Malaysian Journal of Analytical Sciences (MJAS)
Published by Malaysian Analytical Sciences Society

COMPARATIVE STUDY ON THE EFFECT OF GAMMA RADIATION ON BIOPLASTICS

(Kajian Perbandingan Tentang Kesan Sinaran Gamma Terhadap Bioplastik)

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Received: 20 January 2023; Accepted: 9 May 2023; Published: 23 June 2023

Abstract

Conventional plastic is made from petrochemical substances. Its non-biodegradable properties negatively impact the environment, causing landfill pollution and affecting wildlife and aquatic life. It uses non-renewable sources that will be depleted in a few years if there is no solution to this situation. This study was carried out to produce bio-plastic made from renewable sources: 10 g of corn starch, 5 ml of glycerol and vinegar, and 60 ml of distilled water. The produced bio-plastic was exposed to gamma radiation to observe the difference between irradiated and non-irradiated bio-plastics. Surface morphology and chemical properties like absorption spectra, functional group and chemical bonding were characterized using Opto Digital Microscope, Ultra-Violate Visible (UV-Vis) Spectrophotometer and Fourier Transform Infrared Spectroscopy (FTIR). The results indicate that exposure to gamma radiation causes the bio-plastic surface to be darker with some degradation. Irradiated bio-plastic degrades faster than the ones not irradiated. The samples were characterized using UV-Vis, prepared in different glycerol concentrations to determine which concentration gave the best absorption peak. The absorption peaked at 270 nm wavelength in the highest glycerol concentration. Functional groups like alcohols, carbonyl and ester were also found in the sample's solution. All functional groups that contribute to the bio-plastic properties could be stretched and bent to enhance their performances as a packaging application. Overall, exposure to gamma radiation improves the bio-plastic quality as a packaging application that can be used in the food and pharmaceutical industries.

Keywords: Effect of gamma radiation, bioplastics, radiation, ionizing

Abstrak

Plastik konvensional diperbuat daripada bahan petrokimia. Sifat tidak terbiodegradasinya memberi kesan negatif kepada alam sekitar, menyebabkan pencemaran tapak pelupusan dan menjejaskan hidupan liar dan akuatik. Ia menggunakan sumber tidak boleh diperbaharui yang akan habis dalam beberapa tahun jika tiada penyelesaian kepada keadaan ini. Kajian ini dijalankan untuk menghasilkan bio-plastik yang diperbuat daripada sumber boleh diperbaharui: 10 g kanji jagung, 5 ml gliserol dan cuka, dan 60 ml air suling. Bio-plastik yang dihasilkan telah didedahkan kepada sinaran gamma untuk melihat perbezaan antara bio-plastik yang disinari dan tidak disinari. Morfologi permukaan dan sifat kimia seperti spektrum serapan, kumpulan berfungsi dan ikatan kimia telah dicirikan menggunakan Mikroskop Opto Digital, Spektrofotometer Cahaya Nampak-Ultraungu (UV-Vis) dan Inframerah Transformasi Fourier (FTIR). Keputusan menunjukkan bahawa pendedahan kepada sinaran gamma menyebabkan permukaan bio-plastik menjadi lebih gelap dengan sedikit degradasi. Bio-plastik yang disinari merosot lebih cepat daripada yang tidak disinari. Sampel telah dicirikan menggunakan UV-Vis, disediakan dalam kepekatan gliserol yang berbeza untuk menentukan kepekatan

yang memberikan puncak penyerapan terbaik. Penyerapan memuncak pada panjang gelombang 270 nm dalam kepekatan gliserol tertinggi. Kumpulan berfungsi seperti alkohol, karbonil dan ester juga ditemui dalam larutan sampel. Semua kumpulan berfungsi yang menyumbang kepada sifat bio-plastik boleh diregangkan dan dibengkokkan untuk meningkatkan prestasi mereka sebagai aplikasi pembungkusan. Secara keseluruhan, pendedahan kepada sinaran gamma meningkatkan kualiti bio-plastik sebagai aplikasi pembungkusan yang boleh digunakan dalam industri makanan dan farmaseutikal.

