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Influence of Cold Plasma Exposure Time on Structural and Optical Properties of Chemically Deposited CdSe Thin Films

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The influence of cold plasma at various intervals on the optical and structural characteristics of cadmium selenide (CdSe) films, produced via chemical bath deposition, was evaluated using X-ray diffraction (XRD) and UV-visible spectroscopy methods. The observed decrease in optical energy gap along with the increase in Urbach energy and extinction coefficient suggests that cold plasma irradiation introduced additional localized states and structural defects within the CdSe films. These defect states broaden the Urbach tail and enhance photon absorption, leading to reduced optical transmittance. The results indicate that plasma treatment significantly modifies the electronic and crystalline structure of the films. The reduction in crystal stress after cold plasma irradiation can be attributed to partial relaxation of the crystal structure and improved atomic rearrangement, which contributes to grain growth and reduces internal distortions within the CdSe films. These results indicate that cold plasma significantly affects the structural and crystalline properties of the films.

Keywords: Cadmium selenide; energy gap; Urbach energy; extinction coefficient; cold plasma
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1. Introduction

One of the most significant substances that has caught researchers' interest is semiconductors, since the majority of them have a direct band gap and possess two crystal structures: either hexagonal (of the wurtzite type) or cubic (of the zinc lattice type). Group II–VI of the periodic table contains the semiconductor chemical cadmium selenide. Due to its broad and direct bandgap energy of 1.74 eV, CdSe is widely used in the optoelectronics industry [1,2], which allows its use in solar cell applications, photodetectors, transistors, lasers, gas sensors, gamma ray detectors, chemical photovoltaic (PEC) cells, and optoelectronic devices [3-5]. The chemical bath deposition (CBD) is the simplest technique, often referred to as solution growth, which offers a low precipitation temperature of less than 90°C and a wide range of possibilities for the production of huge regions of films. The thickness of the film can be controlled. This technique is based on the slow release of metal ions [6]. Another advantage of the CBD method is that films can be deposited on any substrate [7]. The optical characteristics of CdSe thin films can be modified through thermal processing or by altering deposition parameters, including the temperature of the chemical processes [8], deposition time, pH, and the complexing agent effect [9]. Optimizing the deposition conditions resulted in the preparation of nanoparticles of varied size and shape with different electrical, optical, structural and properties [10]. The thin film's thickness varies from one nanometer to several micrometers, and its structure has a significant impact on its optical, mechanical, thermal, and electrical characteristics [11]. There are several techniques for depositing thin films, such as metal organic chemical vapor deposition and vacuum evaporation [12], thermal spray deposition [13], metal chemical vapor deposition [14], chemical bath deposition [15,16], plasma-enhanced chemical vapor deposition [17], atomic layer deposition (ALD) [18], rf sputtering [19], and electrodeposition (ED) [20]. The chemical bath deposition has a number of benefits, such as the absence of the need for deposition on a conductive substrate, low processing cost, the technique works at low temperatures, and the technique supports deposition on large surfaces with improved homogeneity [21]. Cold plasma is classified into two categories: low-pressure plasma and atmospheric plasma. Plasma is an ionized gas composed of electrons and ions. It is electrically conductive and affected by magnetic and electric fields. Cold (non-thermal) plasma is characterized by electrons possessing high energy while the ions remain close to room temperature, allowing its use in tissue treatment without burning and in medical sterilization [22,23]. In the experiment, the CdSe thin films are treated with atmospheric plasma. Cold plasma modifies the structural and optical properties of CdSe films through the interaction of high-energy ions and reactive oxygen and nitrogen species with the film surface. These interactions can distort the crystal lattice, generate defects, promote grain growth, and create localized electronic states, leading to changes in crystallinity, optical absorption, and energy gap. This research contributes to clarifying the mechanism for modifying CdSe properties using cold plasma as a low-temperature and environmentally friendly processing method. It demonstrates the possibility of controlling film properties solely through irradiation time, without the need for complex chemical additives or high temperatures. Cold plasma treatment generates reactive oxygen and nitrogen species capable of modifying the surface morphology, defect states, and crystallographic structure of CdSe thin films without causing significant thermal damage.

