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Material properties

Mechanical response of high density polyethylene to gamma radiation from a Cobalt-60 irradiator

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ABSTRACT

The response of High Density Polyethylene (HDPE) to gamma irradiation is important for a variety of applications, from the durability of hip replacements after gamma sterilization to the degradation of power cable insulation that guides the licensing and regulation of nuclear power plants. HDPE samples are irradiated with up to 58.8 kGy from a Cobalt-60 gamma irradiator, and mechanical properties are examined using models assuming exponential behavior. Increasing the radiation dose led to increases to the ultimate strength and the Rockwell hardness with a corresponding reduction in the maximum elongation at ultimate strength, supporting the hypothesis that the samples increased their strength and brittleness.

1. Introduction

Irradiated polymers are of interest for a variety of reasons. As a large component of all plastic waste due to its low production cost [1,2], they have a role in sustainability and recycling [3]. Medical applications include the response of High Density Polyethylene (HDPE) in hip replacements to gamma sterilization [4,5], and the accelerated aging of HDPE-based nano-composites that replace bone [6].

Degradation mechanisms in polymers due to gamma irradiation also impact the reliability of instrumentation and power cable systems necessary to maintain safe operation and extend the lifetimes of Light Water Reactors (LWRs) for nuclear power. In support of the Light Water Reactor Sustainability Program [7] for the United States Department of Energy (DOE) and the Nuclear Regulatory Commission (NRC), a recent Extended Materials and Degradation Assessment [8] addressed the research needs for the effects of gamma radiation with other stressors on polymers like HDPE that may be used in power cable insulation.

The present study examines the effects of gamma radiation from a Cobalt-60 irradiator on the mechanical properties of HDPE.

2. Theory

On interacting with materials like HDPE, gamma radiation can produce free radicals or chemical species with unpaired electrons which can be extremely reactive, resulting in complex structures and irreversibly broken covalent bonds [9]. Free radical reactions in plastics cause crosslinking between polymer chains, chain scission and oxidations of the carbon chains [10]. When crosslinking occurs, new chemical bonds between the atoms of adjacent polymer strands may be formed. These bonds can alter a polymer's mechanical and chemical properties [11,12].

For example, the modulus of elasticity and micro-hardness properties decrease as crosslinking increases in polyvinyl alcohol exposed up to 30 kiloGray (kGy) [13]. The hardness of HDPE has been shown to increase with the gamma ray dose, the annealing time and the annealing temperature [14]. Increasing the temperature tends to decrease of the modulus of elasticity, yield stress and maximum stress of HDPE [15]. The reduction of degradation from adding antioxidants into the plastics indicates that oxidation is a factor even at low energy radiation exposure [16]. Volatiles from polyethylene can be detected after gamma irradiation [17].

Tensile strength properties that characterize a response to irradiation are acquired from a stress versus strain curve. Initially, this curve is linear due to the reversible nature of the elastic deformation and the slope is the modulus of elasticity. The maximum stress in this region is the yield stress. After the yield

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stress point has been passed, the material undergoes plastic deformation that is irreversible. The slope of the stress vs. strain graph will continue to decrease until it becomes negative or the material fractures, defining failure [18].

The integral of the stress versus strain during plastic deformation is the strain energy absorbed prior to fracture. The less energy absorbed prior to fracture, the more brittle the material [19].

Hardness is also a measure of a material's resistance to plastic deformation. This can be measured as a value of Rockwell hardness number, which is the permanent increase in depth subtracted from a constant that depends on the indenter used [20]. The flexural testing or 3-point bending is also used to provide values of the modulus of elasticity in bending, flexural stress, flexural strain and flexural stress vs. strain response of materials. By measuring changes in these properties, the impact of the gamma irradiation can be determined.

3. Materials

High Density Polyethylene sheets with 1.27 cm thickness and 122 cm × 122 cm area were purchased from US Plastics with a density of 0.96 g/cm^3. Test samples were cut to specifications [21–23], using a water jet cutting technique by Brookings Machine and Engineering in Brookings, South Dakota.

The effects of gamma irradiation are often correlated with the total radiation dose, which is equivalent to the energy absorbed per unit mass of the sample. Six samples were irradiated by a Cobalt-60 gamma-ray source operated by the 3M Corporation for each dose.

The radioactive decay of Cobalt-60 results in two gamma rays with energies of 1173.2 keV and 1332.5 keV (1000 keV = 1 MeV = 1.602 E−13 J). These energies are large enough to break chemical bonds but too small to produce artificial radioactivity.

