C; deposition time = 40 minutes. Pure CdS film = 122nm; CdS : Al film = 147nm.

Sample No.	Wt% Al.	Molar Concentration (M) of Al	γ - radiation dose (Gy)	Optical band gap [~] (eV)	Dark Electrical conductivity $(\Omega\text{-cm})^{-1} \times 10^{-4}$	Optical band gap (eV) (after 4 months)
1	-	-	0	2.43 ± 0.02	0.067 ± 0.005	2.43 ± 0.02
2.	-	-	7.27 x 10 ²	2.44 "	0.059 "	2.43 ± "
3.	-	-	1.82 x 10 ³	2.43 "	0.069 "	2.43 ± "
4.	-	-	3.63 x 10 ³	2.42 "	0.068 "	2.41 ± "
5.	-	-	7.27x 10 ³	2.42 "	0.071 "	2.42 ± "
6.	-	-	1.45 x 10 ⁴	2.42 "	0.073 "	2.42 ± "
7.	-	-	2.04 x 10 ⁵	2.30 "	0.162 "	2.30 ± "
1.	0.001	1.85 x 10 ⁻⁴	0	2.30 ± 0.02	0.68 ± 0.05	2.30 ± 0.02
2.	"	"	7.27 x 10 ²	2.33 "	0.66 "	2.32 "
3.	"	"	1.82 x 10 ³	2.30 "	0.70 "	2.30 "
4.	"	"	3.63 x 10 ³	2.30 "	0.72 "	2.30 "
5.	"	"	7.27x 10 ³	2.27 "	1.24 "	2.31 "
6.	"	"	1.45 x 10 ⁴	2.20 "	2.50 "	2.26 "
7.	"	"	2.04 x 10 ⁵	2.25 "	1.82 "	2.24 "

in the grains, and the lowering of potential barrier heights at the grain boundaries which enhance the electron mobility. These changes are an indication of improvement in the crystallinity of the irradiated CdS film, which is believed to have increased the film grain size, leading to the observed decrease in the optical energy gap of the pure CdS films.

In CdS : Al film, the observed decrease in band gap is believed to be due partly to the increase in donor levels in CdS films brought about by the incorporated aluminium, a shallow donor in CdS. A large concentration of aluminium (0.001wt%Al) in the CdS lattice makes the donor levels degenerate and merge into the conduction band of CdS thereby causing the conduction band to extend into the gap (i.e. tailing) which reduces the band gap as observed in this work. The decrease in the band gap may be due to increase in the grain size of the film during ionization process as suggested above for pure CdS film. The observed increase in the n - type electrical conductivity of CdS:Al film is due to electrons contributed by the incorporated aluminium which acts as a donor in CdS film, and those made available through ionization process in the CdS films by the γ - rays. Incorporated aluminium, a trivalent cation substitutes for a cadmium ion to form a centre, consisting of the trivalent cation, having an electron loosely bound in an orbit about the centre, with ionization energy of $\sim 0.04eV$.



Most of the centres are ionized at room temperature to release electrons that will contribute to the conductivities of CdS films. Therefore, as the concentrations of incorporated aluminium increases, the number of such centres with loosely - bound electrons, that could contribute to the dark current also

increases. The higher dark electrical conductivities of CdS: Al films compare to that of pure CdS films were due to a larger concentration of electrons from the shallow donor (Al) in CdS films. The increase in the electrical conductivities of CdS:Al films with film thickness (or dopant concentrations) (Table 1) due to increase in the charge carrier (electron) density in the films is a manifestation of the presence of excess charge carriers (electrons) in the film grains, and the lowering of potential barrier heights at the grain boundaries which enhance the electron mobility. These changes are an indication of improvement in the crystallinity of CdS films, which may have brought about increase in the films grain size. This in turn can contribute to the observed decrease in band gaps.

Measurements of bandgaps and electrical conductivities of some CdS film samples were repeated after four months. The band gaps and electrical conductivities of the CdS films γ - irradiated at a high dose of 2.04×10^5 Gy were found to be permanent

most probably due to γ - radiation induced permanent changes in the film lattice structures. Below this γ - radiation dose, the bandgaps and electrical conductivities of the CdS films were found to be transitory because E_g and electrical conductivities increased and decreased towards pre-irradiation val-

ues. In a good transparent thin film electrode like CdS thin film, suitable as a window material for solar cell application, the optical transmission and the electrical conduction of the electrode should be as large as possible. However, the interrelationship between optical transmission and the electrical conduction excludes in most cases, the simultaneous achievement of maximum transmission and conduction (Haacke, 1976). Reasonable electrical conductivity values and

