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Physicochemical Characteristic of Polydiacetylene Prepared in Vesicles, Rod-like and Thin Film Upon Gamma Rays

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ABSTRACT

The purpose of this research was to look into the physicochemical properties of polydiacetylene (PDA) supramolecular assemblies made from 10,12-pentacosadiynoic acid after gamma irradiation. PDA can be oriented into any supramolecular form, including vesicle, rod-like and thin film structures. PDA dispersion was exposed to gamma radiation ranging from 0.5 - 5kGy. Pentacosa-10,12-diynoic acid is a diacetylene conjugate material with radiochromic properties. Its structure contains a pi-conjugated electron that efficiently reacts against radiation, transforming its optical properties in the visible region. The prepared PDA was characterised using UV-vis, FESEM, AFM, ^{1}H NMR and FTIR. The colour intensity of the PDA before and after gamma irradiation was measured and observed. The spectra were detected by UV-vis analysis at 649 nm. FESEM analysis revealed the morphology of vesicles as irregular shape particles and rod-like formations, whereas AFM analysis revealed the surface roughness of the formed PDA film. FTIR and ^{1}H NMR analysis confirmed the chemical structure of the PDA prepared in this study. As a result, the characteristics of each PDA supramolecule conformation and their response to gamma radiation were compared.

1. Introduction

Polydiacetylene (PDA) has develop alternative scientific and technological interest in recent years due to its unique chromatic properties. PDA in particular, has been proven to self-assemble into structured vesicles and films, forming an ene-yne conjugated framework that absorbs light in the visible region of the electromagnetic spectrum and appears intensely blue as an outcome [1]. PDA research has resulted in a large number of applications in a wide range of technological and scientific areas of study because of its superior optical, electrical and sensing properties [2-7]. It is broadly acknowledged that the abilities of materials are defined not just by the molecules themselves, but also by the configuration of the molecules [2,8]. PDA materials have just been produced in the form of liposomes [9,10], vesicles [1,11-16], Langmuir-Blodgett (LB) films [2,17,18], casting films [17,19-22] and nano-composites [7,17,23,24] to date. Between these, the LB technique is considered as one

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of the most impactful techniques for regulating molecular orientation and packing through two dimensions and enhancing photopolymerization [7,9,17,18,22,25-28]. Besides this, considering the responsivity of the absorption and fluorescent qualities to utilised stress [28], light [29] or heat [2], changes throughout chemical environment [6,30] and bio-analytes [1,24,31], various PDA materials have been studied in a variation of sensor applications.

However, since the majority of these chromatic responses are irreversible, their application in colorimetric sensing is limited. To achieve complete colorimetric reversibility for PDA materials during repetitive heating-cooling cycles, various functional groups such as hydrazide, urethane and azobenzene have been tried to introduce into PDA systems [32]. Recent studies have discovered that hydrogen-bonding between carboxylic acid head groups and relations among aromatic substituent in lipid assemblies were considered necessary for thermochromism reversibility in PDAs.

The author's goal in this paper was to present the characteristics of each PDA supramolecule conformation and compare their responses to gamma radiation. Each supramolecular assembly was synthesised into three different formations (vesicles, rod-like and thin film) and their responses to gamma rays were studied.

2. Methodology

2.1 Materials

The PDA liposome used in this study was produced from pentacosa-10,12-diynoic acid monomer (97 %). It was purchased in powder form from Sigma-Aldrich. Figure 1 illustrates the structure of its molecule, which has a molecular weight of 374.6 gm/mol. R&K Chemicals supplied laboratory grade chloroform for monomer dissolution. The ultra-pure water system was used to purify deionized water for the hydrolysation technique. In this study, three main types of PDA formation were synthesised: vesicles, rod-like and thin film.

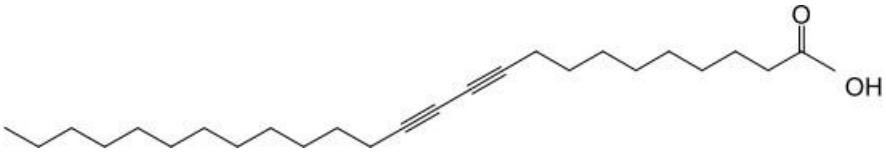

Fig. 1. Structure of 10,12-Pentacosadiynoic acid

2.1.1 Preparation of vesicles formation

The liposome was formed by diluting the PDA monomer in chloroform to produce a solution of approximately 2 mmol. A 20-min sonication process at room temperature enabled the solubility and dissolution rate. Afterwards, the organic solvent was extracted by air drying, resulting in a thin lipid film on the bottom of the round glass flask. For the secure volatilization of chloroform, the procedure was carried out using a fume hood. To quicken up the drying process, a hot water bath with temperatures ranging from 60 - 70°C had been used. The drying process have been further validated by heating it at 70°C at one hour.

