VARIATION OF NEUTRON MODERATING POWER ON HDPE BY GAMMA RADIATION

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High density polyethylene (HDPE) is degraded due to a radiation-induced oxidation when it is used as a neutron moderator in a neutron counter for a nuclear material accounting of spent fuels. The HDPE exposed to the gamma-ray emitted from the fission products in a spent nuclear fuel results in a radiation-induced degradation which changes its original molecular structure to others. So a neutron moderating power variation of HDPE, irradiated by a gamma radiation, was investigated in this work. Five HDPE moderator structures were exposed to the gamma radiation emitted from a ^{60}Co source to a level of 10^5-10^9 rad to compare their post-irradiation properties. As a result of the neutron measurement test with 5 irradiated HDPE structures and a neutron measuring system, it was confirmed that the neutron moderating power for the 10^5 rad irradiated HDPE moderator revealed the largest decrease when the un-irradiated pure one was used as a reference. It implies that a neutron moderating power variation of HDPE is not directly proportional to the integrated gamma dose rate. To clarify the cause of these changes, some techniques such as a FTIR, an element analysis and a densitometry were employed. As a result of these analyses, it was confirmed that the molecular structure of the gamma irradiated HDPEs had partially changed to others, and the contents of hydrogen and oxygen had varied during the process of a radiation-induced degradation. The mechanism of these changes cannot be explained in detail at present, and thus need further study.

Keywords : Neutron Moderator, Neutron Moderating Power, Gamma Radiation, Radiation-induced Degradation, High Density Polyethylene (HDPE)

1. INTRODUCTION

The molecular structure and macroscopic characteristics of polymer materials and electronic components are affected by radiation in nuclear power plants, radiochemical plants, radioisotope handling facilities, and so on. The polymers used in these facilities are exposed to an ionizing radiation leading to a radiation-induced degradation. So far, many studies on a radiation degradation of polymer materials have focused on radiation-induced oxidation phenomena, because most polymers are used in air [1]. The radiation-induced free radicals in a polymer react with the oxygen in air, and their degradation proceeds via a similar process in other environments, such as an elevated temperature, ultra-violet light, a mechanical stress, a chemical initiation involving radicals, etc. It is known that oxidation reactions induced by free radicals progress as follows; 1) initiation of free radicals by absorbing radiation energy, 2) their propagation, and chain branching, which leads to a scission and cross-linking, and ends with 3) a termination stage at which all the free radicals are completely consumed [2,3].

HDPE is employed in the moderator of a neutron counter for a specific nuclear material accounting during a spent fuel processing in laboratories and radiochemical plants. Radiation effects of an HDPE moderator are not serious in the case of a nuclear material accounting without gamma emitting fission products in the final products such as a wet processing. But if gamma emitting fission products are included in a nuclear material such as during a dry processing, the HDPE moderator is affected by the gamma radiation emitting from the fission products in that material.

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Then, the neutron moderating power of the HDPE can be affected due to a molecular structural change, a swelling, a radiation-induced oxidation, etc. Therefore, it is necessary to confirm this variation of the neutron moderating power of HDPE according to a radiation exposure for a correct analysis of the data acquired from a neutron measurement of nuclear materials.

2. THEORETICAL BACKGROUND

The effectiveness of a material as a neutron moderator is measured by means of the product $\xi\sum_S$, known as a moderating power (MP), which takes into account the frequency of the scattering collisions and the loss of energy per a collision.

$$ModeratingPower = \xi\Sigma_S \quad (1)$$

where, ξ(=ln E_{Before} - ln E_{After}) is the average logarithmic energy decrement per scattering collision and $\sum_S$(=$N\sigma_S$) is the macroscopic scattering cross-section. When a moderator is composed of n elements, the moderating power of a compound is given by

$$\xi\Sigma_S = \frac{\rho N_A}{M}\sum_{i=1}^{n} n_i \sigma_{si} \xi_i \quad (2)$$

where ρ is the density of the moderator, N_A is the Avogadro's number, M is the molecular weight, n_i is the number of atoms of element i in one molecule, σ_{si} is the microscopic scattering cross-section for element i and ξ_i is the logarithmic energy decrement for element i [4].

