

International Journal of Nuclear Energy Science and Technology

ISSN online: 1741-637X - ISSN print: 1741-6361

<https://www.inderscience.com/ijnest>

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Khalid Nabaoui, Abdessamad Didi, Hamid Bounouira, Fouad Taous, Samira Mimount, Hamid Amsil, El Mehdi Alibrahmi, Chakir El Mahjoub, Mohammed Darouich, Hicham Jabri, Ilias Aarab, Abdelouahab Badague, Abdeslem Rrhioa, Otmane Allaoui

Article History:

Received:

Accepted:

Published online: 14 June 2024

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Khalid Nabaoui

LPMS, Faculty of Sciences,
Ibn Tofail University,
Kenitra, Morocco
Email: khalid.nabaoui@uit.ac.ma

Abdessamad Didi*

National Centre for Nuclear Energy,
Science and Technology,
Rabat, Morocco
Email: a.didi@ump.ac.ma
*Corresponding author

**Hamid Bounouira, Fouad Taous,
Samira Mimount and Hamid Amsil**

National Centre for Nuclear Energy,
Science and Technology,
Rabat, Morocco
Email: bounouira@cnesten.org.ma
Email: taous@cnesten.org.ma
Email: mimount@cnesten.org.ma
Email: amsil@cnesten.org.ma

**El Mehdi Alibrahmi and
Chakir El Mahjoub**

LPMS,
Faculty of Sciences,
Ibn Tofail University,
Kenitra, Morocco
Email: alibrahmi.elmehdi@uit.ac.ma
Email: elmahjoub.chakir@uit.ac.ma

Mohammed Darouich and Hicham Jabri

National Centre for Nuclear Energy,
Science and Technology,
Rabat, Morocco
Email: darouich@cnesten.org.ma
Email: Jabri@cnesten.org.ma

Iliasse Aarab and Abdelouahab Badague

LPMS,
Faculty of Sciences,
Ibn Tofail University,
Kenitra, Morocco
Email: Iliasse.Aarab@uit.ac.ma
Email: abdelouahab.badague@uit.ac.ma

Abdeslem Rrhious

Faculty of Sciences,
University Mohamed Premier,
Oujda, Morocco
Email: a.rrhious@ump.ac.ma

Otmene Allaoui

LPMS,
Faculty of Sciences,
Ibn Tofail University,
Kenitra, Morocco
Email: otmene.allaoui@uit.ac.ma

Abstract: This article explores the consequences of neutron irradiation on the characteristics of polyethylene used as sample shuttles sent for prolonged irradiation (3 hours) in the core of the TRIGA Mark II reactor. The results reveal alterations in the mechanical, thermal and chemical properties of polyethylene following neutron irradiation. Conclusions drawn from two distinct approaches – infrared and X-ray imaging – underscore the importance of understanding the impacts of neutron irradiation on materials intended to be exposed to neutron fluxes. This understanding is crucial to ensuring the safety and durability of these materials.

Keywords: neutron irradiation; polyethylene; shuttles; TRIGA Mark II; infrared; X-ray imaging; neutron fluxes.

Reference to this paper should be made as follows: Nabaoui, K., Didi, A., Bounouira, H., Taous, F., Mimount, S., Amsil, H., Alibrahmi, E.M., El Mahjoub, C., Darouich, M., Jabri, H., Aarab, I., Badague, A., Rrhioua, A. and Allaoui, O. (2024) 'Neutron irradiation-induced modifications in polyethylene properties within nuclear environments: analysis of shuttles irradiated in the TRIGA Mark II reactor', *Int. J. Nuclear Energy Science and Technology*, Vol. 17, No. 1, pp.85–97.

Biographical notes: Khalid Nabaoui is a Doctoral student at Ibn Tofail University, Faculty of Sciences. His research interests include neutron and gamma radiation.