2. Experimental Part

CdSe films were prepared by CBD method on glass substrates with a concentration of 0.5 M solution at a temperature of 50°C and pH of 9, and a precipitation time of 3 hours. The solution used is a mixture of sodium selenosulfate (Na_2SeSO_3), by mixing selenium with sodium sulfite; and cadmium chloride ($CdCl_2$); and adding ammonia (NH_3) and drops of triethanolamine (TEA) as a catalyst. Glass substrates were cleaned with ethanol and acetone. The prepared samples were then exposed to cold plasma radiation for varied times of 30, 60, and 120 minutes at 0.63 mA of current and 9 kV of voltage and at a frequency of 100 kHz and power 5.67 W. The elements resulting from the reaction are emitted into the air after the plasma is formed under atmospheric pressure and are chemically active, as the electrons ionize and excite the air molecules. The production of a number of nitrogen and oxygen lines is one of the most effective and influential factors on cells and organic materials in general. The intensity of emission of these elements across optical spectra has been found to depend on the density of excited species, the applied voltage, the emission characteristics (light, frequency, spontaneous emission probability, and electrical discharge), and the spectral response of the detection system. Therefore, emission intensity serves as a measure of species concentration for qualitative analysis. Furthermore, operating conditions with a higher voltage result in a greater abundance of reaction products [24].

The crystal strain was calculated using the following equation, $\varepsilon = \beta/4\tan\theta$, where β represents the full-width at half maximum (FWHM) of the diffraction peak

The optical and structural properties of the films were investigated using X-ray diffraction and UV-visible spectrophotometry. The equation $A = \log_{10}\left(\frac{1}{T}\right)$ was used to determine the absorption spectrum from the transmission spectrum that resulted from measuring the transmittance (T) values as a function of the wavelength within the range 340-1000 nm. In addition, the grain size was calculated using the Debye-Scherrer equation $D = K\lambda/\beta\cos\theta$, where K is a constant (0.9 for CdSe films) [25,26].

The optical energy gap was found by determining the absorption coefficient as a function of the photon energy using the Tauc's relationship:

$$(\alpha h\nu)^n = A(h\nu - E_g) \quad (1)$$

where A is a constant whose value depends on the nature of the material, n is an integer whose value depends on the type of electron transition (n=2 for allowed direct transition). Calculating the direct energy gap requires plotting the relationship between $(\alpha h\nu)^2$ as a function of the photon energy (hν), where the extrapolation of the linear part of the curve to intersect the x-axis represents the direct gap energy. The absorption coefficient is calculated using the following equation $\alpha=2.303A/t$, where A represents the absorbance and t represents the film thickness. Plotting the relationship between (Lnα) and (hν) yields the inverse slope, which can be used to determine Urbach energy as

$$E_U = \left[\frac{d(ln\alpha)}{d(h\nu)}\right]^{-1} \quad (2)$$

The extinction coefficient (k) can be calculated from the following equation $k = \frac{\alpha\lambda}{4\pi}$, where λ is the wavelength of the incident rays

3. Results and Discussion

The homogeneous, orange-red, good adhesion, smooth, and reflective CdSe thin films that have been deposited. These CdSe films have either a hexagonal or cubic shape, according to earlier studies [3,27]. The X-ray diffraction (XRD) patterns and distribution were determined using JCPDS data. The structural properties of pure CdSe films and exposed to plasma radiation for different times (30, 60, and 120 min) were studied.

The peaks at (111), (220), and (311) planes are corresponding to diffraction angles of 25.59°, 42.50°, and 50.14°, respectively, in both pure and cold plasma-exposed films. The outcomes were consistent with the standard JCPDS card 19-0191, indicating that the film is polycrystalline, has a cubic structure, and has a clear peak with a high relative intensity at the (111) level [28, 29]. Figure (1) shows the XRD patterns of the samples with a shift of the XRD peaks to the left after cold plasma irradiation. Such shift indicates an increase in the interfacial distances between crystal planes as a result of the expansion of the crystal lattice, partial relaxation of internal stresses occurs after plasma irradiation, and the formation of structural defects due to the effect of active oxygen and nitrogen species.

Figure (2) shows that the energy gap decreases (1.74, 1.7, 1.65, and 1,6 eV) when exposed to cold plasma at different durations (pure, 30, 60, and 120 min, respectively) [26,30]. The reason for the decrease in the energy gap after cold plasma irradiation is due to the relaxation of crystal stress and grain growth with the generation of local defective states within the gap, which causes enhanced light absorption and an apparent decrease in the value of the energy gap [31].