HDPE is widely used as a moderator for moderating the neutrons in a neutron detection system for a specific nuclear material accounting due to its high moderating power, material price, easy formation, and so on. But if HDPE is employed in a radiation environment, the neutron moderating power varies because of a chemical structural change and a new element production or evaporation due to a radiation-induced degradation.

3. EXPERIMENTAL

3.1 HDPE Moderator Structure Preparation

Six HDPE moderator structures were manufactured with the same dimensions for the same measurement conditions, which had five holes for inserting the neutron detectors and neutron source as shown in Fig.1. The hole in the center of the upper cross section is for the neutron source insertion, and the bottom end of the hole is placed in the center of the axial direction by considering the detector active length. The other four holes are for inserting the ^{3}He neutron detectors whose diameter and length are 2.54 cm and 31cm, respectively.

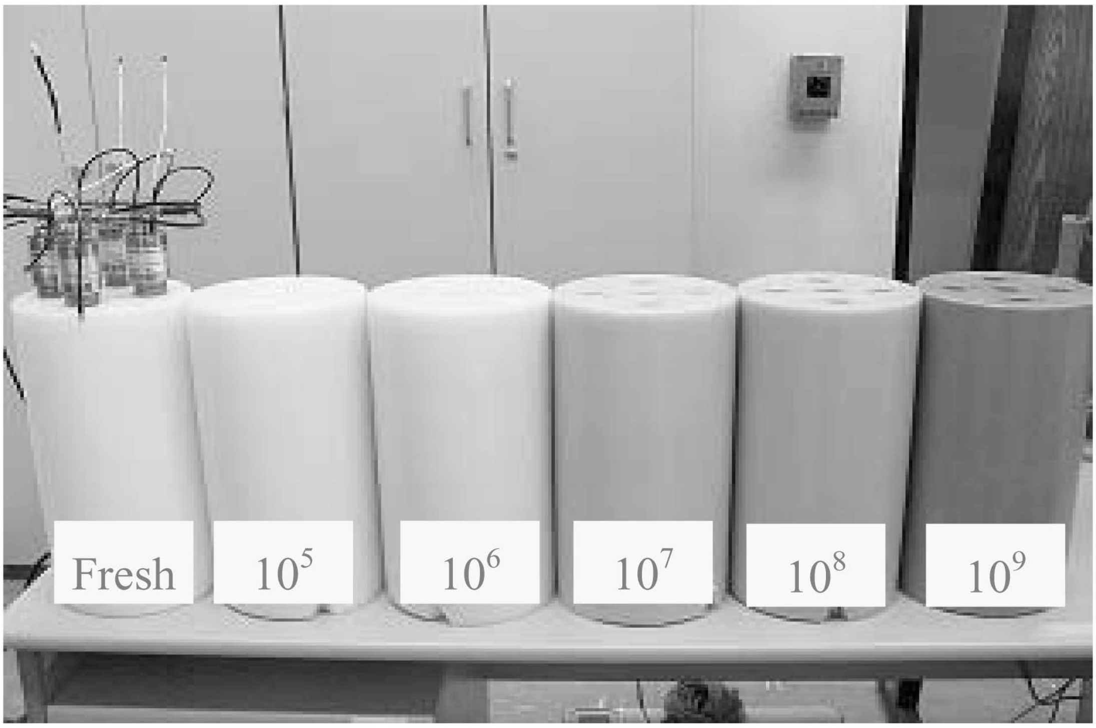

Fig. 1. Photograph of the HDPE neutron moderators irradiated by gamma-ray.

3.2 Gamma Irradiation

HDPE neutron moderator structures were exposed to a gamma radiation in KAERI's (Korea Atomic Energy Research Institute) high level irradiation facility. The five HDPE structures were irradiated in the range of 10^5 - 10^9 rad by the gamma ray emitted from ^{60}Co with an energy of 1,173/1,332 keV. One HDPE structure was not irradiated, so we could use it as a reference for comparison purposes. The structures were rotated on the sample holder in the high level irradiation facility for a homogeneous exposure because of their voluminous structure.

