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Determining Optical Band Gap Energy of Chitosan Biopolymer Film as the Effect of Gamma Rays Irradiation

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ABSTRACT

Dosimeter is one of the apparatus needed by radiation workers. From a dosimeter, radiation workers can control the absorbed dose. This research aims to investigate the properties characteristics of chitosan film for dosimeter usage. Thin-film chitosan polymer blended starch and methyl orange were prepared by phase inversion methods. The spectra were investigated by UV-Visible spectrophotometric in the wavelength range of 400-500 nm, while the optical band gap energy was investigated by the Tauch plot method. The increase in gamma rays irradiation dose affected the optical bandgap energy. It was observed that the value of band gap energy within the direct transition, indirect transition, and direct forbidden transition decreased along with the increase of gamma rays irradiation 3 and 7 kGy doses. These results indicated that gamma-ray irradiation could cause structural defects due to the excitation of non-bonding electrons. These structural defects could reduce the value of band gap energy because of the width localized states.

INTRODUCTION

Nuclear technology has developed very rapidly over time. Various scientific disciplines, nuclear technology used in various fields such as medical diagnosis and imaging, sterilization, therapy treatment, criminal investigation, agriculture, and space exploration. With the higher usage of nuclear technology in the environment, the usage of dosimeter more increase. This method of measuring the quantity of radiation is also known as dosimetry. Meanwhile, a series of tools used to measure the ionizing radiation exposure received by each person in a radiation field is called a dosimeter. The dosimeter study focuses on the development of active ingredients capable of responding to exposure to either particle radiation or electromagnetic waves (American Nuclear Society, 2014).

Dosimeter is one of apparatus must be used to radiation employee. Radiation exposure can affect healthy damage such as skin burned, cancer, cataract, etc. Nowadays, the Indonesian government is still import dosimeters from abroad. So, this research will explore about characteristics of synthesized dosimeter from polymer. Polymers in the form of thin films (layer) are one of the materials that can be used for the manufacture of dosimeters. The performance effects of this thin-film polymer are influenced by several factors including resistance to chemicals and most importantly the response it generates when interacting with radiation (Suman et al., 2015). This research will discuss the characteristics of the thin film chitosan polymer blended starch and methyl orange as dosimeter potential ingredients against gamma radiation. Chitosan and starch are compounds that are very abundant and cheap in Indonesia, with the hope that the use of these two compounds can increase the added value of both.

Various studies have been developed to obtain dosimeter-like potential active ingredients such as gels from metal complex Cr(III) and 1,5-diphenyl carbazon (DPC) (Gafar et al., 2018); Reactive Dye Red 120 (RR-120) (Paul et al., 2014); tetrabromophenol blue-dyed poly(vinyl alcohol) (Beshir, 2013); 2,6-Dinitrophenol (Gafar & El-Ahdal, 2014); toluidine blue O-Gelatin (Gafar et al., 2014); polyvinyl chloride dyed with bromocresol purple (Kattan et al., 2011); flexible poly(chloroprene)/methyl red film (Suman et al., 2015); organic films based on a diarylethene molecule (Asai et al., 2020); etc. Research on dosimeter potential active ingredients is still developing until now to get dosimeters with precise and accurate performance.

The characteristics of the thin film chitosan polymer blended starch and methyl orange that will be studied in this study are optical characteristics as a result of the interaction or response produced due to exposure to gamma radiation. Optical characteristics can be studied through the use of UV-Visible spectrophotometry instruments. One of the optical characteristics that can be identified from the UV-Visible spectrophotometry instrument is the optical bandgap energy. UV-Visible can identify the absorption spectrum of the thin film chitosan polymer blended starch and methyl orange material. The resulting spectrum can be determined by the bandgap or energy gap through Tauc plot methods. The value of the bandgap is the difference or gap between the valence band and conductivity band. Optical absorption involves the absorption of photons accompanied by changes in electron energy. If enough energy is absorbed, this energy is used to transfer negative charge or electron from valance band to conduction band, the gap energy required is the minimum energy to excite negative charge from valance band to conduction band (Mergen & Arda, 2020).