Kata kunci: Kesan sinaran gama, bioplastik, sinaran, pengionan

Introduction

Radiation processing is used for packaging applications, especially for medical products and in the food industry [1]. Presently, packaging materials are mostly made from synthetic plastic, making it crucial to see the effect of irradiation on these materials. Apart from radiation treatment, a few other methods can be applied for packaging applications, such as UV-ray, ionizing ray, and light pulse. There was an attempt to use UV-ray for surface sterilization. It depends on the intensity rate applied for the packaging applications. However, the UV ray's penetration ability is limited [2]. Hence, it is only suitable for packaging materials with very low contamination.

Using an ionizing ray is another method for packaging applications. It uses Cobalt-60 as a source that emits gamma radiation. It is very convenient for killing bacteria [2]. Ionizing radiation is commonly used for bulk packaging of acidic foods such as aseptic bag-in-box. This method applies a radiation dose of 25 kGy equivalent to 2.5 mrad. However, the effects of gamma radiation on food have been a huge concern in the manufacturing process because it needs heavy shielding to prevent radiation leakage. It also requires careful process design, high maintenance, and expensive equipment.

Radiation treatment is chosen to radiate packaging materials because it does not leave any residue on the treated surface nor affect the immediate environment, making it the perfect solution for packaging materials. Packaging materials are any material used to protect something. Many sources have been used in packaging materials, including glass, metals, papers and plastics. However, a special variety of plastics have been introduced in the manufacturing industry for packaging. These plastics are made from petroleum and other chemicals with unhealthy side effects for the product and its surrounding, leading to the invention of bio-plastic, a more convenient and safer packaging material.

Plastics have been widely used for packaging applications for 70 years [3, 4]. Its versatility and durability give it an advantage over other materials. It is inexpensive, lightweight, heat-sealable and has good flexibility for fabrication into various shapes [5]. However, petroleum-based plastics are detrimental to the environment. Conventional plastics are made from petroleum, making them non-biodegradable and non-renewable [6, 7]. This disadvantage leads to huge environmental problems and municipal waste. The solution to this problem is to produce a new type of plastic made from bio-based materials. Unfortunately, recent studies found that no bio-based plastics could combine all the features and functionalities of conventional plastics [8]. Hence, there is ongoing research on producing bio-based plastics that achieve all the required properties of conventional plastics.

Another definition of bio-plastic can be explained from two perspectives. The first one is its functionality. Bio-plastic is biodegradable plastic from organic waste, while the second one is by its source materials. It uses renewable or biomass materials that are processed into plastic. Bio-plastics are a type of biological material that is degradable and eco-friendly in its chemical nature. They are produced using a few methods, such as microorganisms cultured under different nutrients and environmental conditions [9]. Bio-based plastics are man-made or man-processed, derived from biological resources. Bio-plastics are classified into two types: photodegradable and semi-biodegradable [10]. The history of bio-based plastics is much earlier than our conventional plastics. The first bio-based plastics, called thermoplastics, were made from celluloid in the 1860s [11]. Unfortunately, the numerous inventions of bio-based plastics stayed in the laboratory and was not

commercially exploited because, in the 1950s, the petrochemical industry had developed cheap and synthetic polymers made up from crude oils.

What we have now are synthetic plastics that are usually made from petroleum. 4% of the world's petroleum sources are reserved for manufacturing plastics [12]. More than 200 billion pounds of plastics are produced yearly [13], indicating how crucial plastic is worldwide. Within a few years, sources will deplete if nothing is done about it. These synthetic plastics contribute to environmental pollution because they are not eco-friendly. The chemicals used to make synthetic plastics are non-biodegradable, taking a long time to decay. Some plastics take up to 1000 years to decay [14]. On top of that, plastics also affect wildlife, destroying habitats, especially the marine ecosystem. Aquatic life often mistakes plastics for food, causing them to suffocate or entangled by the plastics [15].

Conventional plastics are compounded with several additives to improve their product performance and tensile strength. These factors lead to health hazards for humans. In addition, a very cautious handling process is needed during the plastic manufacturing, which may cause industrial accidents [16]. Our study is important for several reasons. Firstly, inventing a bio-plastic requires a study of its properties and functions to ensure it is similar to or better than conventional plastic for use as packaging. Hopefully, over time, bio-plastic will replace conventional plastic. Secondly, bio-plastic can help lessen global environmental pollution because it uses natural sources as its base. Therefore, bio-plastics are eco-friendly (bio-degradable) and take less time to decay. Thus, overcoming conventional plastic adverse effects on the marine ecosystem.