3.3 Neutron Measurement

A neutron moderating power is directly proportional to the number of neutrons detected in a neutron counting system. And it mainly depends on the neutron moderator material used in the detection system. As a neutron counting rate, the singles rate means the total number of neutrons detected from the (α, n) reaction and spontaneous fission neutrons, and the doubles rate represents the number of neutrons detected from ^{235}U and ^{239}Pu of the total neutrons detected in the detection system. The shift register plays an important role in distinguishing the neutrons detected.

The neutron moderating power tests on the fresh and irradiated HDPE structures were carried out by ^3He neutron detectors (Reuter Stokes, 1 inch in diameter) and electronic equipment such as a PSR (Portable Shift Resister manufactured by Aquila Technologies Group, Inc), a preamplifier/amplifier unit (Precision Data Technology, Inc : PDT) and the INCC (IAEA Neutron Coincidence Counter) software developed by LANL (Los Alamos National Laboratory). The neutron measurement tests were performed by using a ^{252}Cf neutron source, where the measurement time and number were 120 s and 10 cycles respectively, and the high voltage applied to the ^3He detectors was 1,700V and the Vcc (current circuit voltage) applied to the preamplifier/amplifier unit was 5V. The tests were conducted in the same conditions for each structure.

3.4 Element Content

Element Analyzer of CE Instruments (Carlo Erba)/Model EA1110 CHNS/O was employed to analyze the elemental contents of carbon, hydrogen, nitrogen, sulfur and oxygen in the six samples taken from the fresh (un-irradiated) and irradiated HDPE moderator structures.

3.5 FTIR Spectroscopy

Fourier transform infrared spectroscopy (FTIR) has become one of the favored techniques, from among a nuclear magnetic resonance spectroscopy (NMR), an electron spin resonance spectroscopy (ESR), a gel permeation chromatography (GPC), a swelling analysis, and so on, for analyzing the effects of an ionizing radiation on polymer materials. In this test, a broadband infrared source was transmitted through a thin section on to the fresh HDPE and irradiated HDPEs whose thicknesses were 200-300 μm. Chemical species were activated by light at a specific frequency, so they would absorb the energy at this frequency.

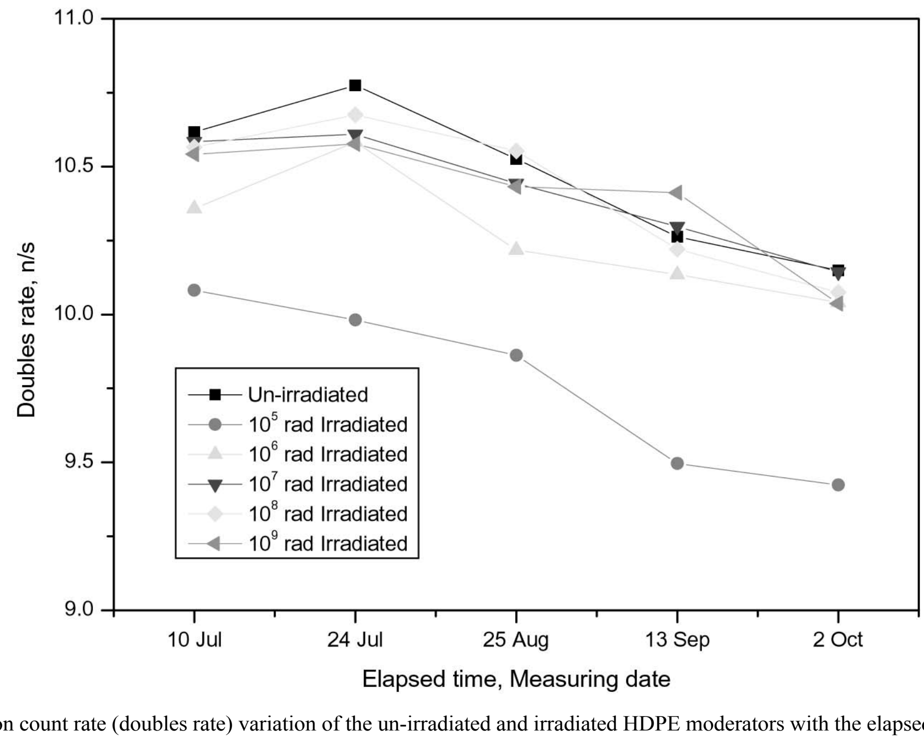

Fig. 2. Neutron count rate (doubles rate) variation of the un-irradiated and irradiated HDPE moderators with the elapsed time.