The calculation of the energy gap can be done using the Tauch plot method as in previous studies (Chikaoui, 2019). Tauch plot method is a method of determining is the minimum energy to excite negative charge from valance band to conduction band. A study of the Tauch gap was carried out on several materials such as boron-doped ZnO thin films (Alsaad et al., 2020); Ag_2O doped sodium antimonate glass-ceramics (Ashok et al., 2020); polymer blend composites PVA-PVP (Aziz et al., 2017); CuO for optoelectronic (Babu et al., 2020); ultrathin films TiO_2 (Bouzourâa et al., 2019); CdSe nanostructured thin film (Ghobadi et al., 2020); thin films Pb-Se-Ge (I. Sharma et al., 2019); yttrium aluminum iron (Kumar et al., 2020); kesterite $Cu_2ZnSnS(Se)$ (Mamedov et al., 2020); RGO (Merazga et al., 2020); $As_{40}Se_{53}Sb_{07}$ thin films (Pradhan et al., 2018); LiF-SrO-B_2O_3 glasses (Ramesh Babu & Yusub, 2020); semiconductor material (Sangiorgi et al., 2017); $BiFeO_3$ nanoparticles (S. Sharma & Kumar, 2020); $Cr_{2-x}Mg_xO_3$ (Singh et al., 2019); heterometallic La (Wang et al., 2020); etc. Research conducted by Chikaoui (2019) on the gamma irradiation effect on structural properties of thin-film PET showed that the value of absorbance in the range of 310-355 nm increases with the higher gamma irradiation dose in the range of 0.05–5 MGy. Both direct (m = 1/2) and indirect (m = 2) energy band gaps are observed decreasing with increasing of gamma irradiation dose. Thus, the new electronic state transitions exist (Chikaoui, 2019).

In the present investigation, this study aims to determine the characteristics of the optical gap as an effect of exposure to gamma rays on thin-film chitosan polymer blended starch and methyl orange. Unirradiated samples will be compared thus optical gap with samples irradiated over several dose ranges. After the irradiation process, the absorbance of unirradiated and irradiated thin-film chitosan polymer blended starch and methyl orange were calculated using a UV-Visible spectrophotometer in the 400-500 nm.

METHODS

The type of this research is experimental research which takes a period of about six months and needs to be analyzed by the quantitative method by plotting it in Matrix Laboratory (MatLab) software. Ordinate of $(\alpha h\upsilon)^{1/m}$ $(cm^{-1}eV)^{1/m}$ on the axis of $h\upsilon$ (eV) must be input in MatLab software to analyze the value of optical gap (Eg). Synthesis of irradiated thin-film chitosan polymer blended starch and methyl orange were 25 ml of Chitosan 1% w/v, 7.5 mL of 1% w/v starch solution, and 5 mL of 0.01% v/v dyes methylene orange added together. The mixed solution was stirred at 52 to 64^0C. After gelatin formed, the solution was cast on flat glass and dried until 70^0C. This method was called phase inversion. Each film was shaped to a size of about 1x 4 cm with 1 mm thickness (Ariyanti, 2020). Then, the sample was irradiated in gamma irradiator. The irradiation source is ^{60}Co with a dose rate of 4 kGy/hour in 3 and 7 kGy doses (at Polytechnic Institute of Nuclear Technology Yogyakarta, National Nuclear Energy Agency of Indonesia). The absorbance in wavelength between 400 nm and 500 nm of unirradiated and irradiated thin-film chitosan polymer blended starch and methyl orange determined by Shimadzu UV Visible spectrophotometer. Plotting ordinate of $(\alpha h\upsilon)^{1/m}$ $(cm^{-1}eV)^{1/m}$ on the axis of $h\upsilon$ (eV) on in MatLab software and draw a straight line until cut off-axis could represent the value of band gap energy of chitosan biopolymer film as the effect of gamma rays irradiation.