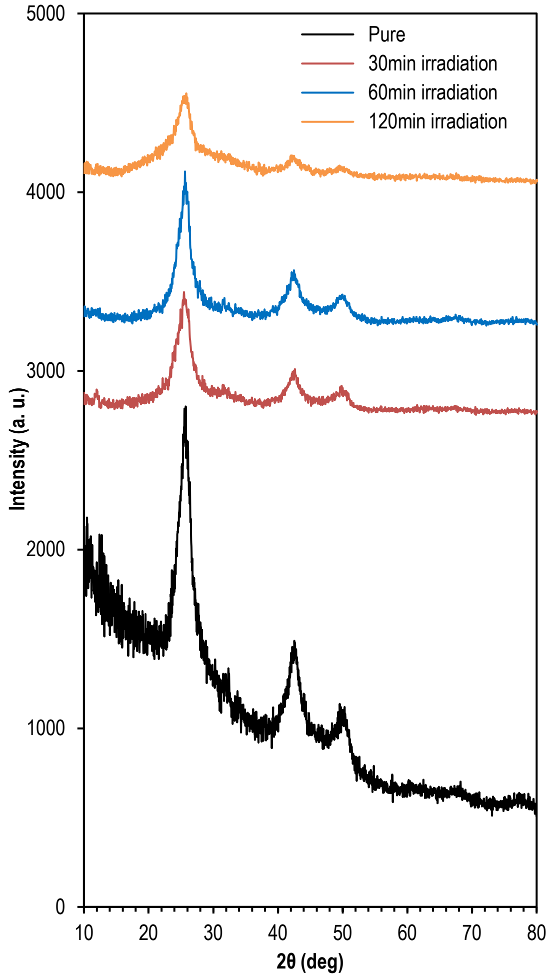
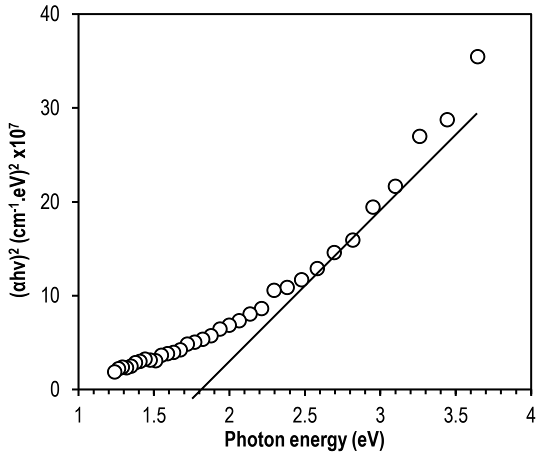
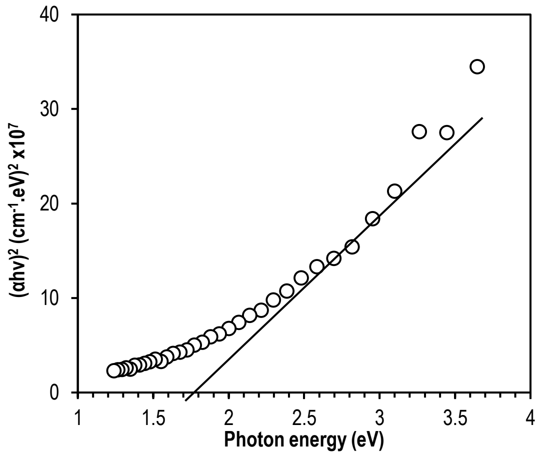


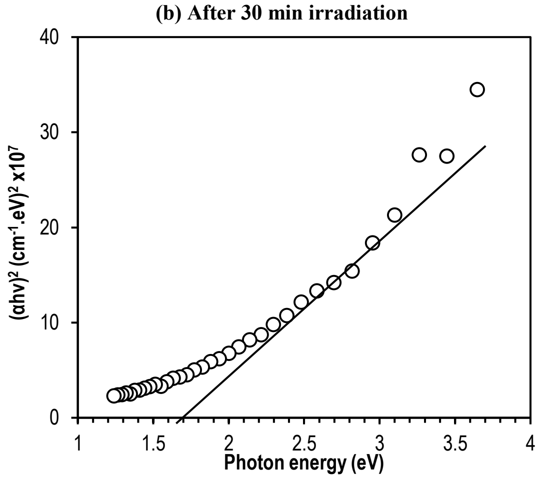
Fig. (1) XRD patterns of CdSe films subjected to cold plasma at various times (a) pure, (b) irradiated 30 min, (c) irradiated 60 min, and (d) irradiated 120 min