Hydrolysis of the fermented lipid film generated a liposome with a total concentration of 1 mM. A sonication process was employed to assist and support with the hydration process and the formation of liposomes. When a relatively homogenous turbid solution formed at the end of the process, the liposome was fully prepared.

The sample mixture was divided up into two vials after being filtered through a 0.8 mm filtration. One vial was kept at 4°C, whereas the other was kept at 25°C. PDA dispersion was irradiated using

gamma irradiation for 0.5 - 5kGy at the Malaysian Nuclear Agency's Gamma cell Irradiation Facility. To avoid any possible exposure to surrounding lights all through storage, all samples were sealed in aluminium foil. Before any further analysis, the samples were kept on a regular basis for 38 days.

2.1.2 Preparation of rod-like formation

At room temperature, oxalyl chloride was added dropwise to a solution of 10,12-pentacosadiynoic acid (2.00 g, 5.34 mmol) in dichloromethane (20 mL) (2.03 g, 16.02 mmol). The resulting solution was then stirred at room temperature for approximately 30 mins. An additional hour of stirring was considered necessary since having to add a catalysed portion of dimethylformamide (at about 0.2 g) to the solution. After already being concentrated in order to evaporate, the residue has been redissolved in tetrahydrofuran (15 mL). The resulting solution was combined with 1.10 g of 4-aminobenzoic acid (8.01 mmol) and 2.16 g of triethylamine (TEA) (21.36 mmol) in tetrahydrofuran (15 mL). Upon stirring at room temperature for 24 hours, the mixture was diluted into water as well as extracted with ethyl acetate. Prior to getting dried with magnesium sulphate and filtered, the organic layer was cleaned three times with water in order to remove leftover 4-aminobenzoic acid. A vacuum was utilized to extract the solvent. The desired diacetylene monomer PDA-pBzA was managed to obtain like a white solid after recrystallization of the resulting solution from methyl alcohol and isopropyl alcohol. The technique was adapted from Lee *et al.*, [33] with minor modifications. The PDA-pBzA distribution was irradiated using gamma irradiation for 0.5 - 5kGy at the Malaysian Nuclear Agency's Gamma cell Irradiation Facility.

2.1.3 Preparation of thin film formation

PDA monomers were ready in a water subphase until being relocated to a pre-fabricated PVOH film material through the use of LB film. The monomers had been relocated numerous times to achieve the greatest optical shift on it by gamma rays. An Atomic Force Microscope was employed to observe the molecular packing. PDA dispersion was irradiated utilising gamma irradiation for 0.5 - 5kGy at the Malaysian Nuclear Agency's Gamma cell Irradiation Facility.

2.2 Chemical Structure and Functional Group Characterization

The molecular structure of all measurements was validated using Perkin-Elmer Fourier transformed infrared spectroscopy (FT-IR). This technology was effective for proving PDA polymerization and assessing the modification in PDA molecular structure induced by ionising radiation exposure throughout field testing. The FT-IR technology revealed the functional group display in the irradiated PDA. The ^{1}H NMR data were taken using a Bruker 400 spectrometer set to 400 MHz. The samples were immersed in $CdCl_3$ at a concentration of 10 mg/mL for the analysis methods.

2.3 Field Emission Scanning Electron and Atomic Force Microscopies

The PDA distribution was evaluated using Field Emission Scanning Electron Microscopy (FESEM-Gemini SEM 500) and an Atomic Force Microscope (AFM- XE-70 Park Systems). The prepared sample was attached to a holder before being loaded into the scanning chamber for FESEM analysis. Images were viewed and captured on camera at different magnification and configurations. AFM was a high-resolution scanning probe microscope with a resolving power in percentages of Angstrom that was

approximately 1000 times greater than a conventional optical microscope. AFM systems were used at room temperature to interpret the thin film formation of PDA.

2.4 Optical Density Measurement

The optical density of PDA liposome in responding to gamma irradiation was monitored using a SHIMADZU UV/Vis Spectrophotometer. The maximum absorption spectra were analysed and recorded at wavelengths in the range from 300 - 800 nm.