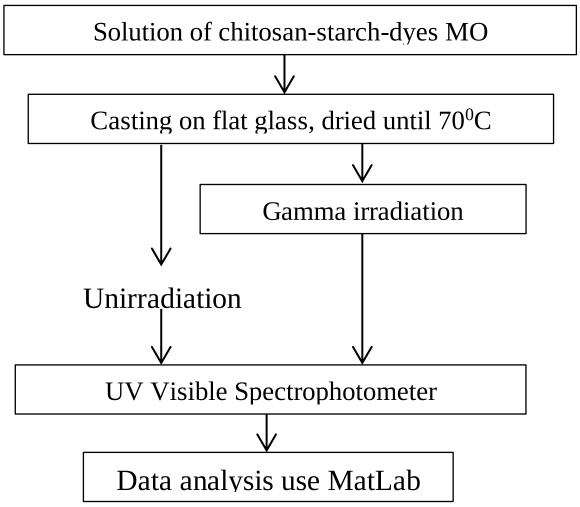

Figure 1. Flow chart method of analysis bandgap energy on chitosan film polymer

RESULTS AND DISCUSSION

Absorption spectra of unirradiated and irradiated thin-film polymer blended chitosan-starch-methyl orange than determined by Shimadzu UV Visible spectrophotometer. The absorbance of unirradiated and irradiated thin-film chitosan polymer blended starch and methyl orange was changed (Figure 2). When the energy of a photon absorbed is less than the width of the energy bandgap, the electron is unable to move to a higher energy level. However, when the photon energy absorbed is greater than the width of the energy bandgap, the electrons in the valence band can move towards the conduction band (Palupi et al., 2019).

Curve plotting of wavelength versus absorption than analyzed by Tauch plot methods to get optical band gap energy value. Tauch plot method is a method of determining the optical band gap by looking at the linear relationship graph Energy (eV) is in axis dan $(\alpha h\upsilon)^{1/m}$ is in ordinate. Relation between photon energy ($h\upsilon$) and absorption coefficient (α) determined by equations 1 (Chikaoui, 2019):

$$(\alpha h\upsilon)^{\frac{1}{m}} = c(h\upsilon - Eg) \quad (1)$$

Where h is Planck's constant 6,63 x 10-34 J.s. c, Eg, and m represent a proportionally constant. The m parameter takes 1/2 for the direct gap (direct transitions), 2 for the indirect gap (indirect transitions), and 3/2 for the direct forbidden gap (the direct forbidden transitions). The bandgap energy Eg was investigated from the plot of $(\alpha h\upsilon)^{1/m}$ versus $h\upsilon$ (Chikaoui, 2019).

Increasing the irradiation dose causes an increase in the absorbance value of the material. Figure 3 investigated that the curve of $(\alpha h\upsilon)^{1/m}$ $(cm^{-1}eV)^{1/m}$ (axis x) on photon energy $h\upsilon$ (eV) (axis y) for unirradiated film chitosan polymer blended starch and methyl orange is calculated in three states, $m = 1/2$ direct transition (A), $m = 2$ indirect transition (B), and $m = 3/2$ direct forbidden transition (C). While Figure 4 and 5 show that the curve of $(\alpha h\upsilon)^{1/m}$ $(cm^{-1}eV)^{1/m}$ (axis x) on $h\upsilon$ (eV) (axis y) for irradiated film chitosan polymer blended starch and methyl orange in 3 kGy and 7 kGy, respectively, when calculated in three states $m = 1/2$ direct transition (A), $m = 2$ indirect transition (B), and $m = 3/2$ direct forbidden transition (C). Accumulation of Eg of unirradiated and irradiated thin-film polymer blended chitosan-starch-methyl orange in 3 kGy and 7 kGy shows in Table 1 and Figure 6.

Several studies conducted studies on the effect of gamma-ray irradiation on the energy bandgap (Eg) in thin-film PET (Polyethylene Terephthalate) (Aarya et al., 2012). Similar studies have been also resulted in other materials, as reported of defect on PADC (Poly Allyl Diglycol Carbonates) (Zaki & Elmaghraby, 2012). The result of the previous experiment was similar to the result of this experiment. Based on the research that has been done, the results show that the irradiation dose of thin-film chitosan polymer blended starch and methyl orange is inversely proportional to the value energy bandgap (Eg). The higher the irradiation dose, the lower the energy bandgap (Eg) value produced. An increase in irradiation dose which is inversely related to the energy band gap value was also reported in previous studies.

Aarya et al., (2012) stated that gamma radiation can affect electronic disorder. This statement is appropriate with this experiment result. Figure 6 indicates that the existence of direct transition or direct band gaps, indirect transition or indirect gap, and direct forbidden transition in the thin film chitosan polymer blended starch and methyl orange with decreasing at higher gamma irradiation dose. These results showed that the irradiation gamma produces faults in film chitosan polymer blended starch and methyl orange structure such as a broken bond, free radical, etc. Termination or broken bond and formation of free radical made increase the electronic disorder. The electronic disorder can disturb the valence band which creating a permitted state of the forbidden gap.