Thirdly, although producing bio-plastics may require a higher capital, it should not be a problem due to ample bio-based sources. Currently, there is a high demand for bio-plastic from the industry. Moreover, petroleum price is expected to increase due to its depletion and high demand. Finally, bio-plastic is irradiated using gamma radiation by emitting a high photon energy that can penetrate the material, ensuring the entire packaging has been radiated. It is a non-polluting radiation, making it safer for the products inside the packaging [9]. Furthermore, the properties of the packaging application were studied so that they could be customized for use in different applications, making it particularly beneficial to the food processing industry.

The objectives of this research are including produce biodegradable plastic using corn starch, analyze the chemical properties of irradiated bio-plastic by using ultraviolet-visible spectrophotometer (UV-Vis) and Fourier transform infrared spectroscopy (FTIR), and study the bio-plastic surface morphology using Opto Digital Microscope.

Materials and Methods

This study employed 10 g of corn starch, 60 mL of distilled water, 5 mL of glycerol, and vinegar. The sample was prepared by adding 60 ml of distilled water and corn starch. The mixture was stirred until it formed a uniform and homogenous suspension. Following that, glycerol was added to the mixture and stirred well. Then, vinegar was added to the mixture. After thoroughly combining all materials, the heat was turned on, and the stirring procedure was resumed. During the stirring process, the temperature was raised from 30ºC to 95ºC. After a while, a thick viscous glue-like substance formed. The mixture was then poured and spread over an aluminum foil. The mixture was left to dry for a few days, as shown in Figure 1. The bio-plastic sample was cut into smaller pieces and sliced into 5 cm x 5 cm squares to facilitate the radiation procedure. They were divided into radiation and non-radiation samples. The samples were then irradiated.

The shielding blocks were arranged properly for irradiation purposes to ensure no radiation leakage while running the experiment. Cobalt-60 was chosen as the radiation source. The samples were left for a week for irradiation. Finally, all samples were sent for analysis before and after each radiation process. The radiated samples were sent for analysis to study their properties and investigate the radiation effects on the sample using Ultraviolet-Visible Spectrophotometer (UV-Vis), Opto Digital Microscope and Fourier Transformation Infrared Spectroscopy (FTIR).

(a) (b) (c)

Figure 1. (a) Corn starch, glycerol, vinegar and distilled water were measured (b) The mixture was mixed together and stirred on heating stove with medium heat until thick viscous glue-like solution formed (c) The mixture was then poured onto aluminum foil and spread out to let it dry. The drying process took several days

Results and Discussion

Ultraviolet-Visible spectrophotometer analysis

10 samples were analyzed using UV-Vis spectroscopy. The samples were divided into radiated and non-radiated samples. Five samples were radiated using Cobalt-60 and exposed to gamma radiation for a week. Then, the samples were prepared for analysis using a UV-Vis Spectrophotometer to observe the bio-plastics absorbance range under different glycerol concentrations at 200-800 nm. UV-Vis uses visible and ultraviolet lights to analyze the bio-plastics chemical structure.

Table 1. Wavelength and absorption peak of non-radiated and radiated bio-plastics

Samples Glycerol (mL)	Non-Radiated Bio-Plastic		Radiated Bio-Plastic	
	Wavelength (nm)	Absorption Peak (a.u)	Wavelength (nm)	Absorption Peak (a.u)
1.0	280	0.031	270	0.057
0.8	270	0.022	270	0.030
0.6	270	0.019	270	0.028
0.4	270	0.018	270	0.012
0.2	270	0.016	270	0.010

Table 1 shows that glycerol influenced the UV absorptivity in bio-plastics [17]. This result also indicates that glycerol concentration is directly proportional to the bio-plastics ability to absorb UV (Figure 2). The highest peak for the non-radiated bio-plastics is 0.031 a.u with 1.0 ml presence of glycerol, found at 280 nm wavelength. The glycerol is directly proportional to the level of bio-plastic transparency [18]. Studying the bio-plastic absorbance range is important because UV absorption has a significant relationship to the plastic's UV degradation. Plastics that did not absorb UV radiation are insusceptible to photo degradation [19].

