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Assigning the Radiation Damage in Ultrahigh Molecular Weight Polyethylene by using Mueller Coherency Matrix

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ABSTRACT

In this work, we propose the use of Mueller matrix of ultra high molecular weight polyethylene (UHMWPE) in order to extract the optical properties (i.e. transmittance, retardance, diattenuation) of sample at optical spectral range of 630nm and irradiated with 0kGy, 30kGy, 65kGy, 100kGy. The deep understanding of UHMWPE polarization properties will pave the way for applications in optical and medical field. Mueller coherency matrix can be extracted from the usual Mueller matrix by means of the $SU(4) \leftrightarrow O^{+}(6)$ homomorphism. By means of this mathematical transformation, the Mueller coherency matrix is obtained from the Mueller matrix. From the Mueller coherency matrix, we extract the eigenvalues using these eigenvalues we calculate the entropy for the quantification of degradation in a sample. When radiation dose is increases in a sample, we notice that disorderedness increases. The result reported in this study suggested that the degradation / modification in UHMWPE can be probed with more sensitive physical quantity of light I.e. its polarization state.

Introduction

The demand for polymer and polymer-based products is growing every day because of many advantages. A useful polymer from the polyethylene (PE) family is Ultra high molecular weight polyethylene (UHMWPE) which possesses excellent properties including strong impact resistance, high wear resistance, good abrasion resistance, good chemical resistance, low friction, and biocompatibility[1]. The homo-polymer UHMWPE, which is made of ethylene monomers, is what gives it its extremely cross-linked structure. The structure also produces higher molecular weight polymers, including different morphologies like fibres, plates, or tubular structures depending on the type of medicinal grade UHMWPE is employed in a variety of medicinal applications. UHMWPE has been utilized extensively among the several materials that are accessible for medical implants, which also include metals, ceramics, and polymers[2]. The UHMWPE have wide applications including medical field, micro-electronic devices, insulation for electrical equipment, is used for the production of sports equipment and also used in the food processing industry. We radiate UHMWPE for the purpose of sterilization, crosslinking and surface modification. These processes generate free radicals, which result in the creation of inter-chain covalent bonds and crosslinking. This subsurface oxidation area can cause delamination, and failure frequently results from subsurface fracture initiation and propagation. The radiation-induced cross-linked UHMWPE has strong wear performance, although UHMWPE's mechanical performance is decreased by oxidation. The crystallinity and oxidation of UHMWPE are significantly influenced by the radiation dose and dose rate [3]. The limitation of UHMWPE is the production of free radicals during gamma irradiation, which are prone to react with oxygen and break polymeric chains when exposed for longer periods of time. The formation of free radicals is typically prevented by bolstering antioxidants such as vitamin E[2]. High-dose radiation (gamma radiation and electron e-beam radiation) was used to create radiation crosslinking in UHMWPE, which decreased the polymer's ductility, increased its hardness, and enhanced its resistance to wear [4].

Much effort has been made to quantify the degradation using FTIR, XRD and ultraviolet - visible spectroscopy[5]. Although these approaches are now in use but efforts are still in progress to have more delicate, precise and straightforward methods for quantification of degradation. According to the best of our knowledge, there are just a few studies in the literature that use polarized light to characterize UHMWPE. Both of the reported studies are carried out by[6] with Mueller matrix polarimetric technique for the characterization of UHMWPE[6]. The observed values of absorbance of polarized light over the wavelength range of 400 nm-800 nm, transmittance, retardance, polarizance, birefringence, and linear and circular polarization are included in the presented results. The results indicates that due to UHMWPE's highly scattering nature, linear retardance was found to be greater than circular. It was clear that circular polarization dominated linear polarization. It was discovered that the optical depth and scattering increased with wavelength. Depolarization index (DI) and degree of polarization (DoP) both showed a considerable decline. Overall, they came to the conclusion that the incoming photons are rotated by the high refractive index, low mean free path, and tight bonding within the UHMWPE matrix, leading to the aforementioned observations in terms of the polarization properties of UHMWPE [5]. Additionally, the results in terms of linear and circular retardance demonstrated the compatibility, strength, and compactness of UHMWPE as a material employed in high strength orthopedic and industrial applications. Unfortunately, both investigations attempted to characterize the UHMWPE while dissolving it in Xylene; as a result, the results are problematic to our knowledge and may not entirely belong to UHMWPE. Nevertheless, both studies give the framework for using polarized light spectroscopy to characterize the material. No more experimental evidence is also offered to support these findings. Additionally, nobody has yet paid attention to the consequences of radiation-induced alterations inside UHMWPE on the aforementioned prospects or the radiated/modified UHMWPE.