3. Results and Discussion

A dose response curve graph depending on UV-Vis characterization at 649 nm is presented in Figure 3. The absorption spectrum peak of PDA seemed to be 0.18 - 0.5 kGy and risen to 2.3 at 3kGy. Since diacetylene supramolecular polymers results in the modification of alkyne carbon atoms from sp hybrid to sp^2, the absorbance reading dramatically changes. The force in between densely packed polymer backbone, in contrast side, inhibits PDA from twisting and modifying the angular position of the PDA polymer backbone all through synthesis, creating tension in PDA and letting it to appear blue [32]. The finding demonstrates that the greater the gamma radiation dosages, the greater the maximum absorption peak. Figure 2 indicates how well the appearance of PDA vesicles improved with varying gamma ray radiation doses.

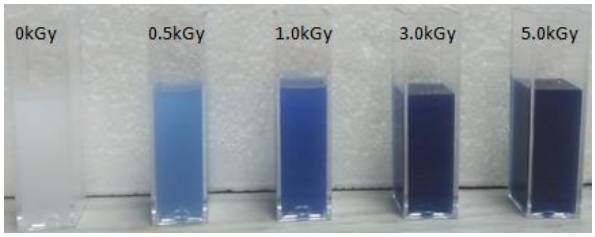

Fig. 2. PDA vesicle colour changes in response to gamma ray radiation doses

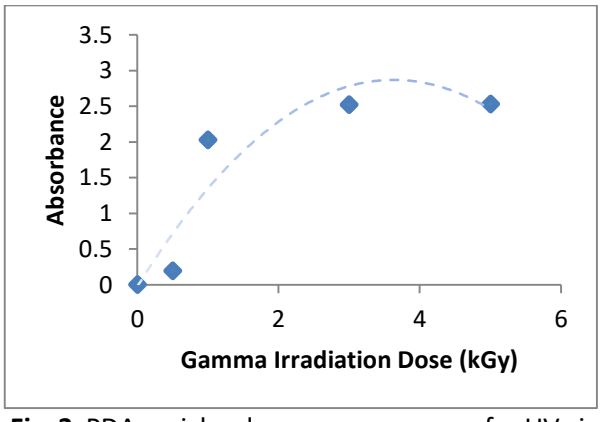

Fig. 3. PDA vesicles dose response curve for UV vis spectrum at 649 nm

Visible color changes were discovered once the PDA vesicles were subjected to gamma rays. The PDA seemed to be cloudy-white until being exposure to radiation. After being subjected to gamma radiation, the PDA started to turn dark blue. The intensity of the colour was proportional to the quantity of radiation absorbed (Figure 2). Due to this attribute, we could perhaps categorized the

PDA assemblies as radiation dose sensing devices and start investigating them further as potentially valuable gamma rays dosimeters. Focusing on the spectroscopy, the specular reflection was assessed at the wavelengths with the greatest decrease in optical properties, and it was closely linked to the absorption dose of gamma rays [19]. As a consequence, a dose response curve for calibration was generated (Figure 3). The PDA absorbed the most light at 650 nm and absorbed the least at 550 nm, providing it an observable blue colour to the naked human eye [34]. Within these studies, PDA films produced from diacetylene monomers with widely differing alkyl chain characteristics were deeply involved to polymer chains to determine the impact on thermochromism of the derived blue PDA [26].

To analyse and evaluate the photopolymerization of the prepared PDA, FT-IR analyses were done and the spectra (Figure 4) were obtained. PDA's solid-state FT-IR measurements reveal bands in the 3000 - 2800cm^{-1} range which then represent the $\nu(CH_3)_{asym}$, $(CH_2)_{asym}$ and $(CH_2)_{symm}$ stretches. The band at 1692 cm^{-1} is attributed a (C=O) stretch of the free acid. This IR peak depicts the interaction of PDA monomer units and PDA polymers. This peak's lowering frequency validates increasing interaction respectively PDA monomers upon force twisting as well as between PDA polymers upon polymerization process [25]. A CH_2 bending mode is represented by an intense band at 1467cm^{-1}, as are numerous different CH_2 wagging bands between 1350cm^{-1} and 1194cm^{-1}. Further to that, the sharp band at 722cm^{-1} is related to PDA's CH_2 hanging mode. The PDA spectrum displayed in the FT-IR configurations was reliable with PDA FT-IR spectra as in literature [4,17,35].

The FT-IR spectrum demonstrated that when subjected to heat, the cross-linked double and triple bonds inside the chain length segment reported experiencing a reversible conformational transition. As an outcome, the double and triple bonds were started changing from across all to partly-cis configurations as both the movement of the chain length improved throughout thermal treatment [36].