Abdessamad Didi is a highly accomplished Researcher specialising in Nuclear and Medical Physics, as well as Monte Carlo methods. He received two PhD degrees, one in Materials Science for Energy and Environment from the Faculty of Sciences in Fes, Morocco and the other in Nuclear Physics from the Faculty of Sciences in Oujda, Morocco. He is working as Researcher the National Centre for Nuclear Energy, Science and Technology in Morocco. His research interests include spallation physics and its applications in nuclear reactors and medical contexts. He has published extensively in prestigious journals and conferences, primarily in nuclear energy, simulation and spallation physics. He is an Active Reviewer and holds Editorial Roles for various journals, and he has contributed significantly to over 30 international conferences in different committee capacities.

Hamid Bounouira is a nuclear physicist with a PhD in Nuclear Physics and Applications, with over 19 years of experience at CNESTEN, he has published close to 60 international journal articles and contributed to scientific committees, serving as a Referee in research reactor application and neutron activation analysis and applications.

Fouad Taous is a Head of Stable Isotopes Laboratory and is a Nuclear Techniques Physicist with more than 20 years of experience at CNESTEN.

Samira Mimount is a Head of Non-Destructive Testing Department in Industrial Applications Division, Directorate of Studies & Scientific Research at the National Centre for Energy, Sciences and Nuclear Techniques.

Hamid Amsil is a Nuclear Physicist with a PhD in Nuclear Physics and Applications, as well as two MSc degrees in Particle Physics and Radiation Technologies from France and Morocco, respectively. With over 15 years of experience at CNESTEN, he has published close to 70 international journal articles and contributed to scientific committees, serving as a Referee in research reactor, reactor physics and applications.

El Mehdi Alibrahmi is a Professor at Faculty Member at Ibn Tofail University, Faculty of Sciences in Kenitra. He has directed numerous scientific research projects in the fields of Radiation Detection, Radioactivity, Applied Statistics, Radiation Protection, Radiation Physics, Ionising Radiation, Environmental Radioactivity and Radiation Dosimetry. He has authored many papers and communications in this field, and is an Associate Professor at the Higher School of Education and Training.

Chakir El Mahjoub is a Professor at Ibn Tofail University, Faculty of Sciences in Kenitra. He earned his PhD degree in Nuclear Physics from the Faculty of Sciences at Mohammed V University, Rabat, Morocco in 1994. Currently, he

holds the position of Research Professor at the Faculty of Sciences, Ibn Tofail University in Kenitra, Morocco. Additionally, he serves as the Director of the Materials and Subatomic Physics Laboratory at the same institution. His research interests include reactor physics, medical physics and science pedagogy. He has a substantial publication record with numerous research articles in indexed international journals, covering both nuclear physics and science pedagogy.

Mohammed Darouich works in Non-Destructive Testing Department, Industrial Applications Division, Directorate of Studies & Scientific Research at the National Centre for Energy, Sciences, and Nuclear Techniques.

Hicham Jabri is an Executive at the National Centre for Energy, Sciences and Nuclear Techniques.

Ilias Aarab is a PhD student in Nuclear Physics at the Faculty of Sciences of Ibn Tofail University in Kenitra. He is also a Member of the Neutron Activation Analysis Team at the National Centre for Energy, Science and Nuclear Technology in Rabat, Morocco, since 2010. He holds a Master's degree in Nuclear Sciences and Techniques and his research interests include application of nuclear techniques in the field of environmental studies.

Abdelouahab Badague is a Member of the Neutron Activation Analysis Team at the National Centre for Energy, Science and Nuclear Technology in Rabat, Morocco, since 2010. He holds a Master's degree in Nuclear Sciences and Techniques, and his interests include the application of nuclear techniques in the field of environmental studies.

Abdeslem Rhioua is a Professor of Physics at the Mohammed First University of Oujda, Morocco. He received PhD degree in Particle Physics from Mohammed First University. His current research interests include radiotherapy, radioactivity and radiation detectors.

Otmane Allaoui is a Doctoral student at Ibn Tofail University, Faculty of Sciences. His research interests include neutron and gamma radiation.