This research found that bio-plastic with 1.0 ml glycerol concentration had the highest peak. We conclude that glycerol in bio-plastic resulted in better UV absorption (Figure 3). The bio-plastics ability to absorb UV is very beneficial, especially if it is used for food packaging, due to their ability to protect food products from UV radiation. It could prevent photo-oxidative degradation that might change the product's taste and smell, and induce free radicals formation. The highest peak obtained is at wavelength 270 nm with a peak 0.057 a.u. The highest peak obtained had the same glycerol concentration as non-radiated bio-plastic. This signifies

that with exposure to gamma radiation, glycerol's presence in bio-plastic plays an important role in absorbing UV radiation.

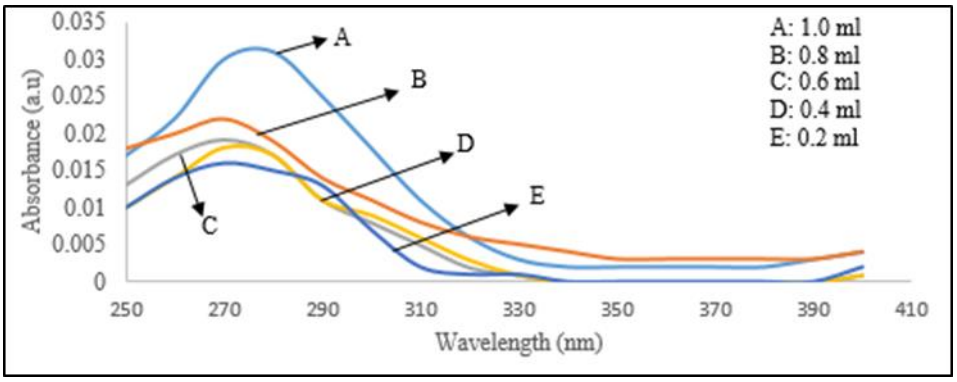

Figure 2. Concentration of different glycerol in non-radiated bio-plastics

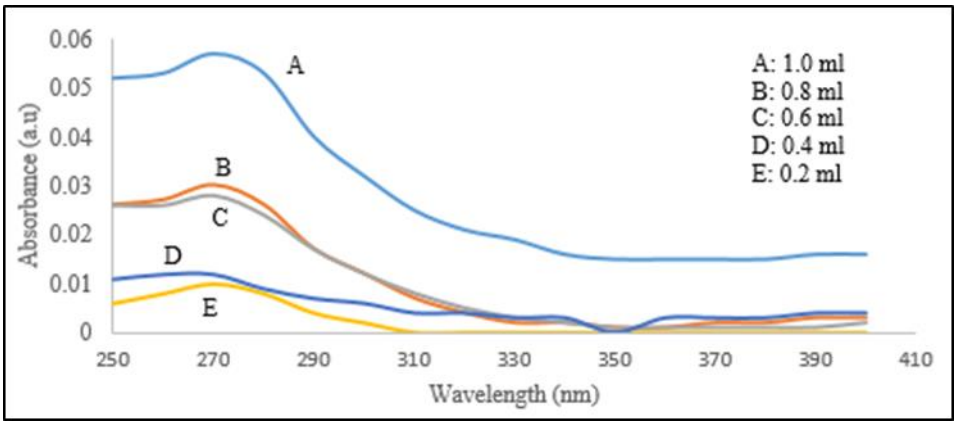

Figure 3. Concentration of different glycerol in irradiated bio-plastics

Opto-Digital Microscope DSX100

The opto-digital microscope is used to characterize the bio-plastic surface morphology. The particle distribution and deterioration effects from radiation could be seen using DSX100. The advantage of using this microscope is it magnifies and measures images in real-time. It also stores and transmits images for future viewing, making microscopy possible for beginners. The bio-plastics produced were transparent, smooth, thick and deformable, whereas conventional plastics were firm, stretchable, thin and transparent. Bio-plastic is quite soft compared to conventional plastics, making it difficult to compare the irradiation effects on both types of plastic.

The samples were analyzed for both irradiated and non-irradiated conditions. Figure 4(a) shows the surface condition of conventional plastic made of petrochemical substances. This sample was not exposed to a gamma source. Hence, the surface morphology was affected by the radiation. The surface was free of degradation effect and had a firm structure.