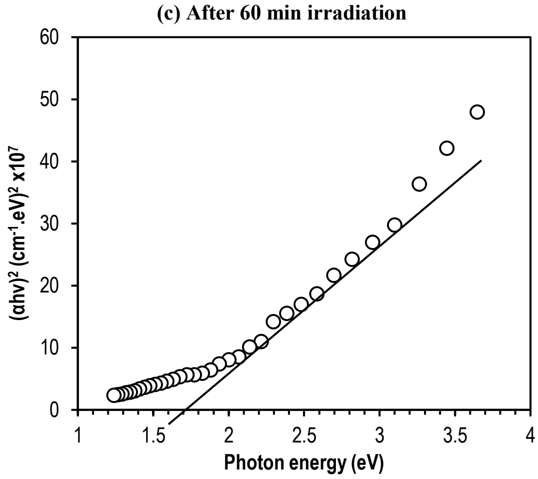
(a) Before irradiation



(b) After 30 min irradiation



(c) After 60 min irradiation



(d) After 120 min irradiation

Fig. (2) The plot between hυ and $(\alpha h\upsilon)^2$ to determine the energy gap for pure films exposed to cold plasma

Figure (3) shows the increased optical absorption accompanied by a decrease in transmittance after irradiation indicates the formation of additional defective states and structural disorder within the CdSe films, which enhances photon absorption and facilitates electronic transitions [32].

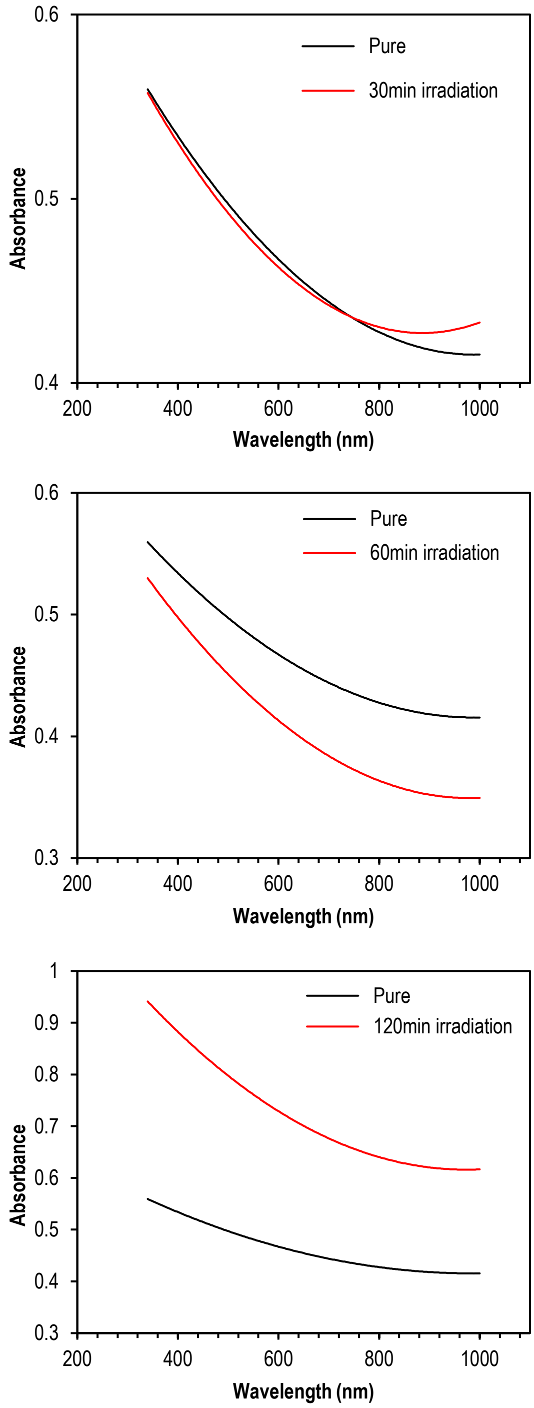


Fig. (3) Absorption spectra of pure CdSe films exposed to plasma at different time intervals

Figure (4) shows that longer exposure to cold plasma increases the Urbach energy. This means that the crystal disorder increases, and the number of local states within the energy gap increases, indicating that the plasma introduced new defects in the CdSe films [33]. Consequently, the structure became disorder increased after exposure to cold plasma. Although the reduction in band gap enhances visible-light absorption, the simultaneous increase in Urbach energy suggests a higher density of localized defect states, which may increase carrier recombination, a decrease in the energy gap and an improvement in optical absorption can be beneficial for optical, electronic, and solar cell applications. Therefore, plasma treatment achieves a balance between improving light absorption and increasing defect density.