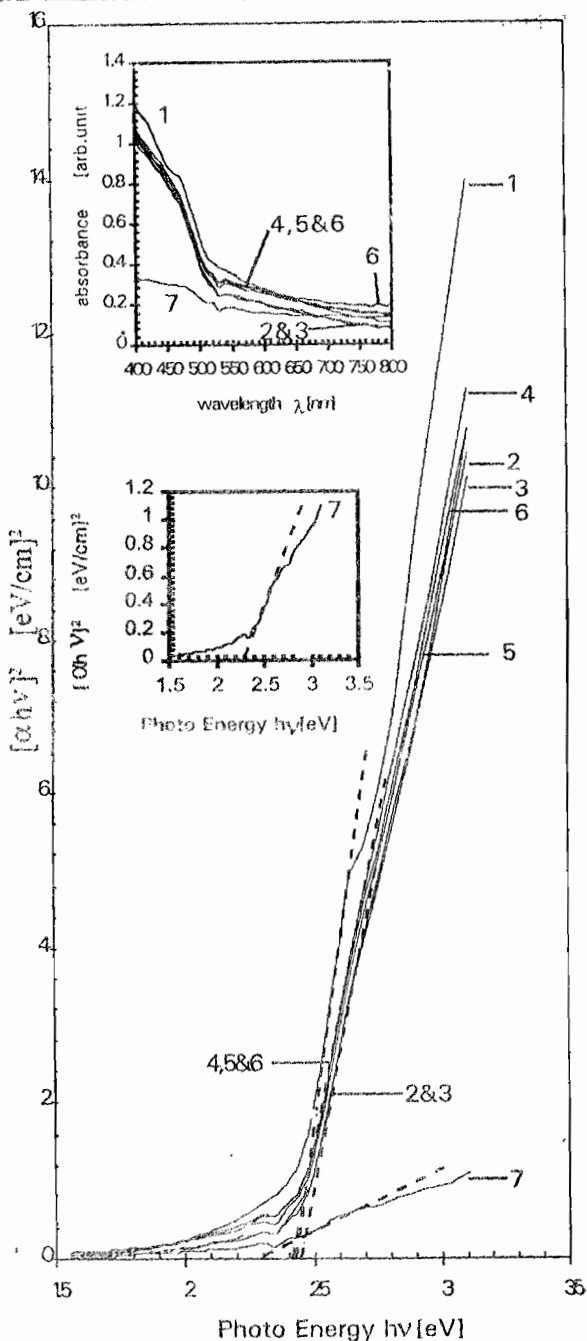


Figure 2: $(\alpha h\nu)^2$ versus $h\nu$ plot of buffer solution grown CdS films for different γ -radiation doses, α is the absorption coefficient and $h\nu$ is the photon energy (eV). (Fig.2 [insets] are the optical absorption spectra of unirradiated and γ -irradiated CdS films, and $(\alpha h\nu)^2$ versus $h\nu$ plot for CdS film gamma irradiated at 2.04×10^5 Gy. $[CdSO_4] = 0.0025M$; $[(NH_4)_2CS] = 0.009M$; $[(NH_4)_2SO_4] = 0.025M$; $pH = 9.5$, deposition time (t) = 40minutes.
 (1) 0 Gy; (2) 7.27×10^2 Gy; (3) 1.82×10^3 Gy; (4) 3.63×10^3 Gy; (5) 7.27×10^4 Gy; (6) 1.45×10^4 Gy; (7) 2.04×10^5 Gy.

high transmittance (i.e low absorption) obtained in this work at high γ -ray doses (2.04×10^5 Gy) will make the CdS films, especially CdS:Al films, useful window materials.

It is pertinent to point out that usually observed change in colour of the microscope glass sub-