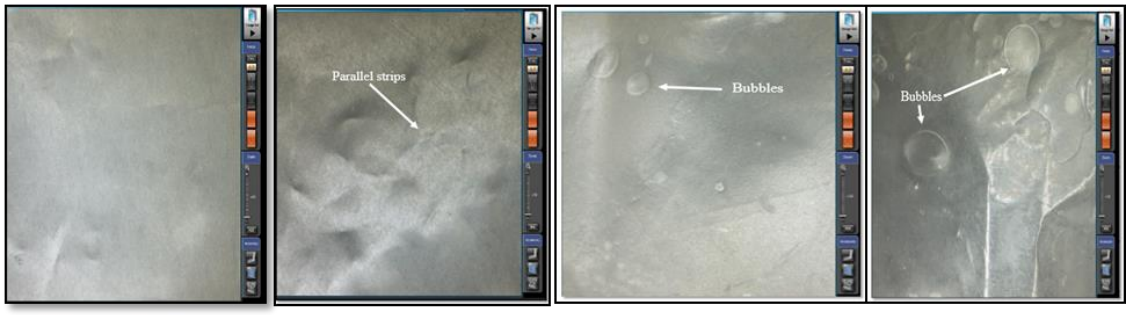

Figure 4. Surface morphology of (a) non-irradiated and (b) irradiated conventional plastic. While (c) non-irradiated (d) irradiated surface morphology of bio-plastic

Figure 4(b) shows the radiation effects on plastic. The plastic exposed to radiation deteriorated on its surface due to the oxygen reacting with the free radicals [6]. The free radical formation could significantly change the molecular structure [20]. Several parallel strips developed on the plastic's surface, possibly due to the radiation effects. Figure 4(c) shows the bio-plastic surface morphology in natural conditions. The surface is rigid due to using glycerol as a plasticizer agent. It shows the fracture surface of dispersed fibers without the incidence of gamma radiation [21]. The sample was brittle due to bubbles, which is problematic for the bio-plastic-making process.

The bio-plastic sample shown in Figure 4(d) has the embrittlement effect due to irradiation. Gamma rays induced chain scission and degradation effect [9], resulting in the melt viscosity and embrittlement observed in Figure 4(d). Because of radiation-induced oxidation, the surface is deepened with oxygen-containing hydrophilic groups [22], indicating that the bio-plastic surface becomes hydrophilic after irradiation. The surface of the irradiated bio-plastic also appeared darker than the non-irradiated ones because of the irradiation degradation effect. The exposure to gamma energy improved the degradation process. When exposed to radiation, the bio-plastics could be degraded by photolysis, giving the samples a lower molecular weight, thus enhancing and quickening the bio-plastics degradation process [23].

The bubbles on the surface resulted from the inadequate method when handling the samples during the drying process. The samples were left at room temperature for several days, promoting the bio-plastic surface's oxidation process. It is more convenient to dry the sample using an oven to avoid bubble formation [24]. The atmosphere in which the radiation took place modifies the effects. The differences are usually observed in the presence or absence of oxygen. However, the findings are inconsistent with some plastics degradation that was enhanced in the presence of air. This research used the natural drying process to manage the equilibrium of moisture content, which was critical to determine the ideal microbe development, material deterioration and degradation circumstances. It also aids in determining food stability at a certain moisture level in a specific environment.

Fourier transform infrared spectroscopy analysis

FTIR is chosen to describe the functional group present at the material surface before and after drying [24]. All samples were recorded in absorbance units from 4000 to 450 cm^{-1}. Four samples were analyzed which are irradiated bio-plastic and conventional plastic also non-irradiated bio-plastic and conventional plastic. Figure 5 shows the band at 3000-3600 cm^{-1} range spectra of irradiated bio-plastic indicate the O-H stretching which is broader than other absorption bands [25]. It is somehow overlaps the N-H stretching in the same region. There are four primary peaks shown in table 2 and 3 as similar to previous study [26]. The peak at 3293.22 cm^{-1} signifies the presence of alcohols group in the sample as glycerol was involved in the process of making the bio-plastic.