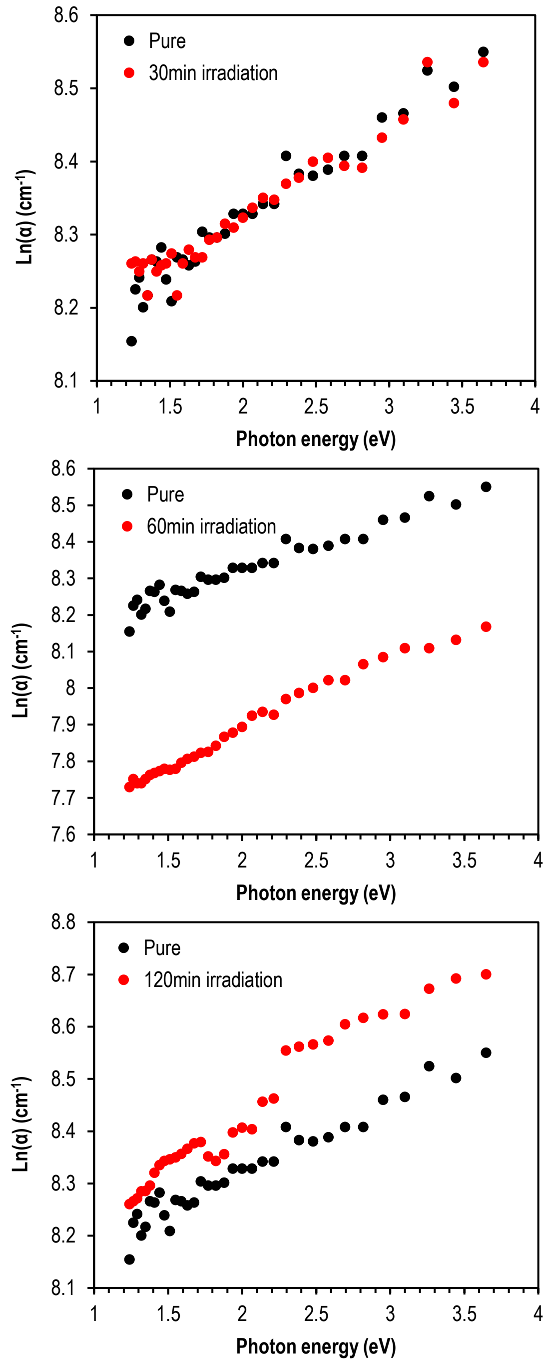
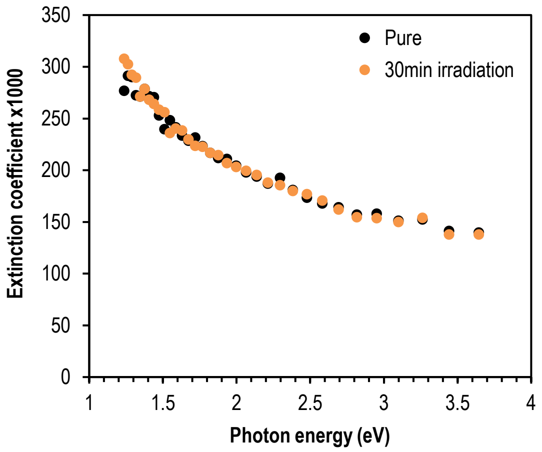
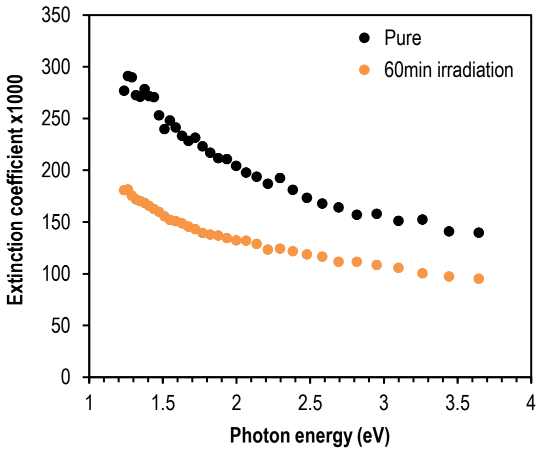