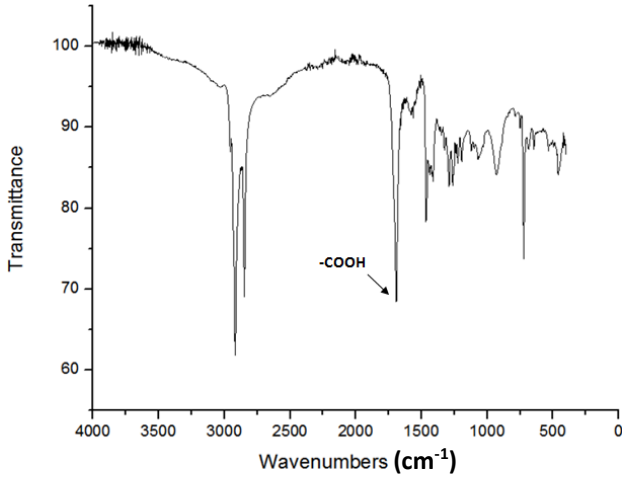

Fig. 4. FTIR spectra of PDA

The ^{1}H NMR spectroscopy of PDA prior to and after radiation exposure have been measured on a 400 MHz instrument with $CdCl_3$ as the eluent (Figure 5). In Figure 5 (a), the resonance frequency peak was observed at 7.50, 7.45 and 7.30 ppm are appointed towards the charged particles mostly on alkyl chain (C=O) well at two ends of both the CC bond. Such diffraction peak transition to the rising around 7.08-6.59 and 6.17 ppm in Figure 5 (b), which indicates the formation of the polymer backbone owing to improved polyene-backbone cross-linking [37]. The observed peaks were classified into the following categories: 0.85 (3H), 1.23-1.63 (2H), 2.26 (4H), 2.51 (2H), 6.68-6.75 (2H), 7.68 (1H) and 10.21 (1H) (1H). The methyl (-CH_3) group is recognized either by peak at 0.85 ppm; the several peaks among both 1.23 and 1.63 ppm were associated to the methylene group (-CH_2-) of the

hydrocarbon chain; and the peaks at 2.26 and 2.51 ppm are closely linked to the methylene group neighbouring to the alkyne (-C) group.

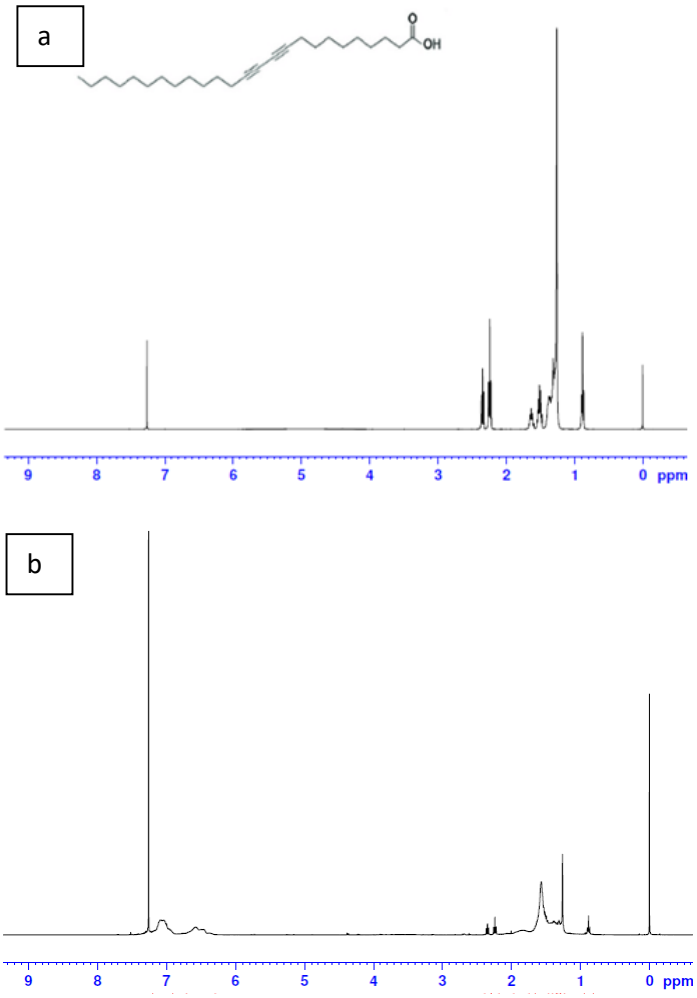

Fig. 5. ^{1}H NMR spectra of (a) Diacetylene monomer and (b) Irradiated PDA at 3kGy gamma radiation doses

FESEM image analysis methods were carried out to validate the research hypotheses. The FESEM image of PDA vesicles assemblies polymerized using distilled water, reveals a tightly packed wrapping framework to infrequent structure molecules (Figure 6 (a)). The incorporation of mixed water-THF solvents ended up causing macroscopic and microscopic conversion to sheet-like micelle cluster centers (Figure 6 (a)). Authors as well start noticing that raising the amount of THF seems to part ways the cluster centre. As a side effect, the mixed solvents assist self-assembly, that further tends to increase the production of PDA vesicles. This deeper understanding may be utilized to keep improving the chemically synthesized process of some of the other diacetylene monomers [22]. Figure 6 (b) clearly demonstrates the rod-like shapes of prepared PDA.