In this work our aim is to investigate the effect of radiation modification on the entropy and correlate the entropy of radiated sample of UHMWPE with irradiated sample. In current research UHMWPE samples were exposed to e-beam ionizing radiation of 30, 65, and 100 KGy. In this work a complex Mueller coherency matrix can be extracted from the usual Mueller matrix by means of the $SU(4) \leftrightarrow O^{+}(6)$ homomorphism. By means of this mathematical transformation, the Mueller coherency matrix is obtained from the Mueller matrix. Additional information can be obtained from the eigenvalue analysis then we calculate the entropy.

Methodology

Materials, sample preparation and irradiation

Laboratory-grade UHMWPE resin powder with an average molecular weight of 3-6 million g/mol purchased from Sigma-Aldrich and pressed under steady pressure of 200 bar while holding for 12 to 15 minutes at different temperature of 150 °C, 160 °C and 190 °C, respectively. Under the same pressure as before, the pressed samples were then cooled to room temperature i.e. 25 °C. After cooling, the surfaces of all samples were cleaned with acetone. The samples were separated into 4 groups after measuring thickness and labeled as PE-0, PE-30, PE-65, PE-100. The thickness of each sample was determined with the help of IR vibration band at 2020 cm^{-1} (an internal PE standard that is still impacted by changes to the PE matrix's structure). The thickness of each sample was roughly 500 µm. One group that was labeled as control (un-irradiated) was placed at shelf and others were sent to Korean Atomic Energy Agency for e-beam irradiation where they were exposed to radiation in open air at ambient temperature for total dose values of 30 kGy, 65 kGy, and 100 kGy, respectively. The main reason for selecting the aforementioned irradiation dose values was the saturation in PE cross linking at or above 100 kGy. The schematic diagram of the experimental apparatus that was used for the measurements is shown in Fig. 1. This was basically a computerized Mueller matrix polarimeter named as Axo-Scan TM and manufactured by Axometric Inc. Readers are directed to the literature for more information on the precise operation of this automated spectrum polarimeter [7]. The setup shown in Fig. 1 is used for polarization characteristics measurements along with refractive index of each sample. FTIR tests of all samples were performed in transmittance mode while using Nicolet 6700 FTIR spectrophotometer made by Thermo Electron Corporation, Waltham, MA, USA in the spectral range from 400 to 4000 cm^{-1} with spectral resolution of 4 cm^{-1}. In order to lower the signal-to-noise ratio, each sample was scanned three times by gathering 16/32 scans per spectrum, and then averaged. Furthermore, for determination of degree of cross-linking gel contents measurements were made in accordance ASTM-D2765 standard. Briefly, Soxhelt apparatus was used, and samples were extracted for 36 h in boiling Xylene. After being cleaned with acetone, these extracted samples were then dried at 80 °C.

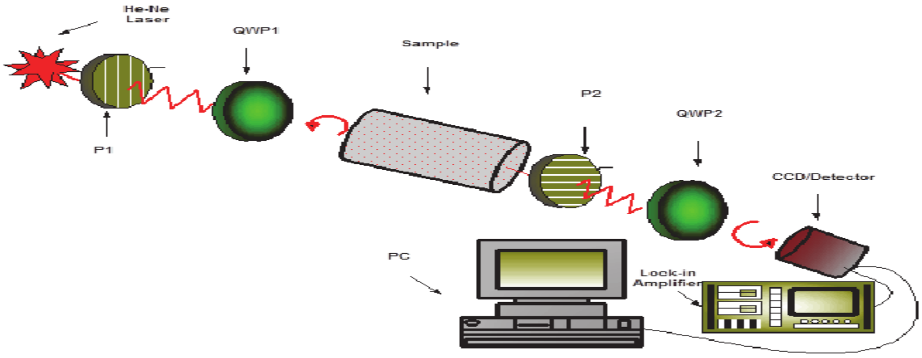

Fig.1. Experimental set up for measurements of Mueller matrix elements.