4. RESULTS AND DISCUSSION

4.1 Neutron Moderating Power by an Experimental Measurement

Neutron measurement tests for an observation of the post irradiation effects on an HDPE moderator were carried out at intervals of an arbitrary period of time. As a result of the tests, the neutron count rate for the 10^5 rad irradiated HDPE moderator was decreased by about 7 % when compared with those of the others as shown in Fig.2. This phenomena seems to be caused by a density change or molecular weight change by a molecular structural change of the HDPE and a neutron scattering cross-section change by the new elements produced from a radiation-induced cross-linking, a scission and an oxidation. Neutron moderating power is in proportion to the density (ρ), average logarithm energy decrement (ξ) and scattering cross-section (σ) of the elements composing the moderator, and is inversely proportional to the molecular weight as shown in equation (2). As deduced from equation (2), the decrease of the neutron count rate for the 10^5 rad irradiated moderator when compared to the un-irradiated and other irradiated ones means that the density, neutron cross-section and average logarithm energy decrement of the moderator (HDPE) have decreased, or the molecular weight has increased. It can be confirmed through a density measurement, an elemental analysis, and a molecular weight and structural analysis whether the density, molecular weight, scattering cross-section and average logarithm energy decrement have decreased or not. Also the decrease of the neutron count rate (doubles rate) for each HDPE moderator according to the elapsed time in Fig.2, is due to a decrease of the neutron intensity by the decay of the ^{252}Cf neutron source.

4.2 Composition of the Elements in HDPE

The pure HDPE (CH_2) originally consists of C and H, but oxygen is attached to it due to a radiation-induced oxidation, due to a gamma radiation exposure of the HDPE material. As a result of the elemental analysis, it appears that the oxygen content is in proportion to the exposure rate for the irradiated HDPE moderators in Table 1. Carbon content was not in proportion to the exposure rate but it was increased for the irradiated HDPE moderators when compared with the un-irradiated one. And the hydrogen content for the 10^5 rad irradiated HDPE was lower than that of the other irradiated ones.

4.3 Neutron Moderating Power by a Theoretical Calculation

Table 2 shows the neutron moderating power (MP) calculated by the measured values and existing nuclear data [5]. Here, the calculated MP can be varied with the scattering cross-section data depending on the neutron energy, and the cross-section at 1 eV was used for this calculation. It shows that the neutron moderating power for the 10^5 rad irradiated HDPE moderator has a lower value than that of the other ones as listed in Table 2. These trends are almost the same as the direct neutron measurement values obtained from a neutron detection by the ^{3}He detection system. It indicates that the neutron moderating power of HDPE varies considerably with a specific radiation exposure rate and this phenomenon causes the changes in the molecular structure and the hydrogen and oxygen contents of the HDPE moderator due to a radiation exposure. MP is largely affected by the hydrogen content because it has the biggest neutron scattering cross-section. It seems to be separated and evaporated by the scission of

Table 1. Major elemental contents of the fresh and irradiated HDPE.

Element	HDPE					
	Fresh	0.1M rad	1.0M rad	10M rad	100M rad	1.0G rad
H, %	14.53	14.67	14.84	14.87	14.83	14.75
C, %	84.01	84.82	85.88	85.95	85.66	85.83
O, %	0.24	0.20	0.27	0.30	0.54	0.88

Table 2. Neutron moderating power (MP) calculated by using the existing nuclear data and the measured values of the HDPE properties.