4. Experimental

The doses provided for this study were measured by the 3M Corporation to be 0 kGy, 14.45 kGy, 31.15 kGy, 43.2 kGy, and 58.8 kGy, where 1 Gy = 1 J per kilogram and 1 kGy = 1 kGy.

The mechanical testing and evaluation was conducted in the Material Evaluation and Testing Laboratory (METLAB) at South Dakota State University. The tensile strength of the materials was evaluated using a MTS 370 Landmark machine. Samples were stretched at a rate of 5 mm/min until they failed. The applied force was measured while a 634.31F/24 axial-multiple gage length extensometer with 20 mm gage length was used to record strain throughout the test.

Six specimens for each dose were tested for better statistical significance. For a given mechanical property, the average mechanical response for specimens with a common radiation dose has been plotted.

Sample outputs from MATLAB for stress versus strain graphs are presented in Fig. 1. The picture on the left shows a break failure and the one on the right shows a necking failure. It is noted that a decrease in ultimate strength in Fig. 1 occurred after a rupture or failure: After plastic deformation, some slip in the clamps occurred.

The maximum elongation was obtained from strain measurements and the geometry of the extensometer.

Three-point bending tests were conducted using a customized bending fixture that was made in METLAB. The crosshead motion was 5.4 mm/min with a strain rate of 0.01 mm/mm/min. A typical flexural stress versus strain response is shown in Fig. 2. The support span L (mm or in) was calculated as $L = \sqrt{6Rd/Z}$, where d is the thickness (mm), Z is the strain rate and R is the cross head motion in mm per minute. L was determined to be 203.2 mm and d to be 12.7 mm.

The corresponding applied forces along with strain values were then used to calculate the flexural modulus of elasticity $E = 2bd^2\varepsilon/(3PL)$. Here, E is the flexural modulus of elasticity (Pascal), and P and ε are the applying force and strain values, respectively. The width of the specimen, b, was 19.05 mm.

A Wilson 2000 Rockwell Macro Hardness Tester was used to evaluate indentation properties. After applying a minor load of 10 kg to a sample, a major load of 60 kg was applied for 15 s, whereupon it reverted to the minor load. The indenter was a 1.27 cm steel ball and R scale was used for Rockwell hardness. The procedure was performed for six samples on both faces at each dose for statistical significance.

5. Results

The results of the tensile strength evaluations were compiled into MATLAB and a stress versus strain graphs like those in Fig. 1 were created for each trial. The properties of each sample were calculated with a hysteresis code and plotted in the following figures.

Exponentials were the primary function used to build an identification model of the response of the polyethylene to irradiation in mechanical testing [24]. While other functions may fit the data, the exponential based models were preferred for their future predictive value. Many properties associated with radiation effects, such as the attenuation of gamma rays through matter, tend to use exponentials [25]. Furthermore, exponential models are commonly used in reliability engineering [26,27].

Functions used to model the data were continuous and incorporated a minimal number of parameters necessary in a continuous to produce a good fit. Said parameters were optimized to produce a reduced chi-square of 1.0 per degree of freedom. Additionally, the sum of the weighted data scatter above the fit curve (positive) and below the fit curve (negative) was constrained to be zero.

5.1. Ultimate strength

One trial associated with the 58.8 kGy dose in Fig. 3 was eliminated by a 99% confidence Q-test. The remaining data were fit by a monotonically increasing function that asymptotically approaches a maximum value. This is consistent with the polyethylene getting stronger as the radiation dose increases. Models using a power law or a natural logarithm with the same number of parameters were also able to satisfactorily fit the data and replicate the monotonic increase.

5.2. Modulus of elasticity (MOE)

The MOE is an effective spring constant that describes the stiffness of the spring-like behavior of the material. The function $y = A{\bullet}\exp(-B{\bullet}x){\bullet}x + C$ generated the best fit to the data.

This model predicts a maximum in the MOE around 7 kGy in Fig. 4. Additional data between 0 and 10 kGy and greater than 60 kGy would improve the model.

5.3. Yield stress

It can be noted that the yield stress in Fig. 5 appears to be negatively correlated to the MOE. In fact, the correlation coefficient between the two data sets was −0.93. Thus a similar-shaped function was attempted for the fit, but a fourth parameter was introduced to shift the function slightly and satisfy our fitting requirements.