Muller matrix

The Muller matrix, which is a 4x4 matrix, shows how polarized light that is incident on a surface change to polarized light that is emitted from it. In plainer language, it describes how light that had a particular beginning polarization is changed as a result of interaction with an optical system. Mueller's matrix operates on the polarization's incoming Stokes vector expression. The Stokes vector is a real valued four element vector.

$$S_{output} = MS_{input}$$

$$S = \begin{bmatrix} I \\ Q \\ U \\ V \end{bmatrix}$$

The Stokes vector, which comprises of measurements for the light's intensity, I, its polarization at 0°/90°, Q, its polarization at 45°/135°, U, and its circular polarization at left and right, V. The output Stokes vector is created by transforming the input Stokes vector. Using the Mueller matrix as a vector. As a four element vector is affected by the Mueller matrix to produce a four. The Mueller matrix must have a size of four-by-four elements.

$$\begin{bmatrix} I_{output} \\ Q_{output} \\ U_{output} \\ V_{output} \end{bmatrix} = \begin{bmatrix} M_{00} & M_{01} & M_{02} & M_{03} \\ M_{10} & M_{11} & M_{12} & M_{13} \\ M_{20} & M_{21} & M_{22} & M_{23} \\ M_{30} & M_{31} & M_{32} & M_{33} \end{bmatrix} \begin{bmatrix} I_{input} \\ Q_{input} \\ U_{input} \\ V_{input} \end{bmatrix}$$

The meaning of a few Mueller matrix components can be deduced by looking at the whole Mueller matrix expression. For instance, the M_{00} element can be thought of as the simple transmittance because it scales the input intensity to the output intensity. The linear polarization at 0°/90° is scaled to the output intensity by the M_{01} element. M_{01} can be understood as the linear extinction at 0°/90° as a result. Extinction refers to the ratio of light intensity input to output in this context. As there are several phrases that are similar to one another, such as transmission/reflectance, absorbance, extinction, etc., the language for polarization may use some clarification [8].

Light transmission is the ratio of light coming in to coming out. Absorption is the linear attenuation in transmission multiplied by the propagation distance. Transmittance is thus the extrinsic measurement of the ratio of light intensity in this context. While absorption aims to gauge a material's transmission-related linear attenuation as an intrinsic feature [8].

Intrinsic property	Extrinsic property
Linear birefringence (LB)	Linear retardance (LR)
Circular birefringence (CB)	Circular retardance (CR)
Linear dichroism (LD)	Linear extinction (LE)
Circular dichroism (CD)	Circular extinction (CE)
Absorption (A)	Transmission (T)

By integrating throughout the distance of propagation through the material, the intrinsic property in each of the aforementioned ideal cases can be connected to the extrinsic property.

$$LE = -\ln\left(\int_0^l LD(Z)dz\right)$$

The circular retardance can be expressed in the following equation.

$$CR \sim \frac{M_{12} - M_{21}}{2}$$

The total retardance scales both the measurable linear and circular retardance by the sinc function.

$$TR = \sqrt{LB^2 + LB'^2 + CB^2}$$

$$CB_{measable} \sim CBsinc(TR)$$

$$LB_{measable} \sim LBsinc(TR)$$

Mueller coherency matrix method

The Mueller matrix has 16 elements, and they contain all the polarization dependent properties. There are techniques that take measurements of Mueller matrix but the information obtained is limited because the direct interpretation of the Mueller matrix element is difficult to relate. The Mueller matrix is transformed into Mueller coherency matrix by using $SU(4) \leftrightarrow O^+(6)$ homomorphism [9]. Then from this matrix we extract the useful information, established the relationship between real and complex sets. Then we extract the eigenvalues and then calculate the entropy.

The eigenvalue analysis can reveal the Mueller Coherency matrix's enormous potential [10]. Its decomposition allows for the extraction of up to four non-zero eigenvalues λ_i and the accompanying eigenvectors or target vectors C_i [11].

$$C_{4\times4} = \lambda_1 C_1 + \lambda_2 C_2 + \lambda_3 C_3 + \lambda_4 C_4$$

Result and discussion

A Mueller matrix is a 4 × 4 matrix providing a complete and accurate description of the polarizing and depolarizing characteristics of a sample for light with any input polarization given by a so-called Stokes vector. The Mueller matrix elements M_{ij} (i, j = 1, …, 4) are normalized to the total transmittance, i.e., element M11, according to $m_{ij} = M_{ij}/M11$. To better visualize retardance features, spectral data are presented as a function of photon energy E related to wavelength λ as $E = hc/\lambda$, where h is Planck's constant and c is the vacuum speed of light. The experimental wavelength range for the data in this work is 630nm and corresponds to the energy range of 0.73–3.54 eV.