1 Introduction

This article focuses on specific polyethylene shuttles, measuring 13 cm in height, 2 cm in diameter and 1 mm in thickness (see Figure 1). These shuttles are deployed as long-term irradiation devices within the core of the TRIGA Mark II reactor, (Aarab et al. 2023). Their precise positioning in the reactor's rotating sample holder is ensured through the use of a fishing-channel-type clamp.

These polyethylene shuttles play a crucial role in maintaining samples such as plant soils and other materials inside the cores of nuclear reactors. However, it is important to note that the special materials used in the design of these shuttles are subjected to high levels of neutron irradiation, [Aarab et al. (2023), Chahidi et al. (2023), Ahmed et al. (2023) and Chahidi et al. (2021)]. This intense exposure can potentially alter the structure as well as the mechanical and thermal properties of these materials.

Figure 1 The polyethylene shuttles numbered 0 not irradiated, 1 irradiated once and 2 irradiated twice (see online version for colours)



Polyethylene, characterised by its resistance to corrosion, is a common choice in nuclear reactor environments, (McKeen 2020). Nevertheless, given the risks associated with neutron irradiation, it is imperative to conduct thorough studies on its implications for the structure of polyethylene ($-(CH_2-CH_2)_n-$), (Chen et al. 2017). Understanding these consequences is essential to ensure the long-term safety and durability of these crucial shuttles in the nuclear environment (Wakai 2021).

2 Materials and methods

The polyethylene shuttles were subjected to a neutron beam with a power of 500 kW in the rotating sample holder within the core of the TRIGA Mark II reactor (see Figure 1). The first irradiation took place in October 2018, and the second one occurred in September 2019.

The shuttles filled with samples contain matrices such as soils, plants or others, according to a protocol involving an irradiation period, followed by a 24-hour cooling phase in a shielded cell (see Figure 2) (El Basraoui et al., 2023). This cooling phase is crucial to avoid any risk of radioactive contamination. The shielded cell is designed to contain emitted radiation and handle samples, using multiple layers of lead to absorb the radiation and prevent it from spreading outside.

Figure 2 Shielded cell for the handling and cooling of shuttles (see online version for colours)



The irradiation technique is crucial for analysing the composition and properties of soil samples. The reactor's rotating sample holder enables uniform irradiation of the samples, ensuring accurate and reliable results (Bounouira et al. 2014).

3 Result

In Figure 1, we present an irradiation experiment involving six polyethylene shuttles. These shuttles are cylindrical samples of high-density polyethylene, with identical shapes and sizes. The objective of this study is to assess the effects of irradiation on the material's properties.

The shuttles are divided into three groups. The first group, labelled '0,' serves as an unirradiated control. The two shuttles in the '0' groups have never been exposed to a radiation source, while the two shuttles in the '1' group underwent a single irradiation. The last two shuttles, labelled '2,' were chosen to study the cumulative effect of irradiation. Consequently, the shuttles in the '2' group underwent two successive irradiations to simulate real usage conditions.

Each shuttle was exposed to a dose of ionising radiation equivalent to a 3-hour-long irradiation. This irradiation duration was chosen to simulate real usage conditions. The irradiation power was set at 500 kW, corresponding to a high intensity of ionising radiation.

Irradiation can have significant effects on the physical and chemical properties of materials. For example, it can induce changes in molecular structure, alterations in colour and transparency, as well as modifications in mechanical strength, according to results obtained through two techniques infrared (IR) and X-ray imaging (RX). This experiment aims to better understand these effects and evaluate the impact of irradiation on polyethylene properties, which may have significant implications for industrial applications.

The exact weights of each shuttle were carefully noted and recorded in Table 1 to ensure precise result analysis. This experiment will provide a better understanding of the effects of irradiation on polyethylene properties.

Table 1 The properties of long-irradiation shuttles

<i>Number of irradiation</i>		<i>Weight (g)</i>	<i>Error (g)</i>
None	0	24.684	0.001
None	0	24.684	0.001
One time (500kW)	1	24.304	0.002
One time (500kW)	1	24.305	0.002
Two times (500kW)	2	24.282	0.001
Two times (500kW)	2	24.291	0.001

The results showed that neutron irradiation had significant effects on polyethylene properties (see Figure 1 and Table 1). Irradiated samples exhibited a decrease in tensile strength and hardness, as well as a reduction in thermal stability. The density of irradiated samples also decreased.