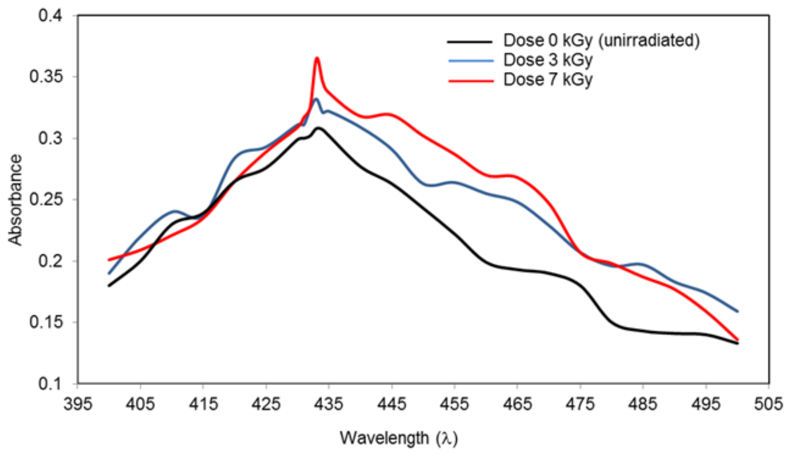

Figure 2. Absorption spectra of unirradiated and irradiated film chitosan polymer blended starch and methyl orange in 3 and 7 kGy

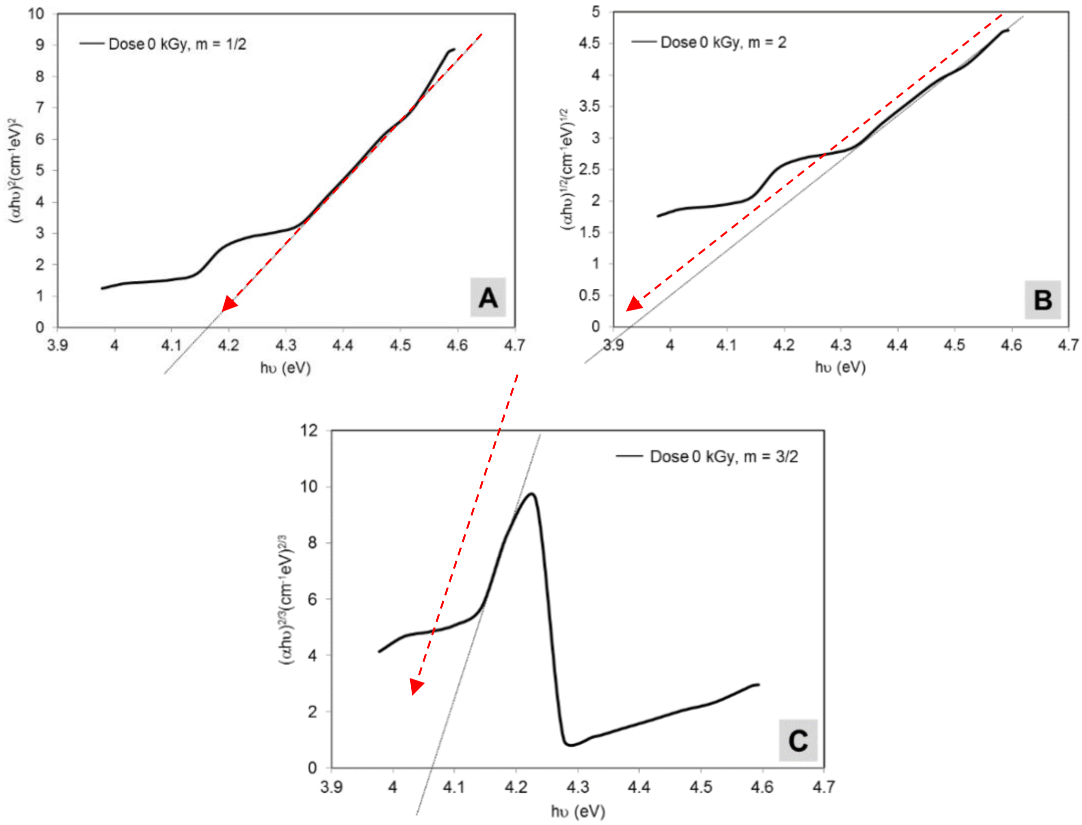

Figure 3. Curve of $(\alpha h\upsilon)^{1/m}$ $(cm^{-1}eV)^{1/m}$ on $h\upsilon$ (eV) on unirradiated film chitosan polymer blended starch and methyl orange: (A) m 1/2 (B) m 2 (C) m 3/2