HDPE	Fresh	10^5 rad	10^6 rad	10^7 rad	10^8 rad	10^9 rad
MP	2.10	1.78	2.12	2.12	2.11	2.10

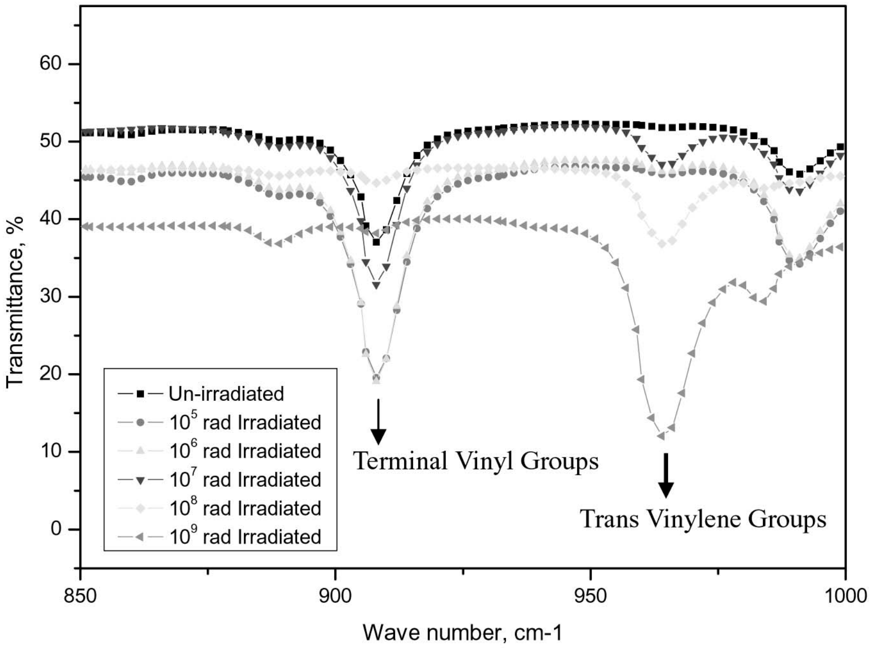

Fig. 3. Extended FTIR trace of the un-irradiated/irradiated HDPE at a range of 500-1,000 cm^{-1}.

–CH_2– during the process of a radiation-induced degradation. Also Its variation according to the gamma exposure rate is thought to be as a result of a separation and a recombination of hydrogen from/with the carbon or radicals. But a detailed mechanism has to be revealed through a further study as to why the hydrogen content for the 10^5 rad sample decreased more compared with the other exposure rates.

4.4 FTIR Spectroscopy

Fig.3 is the extended FTIR trace to observe the differences in detail, and the key frequencies used to identify the radiolytic yields in the HDPE are cited in reference [6]. Trans vinylene group related to a cross-linking at wave number 965 cm^{-1} and a terminal vinyl group related to a cross-linking and scission at wave number 910 cm^{-1} were observed as shown in Fig.3. The un-irradiated HDPE moderator did not change its molecular structure at 965 cm^{-1}, while all of the irradiated moderators had changed their molecular structures at this wave number. It means that the trans vinylene group was produced due to a radiation exposure.

5. CONCLUSIONS

HDPE neutron moderator structures were exposed to a gamma-ray source to a level of 10^5-10^9 rad to investigate their post-irradiation effects. As a result of the neutron measurement test by using these gamma irradiated moderator structures, it was confirmed that the neutron moderating power for the 10^5 rad irradiated HDPE moderator had decreased by about 7% when an un-irradiated pure one was used as a reference. And the others were almost equal to the fresh one. It implies that the neutron moderating power of HDPE does not depend on the integrated gamma dose and that it varies considerably at a specific dose. Also some techniques such as a FTIR spectroscopy, an element analysis, and so on were employed to establish the molecular structures and elemental contents. As a result of the FTIR spectrum analysis, it was confirmed that the molecular structure of HDPE had partially changed to the terminal vinyl groups, trans vinylene groups, and carbonyl groups, related to a cross-linking, scission, and an oxidation. In the element analysis, the oxygen contents appeared to increase with a gamma exposure rate increment due to a radiation-induced oxidation, and the hydrogen content for the 10^5 rad irradiated HDPE was lower than that of the other irradiated ones. The density of each HDPE was changed a little. Consequently, it was confirmed that the molecular structural and elemental changes had the most affect on the moderating power as a result of a gamma irradiation for the HDPE neutron moderators.

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