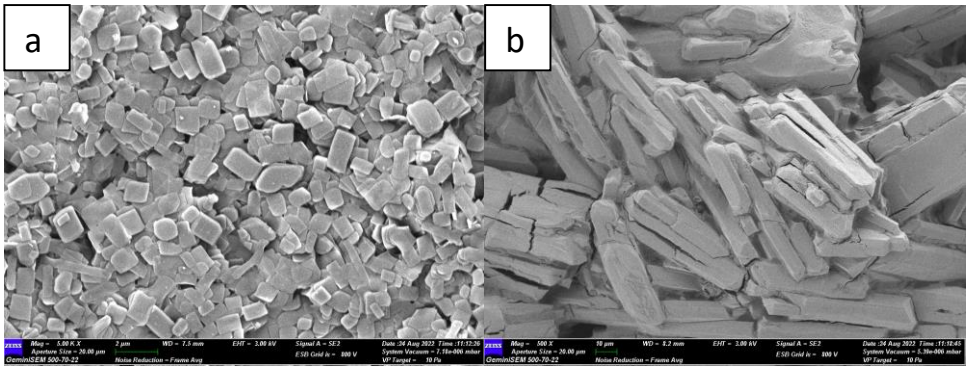

Fig. 6. FESEM images of (a) PDA vesicles formation and (b) PDA rod-like formation

AFM is widely recognized to grossly underestimate the surface area of biological materials due to all the symmetry of both the edge, where it generates illustration broadening [6]. The ability to manipulate the characteristics of PDA films at the nanometer size under geometrical and mechanical demands induced mostly on film by AFM [19] is unquestionably an exciting function of the authors' PDA films.

Figure 7 represents the topography of the PDA film prepared in the water subphase layer by layer method observed by AFM. The overall average distance and length from each single molecule have been analysed upon requiring repeated cross-sections of the individual protein images. Surface roughness of PDA film formed can be seen clearly from the image (Figure 7(a)). The goal of this study was to look into the impact on the qualities of PDA films. It was discovered that as the concentration of water subphase in the samples increased, enough that did the film thickness, but the band gap.

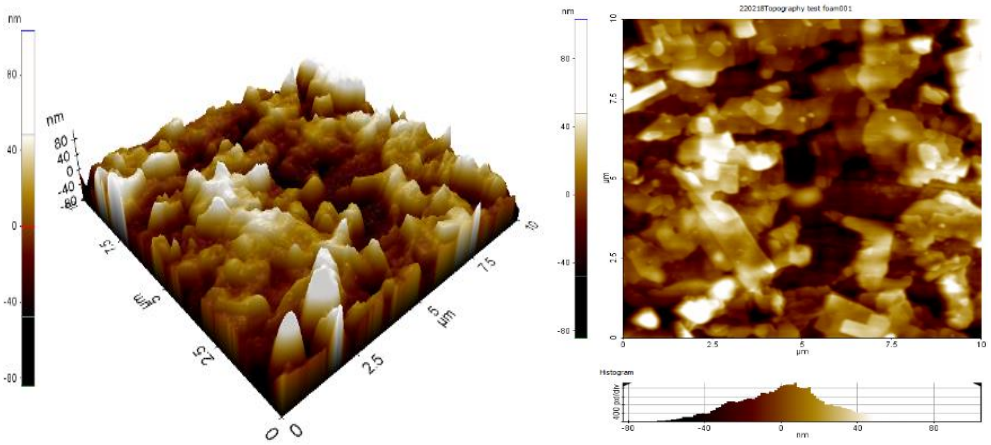

Fig. 7. AFM images of PDA thin film formation

4. Conclusions

Every supramolecular assembly was successfully synthesised and characterised using FTIR, ^{1}H NMR, FESEM, AFM and UV-Vis Spectroscopy. At the end of this study, the morphology of the PDA vesicles and rod-like were successfully viewed and produced, while the topography of the thin film formation was also achieved and analyzed. As a result, the physicochemical properties of each PDA supramolecule conformation were compared in terms of their response to gamma radiation.

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