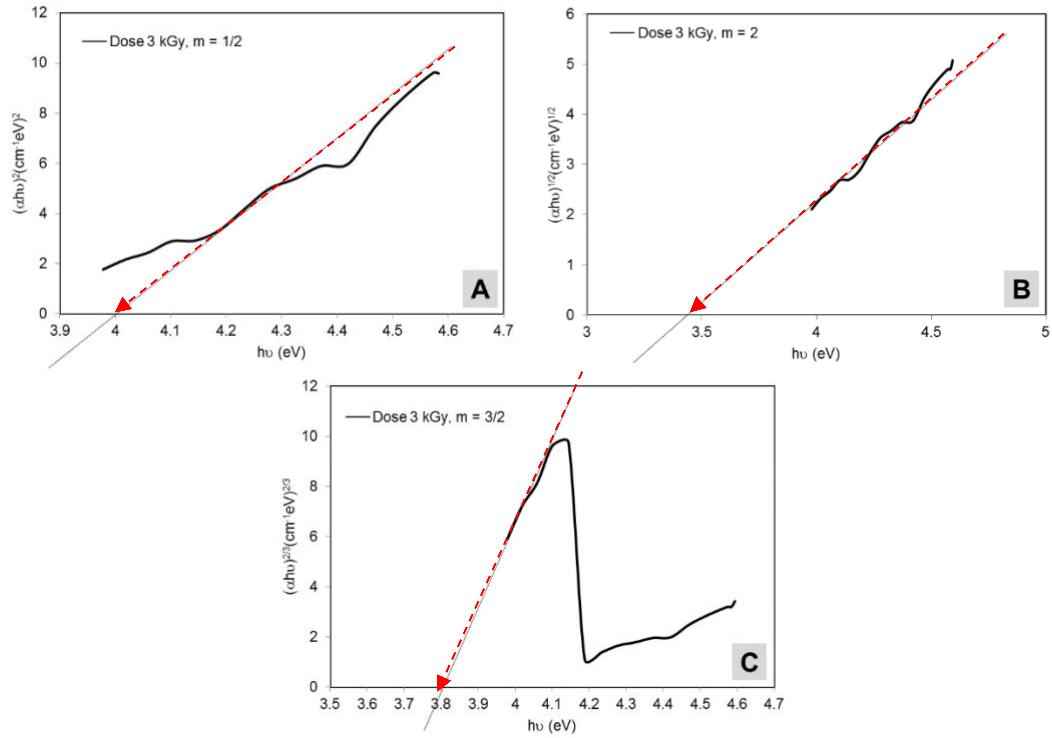

Figure 4. Curve of $(\alpha h\upsilon)^{1/m}$ $(cm^{-1}eV)^{1/m}$ on $h\upsilon$ (eV) on irradiated film chitosan polymer blended starch and methyl orange in 3 kGy: (A) m 1/2 (B) m 2 (C) m 3/2