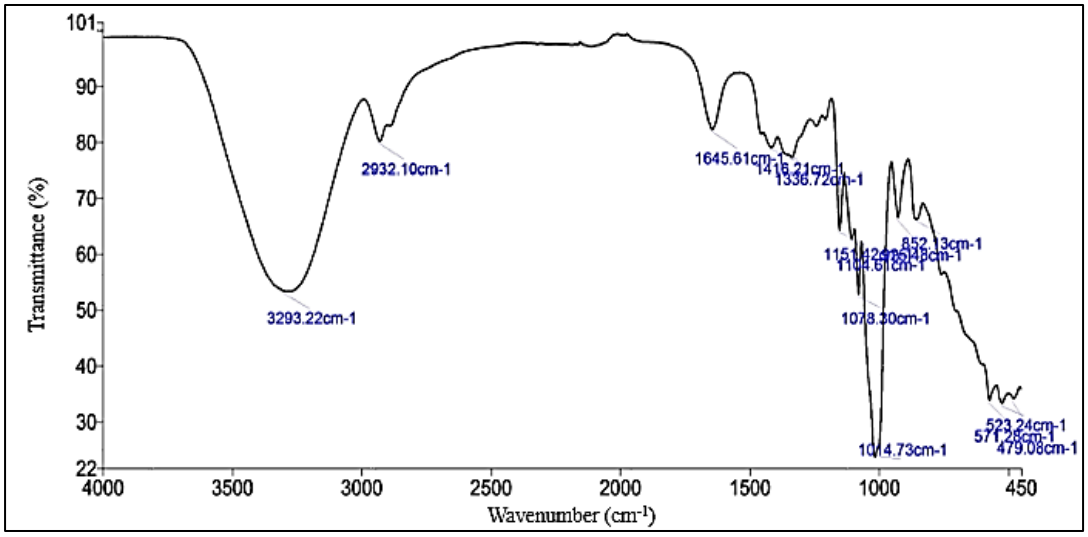

Figure 5. FTIR spectrum for irradiated bio-plastic

Another peak at 2932.10 cm^{-1} indicates the presence of the alkanes group, i.e., the C-H bond, which has medium to strong infrared absorption intensity [27]. The C=O bond at 1645.61 cm^{-1} is attributed to the stretching vibrations of strong carbonyl compounds due to the presence of ketones or aldehydes compounds produced due to thermal decomposition during the sample preparation [28, 29]. Peaks at 1014.73 cm^{-1} were due to the presence of alcohol groups C-O bond involved during the polymerization process, meaning that the starch alcohols group was involved in the hydrogen bond formation. Still, it is not as broad as the O-H bond.

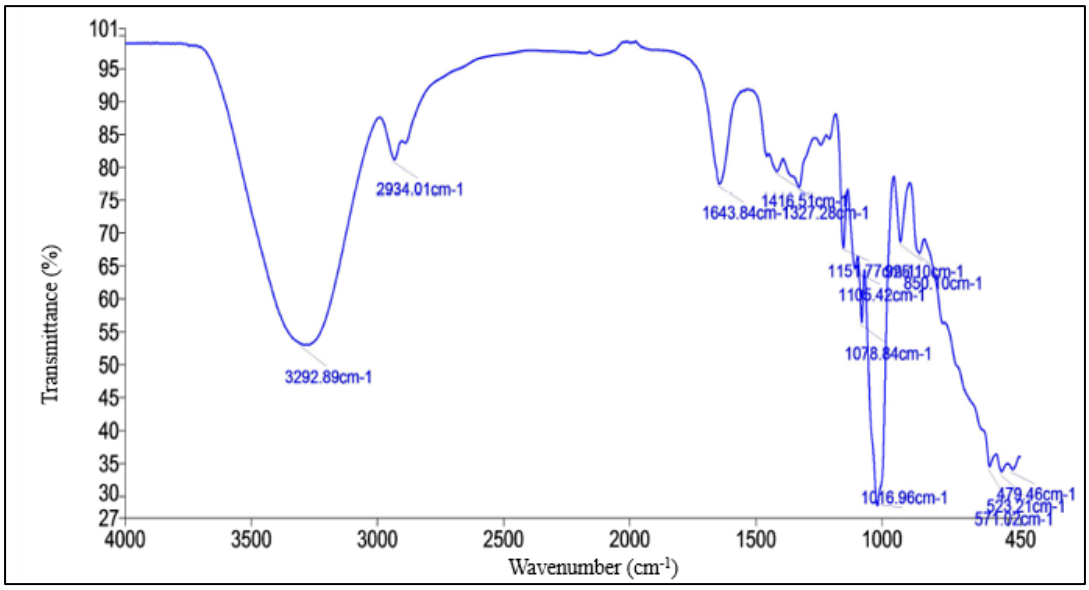

Figure 6. FTIR spectrum for non-irradiated bio-plastic

Table 2 shows the absorption spectrum for non-irradiated bio-plastic. Figure 6 shows no significant changes occurred from irradiated and non-irradiated samples except the slightly different values of wavenumber shown by the four primary peaks. However, a lower wave number indicates a stronger hydrogen bond interaction [30]. Starting from the first peak, there is a decreasing value of the alcohol group