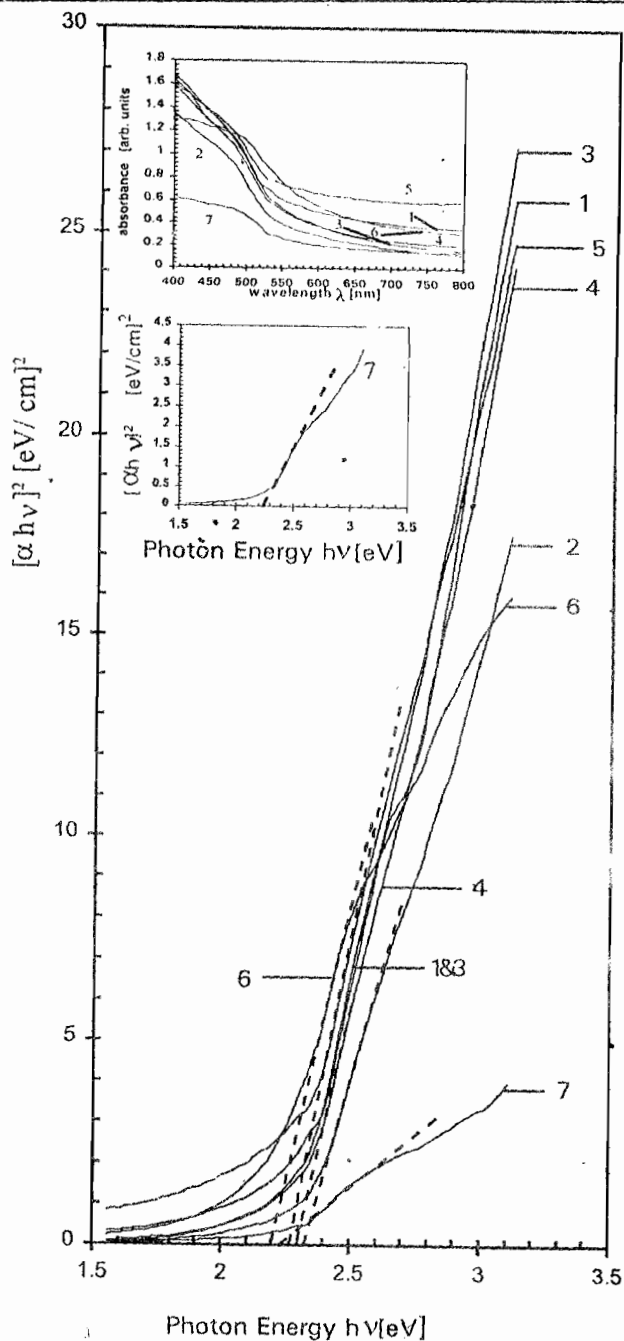


Figure 3: $(\alpha h\nu)^2$ versus $h\nu$ plot of buffer solution grown, CdS:Al films (0.001 wt % Al) for different γ -radiation doses, α is the absorption coefficient and $h\nu$ is the photon energy (eV).
 Fig 3.[insets] are the optical absorption spectra of unirradiated and γ -irradiated CdS:Al films, and $(\alpha h\nu)^2$ versus $h\nu$ plot for CdS:Al film gamma irradiated at 2.04×10^5 Gy. $[CdSO_4] = 0.0025M$; $[(NH_4)_2CS] = 0.009M$; $[(NH_4)_2SO_4] = 0.025M$; $pH = 9.5$; deposition time (t) = 40 minutes
 (1) 0 Gy, (2) 7.27×10^2 Gy, (3) 1.82×10^3 Gy; (4) 3.63×10^3 Gy; (5) 7.27×10^4 Gy, (6) 1.45×10^4 Gy; (7) 2.04×10^5 Gy.

strate occurred upon γ -irradiation to especially high absorbed doses, in line with the observations of other investigators (Attix et al 1966, chap. 13). The glass substrates which were colourless (clear and transparent) turned brown especially at high γ -ray doses. The optical absorption of the glass substrates in-

creased with γ - ray dose. This was, however, taken care of during optical absorbance measurements by subtracting the absorbance of the reference sample from that for the sample.

Post - irradiation fading of glass substrate colour was negligible for high γ - ray dose of 2.04×10^5 Gy, but below this dose, the magnitude of fading increases as the γ - ray dose decreases over a period of four months

CONCLUSION

The results obtained in this work show that;

- (i) The absorbance, optical energy gap and measured dark electrical conductivity of pure and aluminium doped CdS thin films are dependent on aluminium dopant concentration (or CdS film thickness), and γ - ray doses, in the visible wavelength region (400 - 700nm).
- (ii) Changes in energy gap, electrical conductivity, and absorbance induced by γ - irradiation are more pronounced in CdS:Al film than in pure CdS films probably due to possible changes in band structure and film grain size brought about by the incorporated aluminium.
- (iii) High dose γ -irradiation (2.04×10^5 Gy) of pure and aluminium doped CdS films induced electrical conductivity in them which was permanent, and transitory below that dose.
- (iv) Reasonable bandgap value of ~ 2.30 eV, enhanced dark electrical conductivity, and low absorption (i.e. high transmittance) in CdS:Al thin films irradiated at high γ -ray doses (2.04×10^5 Gy) suggest that the CdS films have a comparatively good transparent conducting electrode properties which will make it useful as a good window material for solar cell applications even in a nuclear radiation environment where γ - ray doses lie within the dose range studied in this work.
- (v) Irradiation of pure and aluminium doped CdS thin films at low γ - ray doses ($< 2.04 \times 10^5$ Gy) does not alter the structure or the optical and electrical characteristics of the films.

ACKNOWLEDGEMENT

The author is very grateful to, Mr. S. Adebayo and Dr. A. Adelusi of Botany Department of Obafemi Awolowo University, Ile-ife, Nigeria for the use of a Camspec M201 Spectrophotometer. The author would like to express thanks to Mr. C. Ajele of the Center for Energy Research and Development, Obafemi Awolowo University, Ile-ife, Nigeria for technical support in the γ - irradiation of the films.

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