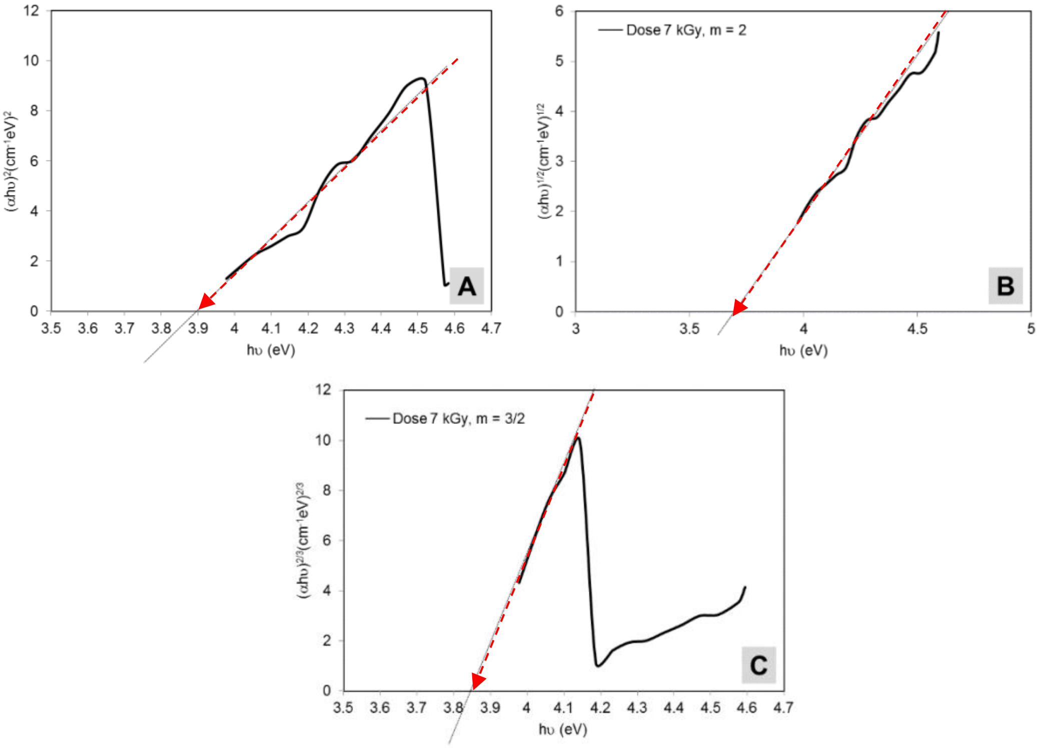

Figure 5. Curve of $(\alpha h\upsilon)^{1/m}$ $(cm^{-1}eV)^{1/m}$ on $h\upsilon$ (eV) on irradiated film chitosan polymer blended starch and methyl orange in 7 kGy: (A) m 1/2 (B) m 2 (C) m 3/2

Table 1. Accumulation of Eg of unirradiated and irradiated thin-film polymer blended chitosan-starch-methyl orange in 3 kGy and 7 kGy

Dose (kGy)	Gap Energy, Eg (eV)		
	1/2	2	3/2
0	4.17	3.93	4.06
3	4	3.45	3.8
7	3.9	3.7	3.85

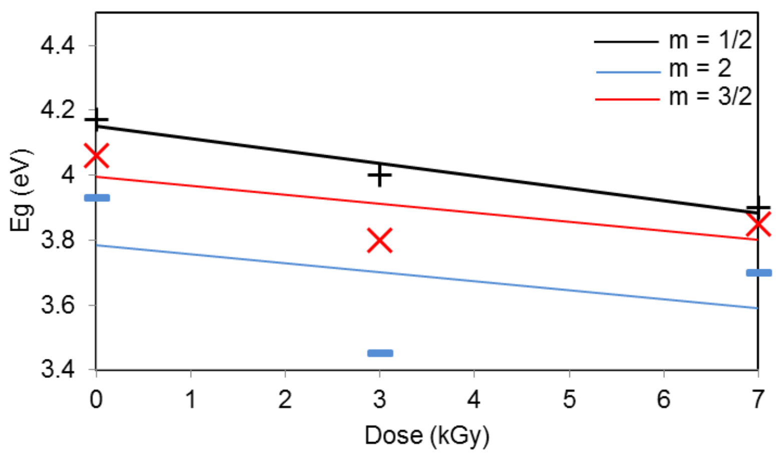

Figure 6. The dependence of Eg (eV) on irradiation dose of thin-film chitosan polymer blended starch and methyl orange

In the other literature El-nahass et al., (2012) and Aarya et al., (2012) states that the presence of gamma-ray irradiation may cause structural defects on excitation of non-bonding electrons because of the formation of free or ions radical into the conduction band. This theory is significant with the result of this experiment. It can be seen from Figure 6 that the energy gap, Eg, decreases linearly because of gamma irradiation. It's appropriate that gamma-ray irradiation could affect the structural defects that lead by could reduce the value of the energy band gap because of increasing width localized states. Increasing width localized states on the formation of free or ions radical would lead to decrease transition probabilities into the extended states. Thus, extended states could enhance absorption and the conductivity of the sheets. Furthermore, the values of the energy gap of indirect transition are observed to be lower than the value of the energy gap within direct transition.

CONCLUSION

Based on the result of the experiment, it can be concluded that chitosan biopolymer film was potentially be used as a dosimeter. Gamma irradiation with the variation of dose exposure has affected the optical properties of film chitosan polymer blended starch and dyes methyl orange. It's observed that the energy gap value decrease as dose gamma irradiation increases. This indicated that gamma-ray irradiation could cause structural defects due to the excitation of non-bonding electrons. This structural defect could reduce the value of the optical band gap because of increasing width localized states.

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