absorption band for the non-irradiated bio-plastic and a decreasing carboxylic acids presence. Table 2 shows a radical cation, likely generated on the carbonyl function under gamma radiation. Another effect was the generation of radical cation-beta on the ester oxygen, providing an alkyl radical and an oxonium ion. The latter collapsed to release acetic acid and a carbocation. The free radical generated an oxyl radical with a carbocation in an alpha position.

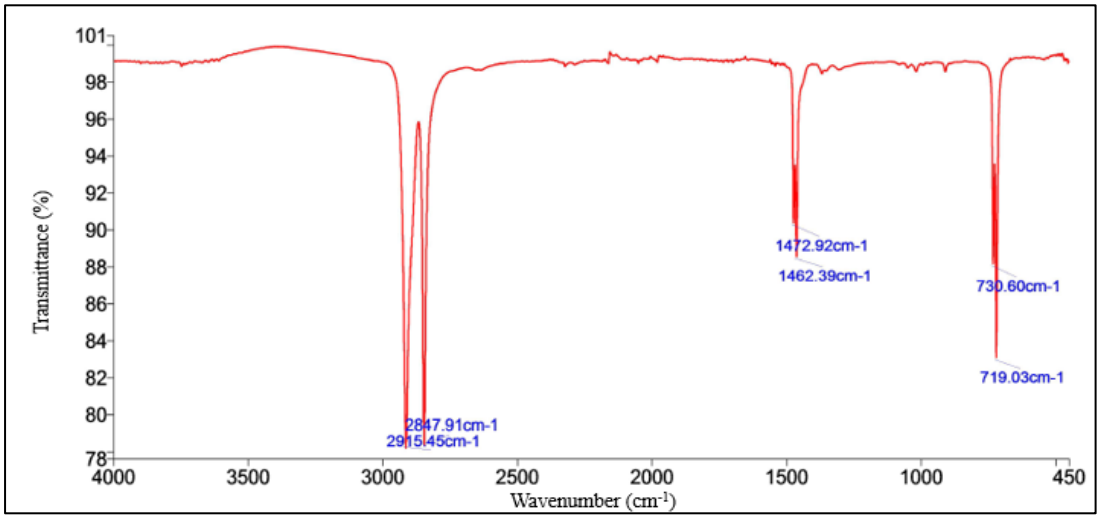

Figure 7. FTIR spectrum for irradiated conventional plastic

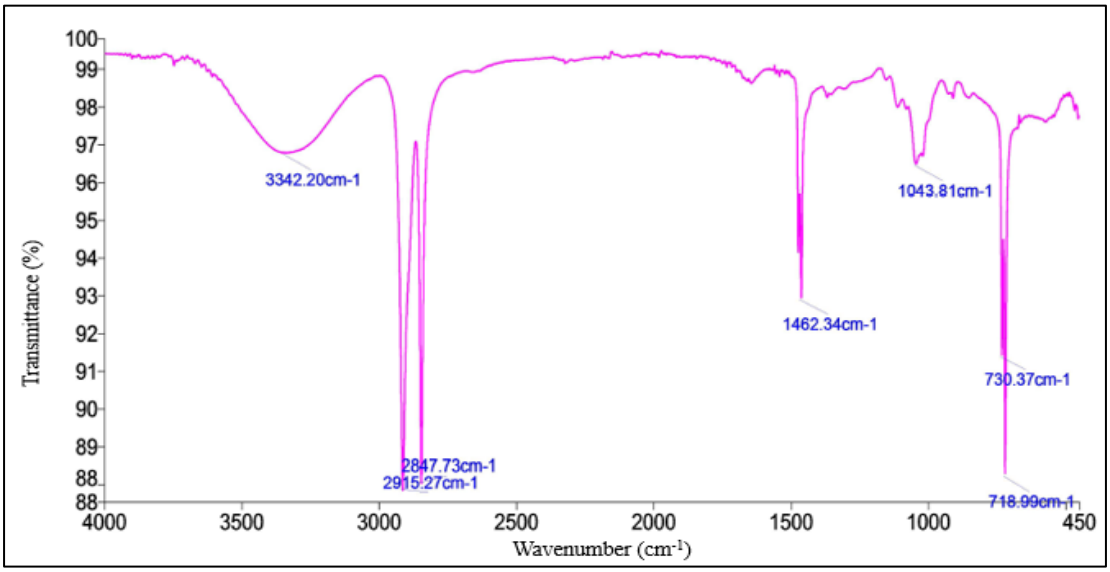

Figure 8. FTIR spectrum for non-irradiated plastic

Table 3 shows the IR spectra for both irradiated and non-irradiated plastics. No significant changes were obtained from both results except a few reduced peaks at certain wave numbers of absorption bands (Figure 7). This agrees with a previous study that there were completely unchanged peaks even after the gamma exposure [30]. The most significant difference is the reduced peak at 3342.20 cm^{-1} of non-irradiated plastic exposed to gamma radiation. The gamma exposure caused O-H scission, resulting in the formation of double bonds. The

peaks in the range of 2847-2915 cm^{-1} are similar in both plastics. These show the presence of phenolic groups O-H, which has a very broad and strong absorption bands intensity. This is due to the plastics being irradiated with gamma exposure, leading to radical oxygen formation caused by the ester linkages breaking and, possibly, forming ester radicals.

Table 2. Summary of functional groups of bio-plastics

Functional Group (Absorption Range)	Wavenumbers (cm^{-1})	
	Irradiated Bio-Plastic	Non-Irradiated Bio-Plastic
Alcohols (O-H stretch)	3293.22	3292.89
Alkanes (C-H stretch)	2932.10	2934.01
Carbonyl compounds (C=O stretch)	1645.61	1643.84
Esters (C-O stretch)	1014.73	1016.96

The oxygen radicals can easily combine with hydrogen radicals to form phenolic groups [29]. There is a shift towards a higher wavenumber from the peak at 1043.81 and 1472.92 cm^{-1} of non-irradiated and irradiated plastics, respectively (Figure 8). This shift is due to the C=O group formation. The peaks at range 718-730 cm^{-1} indicate the plastic's bending vibration bands. It has a medium intensity of alkyl halide groups. Table 2 compares the irradiated and non-irradiated bio-plastic functional groups. The wavenumber differences indicate a radiation effect on the bio-plastics. Table 3 compares the irradiated and non-irradiated conventional plastic functional groups. The most significant difference was reduced alcohol bond in the irradiated conventional plastic.