Fig. (4) Urbach energy curves of pure CdSe films exposed to cold plasma at different time intervals

Figure (5) shows that the extinction coefficient increases after exposure to cold plasma. This means that the absorption coefficient has increased, i.e., the material has become more absorbent of photons. This increase is a result of the formation of new absorption centers due to surface defects.

Cold plasma treatment induces subtle structural changes in materials due to the interaction of particles (ions and electrons) with the sample surface. When these particles collide with the surface, kinetic energy is transferred to the surface atoms, causing partial rearrangement of the crystal lattice and relaxation of residual internal strains, thus reducing the crystal strain. This irradiation displaces some atoms from their lattice positions, creating point defects and localized defect states, particularly at grain boundaries. These defects lead to an increase in localized electronic states within the energy gap, resulting in an increase in Urbach energy and extinction coefficient, and a decrease in the energy gap due to electronic transitions across the localized defect states and increased optical absorption. The results of this study indicate the potential of using cold plasma as an effective means of controlling the structural and optical properties of CdSe films. This opens the door to its application in solar cells for improving optical absorption, photosensitive sensors by modifying the energy gap, and modern electronic and optical devices such as photodiodes and semiconductor materials used in optical systems.





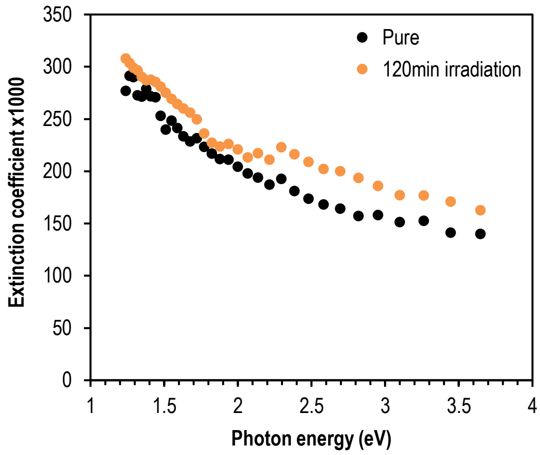


Fig. (5) Variation of extinction coefficient with photon energy of pure CdSe films exposed to cold plasma at different time intervals

4. Conclusion

CdSe films were produced by chemical bath deposition at 50°C and pH 9 for three hours. These films were subjected to cold plasma for varying durations of 30, 60, and 120 minutes at 0.63 mA of current and 9 kV of voltage and at a frequency of 100 kHz and power 5.67 W. Results showed a shift of the diffraction peaks towards lower angles after cold plasma irradiation, indicating expansion of the crystal lattice and the regularity of the crystal structure improved as a result of the reduction in strain. An increase in crystal size was observed, attributed to improved crystal growth and atomic rearrangement within the films as a result of the effective energy generated by the plasma. The results also confirmed the possibility of using cold plasma as an effective and low-temperature method for controlling the structural and optical properties of CdSe films, suitable for electronic and optical applications.

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Table (1) Structural parameters of pure and exposed CdSe films to cold plasma and deposited at a temperature of 50°C

Model type	2θ (deg)	(hkl)	d-spacing (Å)	β (deg)	Crystallite Size (nm)	Strain x10^{-3}
Pure	25.59	(111)	3.477	2.10	3.90	39
	42.50	(220)	2.125	2.05	4.14	22
	50.14	(311)	1.818	2.07	4.15	19
Irradiated for (30 min)	25.56	(111)	3.484	1.272	6.40	24
	42.45	(220)	2.129	0.590	15.00	6
	49.82	(311)	1.830	0.438	22.50	3
Irradiated for (60 min)	25.66	(111)	3.471	1.257	6.70	23
	42.51	(220)	2.126	0.740	12.20	7
	49.96	(311)	1.825	0.576	15.00	5
Irradiated for (120 min)	25.45	(111)	3.499	1.468	5.60	27
	42.17	(220)	2.142	0.941	9.60	10
	50.37	(311)	1.811	0.494	19.20	4