$$\text{P-0} = \begin{bmatrix} 1 & -0.003 & -0.005 & 0.000367 \\ -0.004 & 0.889 & 0.035 & -0.134 \\ -0.004 & 0.038 & 0.844 & 0.277 \\ -9.200 & 0.138 & -0.282 & 0.753 \end{bmatrix}$$

Now, discussed the optical and polarization properties of the sample UHMWPE in visible spectral range i.e. 630nm, firstly discussed the properties for pure sample the transmittance is 0.294666 and the linear retardance with negative magnitude is 21.55793 linear retardance with positive magnitude is 37.72589 circular retardance with negative magnitude is -0.10055, circular retardance with positive magnitude is -0.1759, linear diattenuation is 0.005662, circular diattenuation is 0.000366.

$$\text{P-30} = \begin{bmatrix} 1 & 0.056 & 0.001 & 0.000169 \\ 0.056 & 0.893 & 0.028 & -0.037 \\ 0.001 & 0.024 & 0.429 & 0.593 \\ 0.004 & 0.054 & -0.603 & 0.357 \end{bmatrix}$$

Now discussed all the optical properties for a sample when radiated with radiation dose of 30 kGy, the transmittance is 0.159946 and the linear retardance with negative magnitude is 56.75923, linear retardance with positive magnitude is 99.32866 circular retardance with negative magnitude is 0.401198, circular retardance with positive magnitude is 0.702096, linear diattenuation is 0.055839, circular diattenuation is 0.000175.

$$\text{P-65} = \begin{bmatrix} 1 & -0.00038 & 0.003 & -1.800 \\ -0.00099 & 0.679 & -0.021 & -0.158 \\ 0.001 & -0.005 & 0.633 & 0.251 \\ -0.005 & -0.145 & -0.267 & 0.365 \end{bmatrix}$$

when the sample is radiated with radiation dose of 65 kGy then the transmittance is 0.212424 and the linear retardance with negative magnitude is 30.50922, linear retardance with positive magnitude is 53.39114, circular retardance with negative magnitude is -0.9963, circular retardance with positive magnitude is -1.74353, linear diattenuation is 0.003421, circular diattenuation is -0.000017.

$$\text{P-100} = \begin{bmatrix} 1 & -0.048 & 0.020 & -0.009 \\ -0.045 & 0.627 & -0.267 & -0.063 \\ 0.023 & -0.271 & -0.087 & -0.303 \\ 0.008 & 0.061 & 0.303 & -0.272 \end{bmatrix}$$

when the sample is radiated with radiation dose of 100 kGy then the transmittance is 0.179471 and the linear retardance with negative magnitude is 126.125, linear retardance with positive magnitude is 220.7188, circular retardance with negative magnitude is 0.193081, circular retardance with positive magnitude is 0.337891, linear diattenuation is 0.052709, circular diattenuation is -0.00903.

Sample	λ1	λ2	λ3	λ4	Entropy
P-0	1.8021	0.1083	0.0797	0.0089	0.175458
P-30	1.6701	0.3068	0.1403	0.0905	0.343723
P-65	1.42	0.2302	0.3231	0.0257	0.365841
P-100	5.3201	0.4582	1.166	5.6959	0.461888

For the entropy we convert mueller matrix into mueller coherency matrix by using $SU(4) \leftrightarrow O^{+}(6)$ homomorphism. Then from this matrix we extract the useful information, established the relationship between real and complex sets. Then we extract the eigenvalues and then calculate the entropy. The entropy-factor H is calculated by using formula

$$H = -\sum x_i \, log_4(x_i)$$

The entropy for the pure sample is 0.175458 when radiated with 30 kGy the entropy is 0.343723 for the radiation dose of 65 kGy the entropy is 0.365841 when radiated with dose of 100 kGy the entropy is 0.461888. when radiation dose increases the disorderness in a sample also increases.

Conclusion

The application of structure preserving transformation to characterize the sample of UHMWPE provide an improved tool in the analysis of optical properties of sample. We obtained the data of Mueller matrices of UHMWPE at different radiation doses and discussed all the optical properties of sample. The introduction of polarization analysis based on homomorphism and Mueller Coherency matrix. From these matrices extract the eigen values using these eigen values we calculate the entropy factor. We notice the when we increase the dose rate from 0kGy to 100kGy the disorderedness in a sample increase.

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