Table 3. Summary of functional groups of conventional plastics

Functional Group (Absorption Range)	Wavenumbers (cm^{-1})	
	Irradiated Plastic	Non-Irradiated Plastic
Alcohols (O-H stretch)	-	3342.20
Carboxylic acids (O-H stretch)	2915.45 & 2847.91	2915.27 & 2847.73
Alkanes (H-C-H bend)	1462.39	1462.34
Alkyl halides (C-Cl bend)	730.60 & 719.03	730.37 & 718.09

Conclusion

Results show that the irradiated bio-plastic had a higher absorption peak at wavelength 270 nm than non-irradiated bio-plastic, signifying that glycerol concentration was directly proportional to the absorption peak. Glycerol concentration was also directly proportional to the bio-plastic transparency [19], resulting in better UV absorption. Regarding surface morphology, both types of plastic showed degradation effects on their surfaces after they were irradiated. Several long gashes on the conventional plastic indicated that it absorbed the gamma radiation. Due to the gamma radiation's degradation effect, the irradiated bio-plastic surface was slightly darker than the non-irradiated bio-plastic surface. The gamma radiation caused an embrittlement effect. There were bubbles due to the problematic process of drying the bio-plastic. Irradiated bio-plastic samples displayed slightly decreased absorption spectra at 2932.10 and 1014.73 cm^{-1}. However, increased absorption peaks at 3293.22 and 1645.61 cm^{-1} indicated the presence of alcohols and carbonyl groups in the bio-plastics. For irradiated conventional plastic, there was a drastically reduced peak at 3342.20 cm^{-1}, indicating that the alcohol group was eliminated due to the plastic absorbing the gamma radiation. On the other hand, irradiation did not cause significant changes in other peaks.

Acknowledgement

The authors would like to thank the Faculty of Applied Sciences and Technology, Universiti Tun Hussien Onn Malaysia for the facilities provided that make the research possible.

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