Upon initial visual observation of the shuttles (see Figure 1), a colour difference is noticeable between the different groups. The control group '0' shuttles are white, indicating they have never been used. In contrast, the '1' group shuttles in the middle display a slight pale yellow coloration, while the '2' group shuttles on the right exhibit a more pronounced dark yellow colour. These colour differences result from the effect of irradiation with neutrons and gammas in the core of the TRIGA Mark II reactor on the polyethylene of the shuttles.

Irradiating shuttles with neutrons and gammas is a common method to simulate the effect of ionising radiation on materials. Neutrons are particularly effective at inducing nuclear reactions in atomic nuclei, whereas gammas can ionise atoms by stripping electrons from their orbits.

The effect of irradiation on polyethylene can be explained by the formation of free radicals in the material's molecules. Free radicals are atoms or molecules with incomplete electro- neutrality, making them highly reactive. When formed in polyethylene under the influence of ionising radiation, they can induce chemical reactions with other molecules, thus forming cross-links between polymer chains.

The formation of these cross-links alters the physical and chemical properties of the material. Specifically, it can increase its rigidity, tensile strength and melting temperature. The colour of the shuttles can also be affected by irradiation, as the formed cross-links may absorb certain wavelengths of light.

Observing colour differences among shuttle groups is thus an initial indicator of the effects of irradiation with neutrons and gammas on polyethylene. More detailed analyses will be required to understand the underlying mechanisms of these colour changes and assess their impact on the material's physical and chemical properties.

A more in-depth analysis, such as infrared spectroscopy or other material characterisation techniques could help identify specific chemical changes occurring during irradiation and provide a better understanding of the underlying mechanisms.

Table 1 presents the shuttle weighing results in grams (g) for three different groups with associated errors. The groups were subjected to varying amounts of irradiation.

Group 0 was not irradiated, and two shuttles were weighed, yielding identical results of 24.684 g with a measurement error of 0.001 g. Group 1 was irradiated once, and the two shuttles were weighed, resulting in weights of 24.304 g and 24.305 g, with a measurement error of 0.002 g. Group 2 was irradiated twice, and the two shuttles were weighed, resulting in weights of 24.282 g and 24.291 g, with a measurement error of 0.001 g. Overall, there appears to be a decrease in the weight of shuttles with an increase in irradiation. However, it is important to note that the measurement errors are very low, and there may be natural variations in the weight of shuttles.

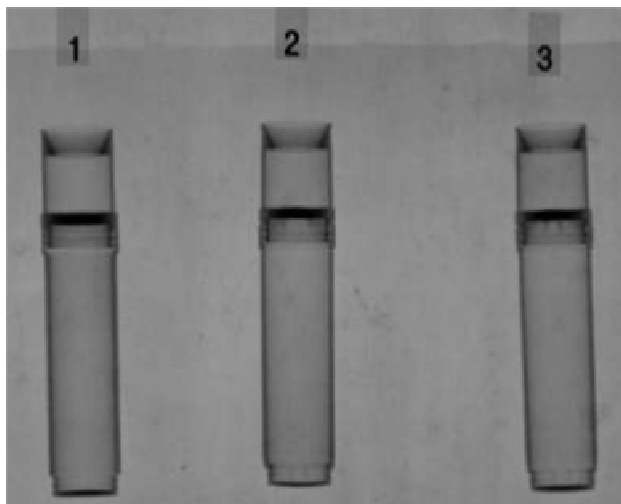
3.1 X-ray imaging

X-ray imaging, a widely used technique in medicine and various scientific fields, utilises X-rays to produce detailed images. An X-ray source emits photons through the body, and a detector records the photons passing through tissues, creating contrast in the image. Dense areas appear white, while soft tissues appear darker.

In this study, we used X-Ray Imaging (XRI) to examine potential anomalies during the irradiation of polyethylene tubes in the core of the TRIGA Mark II reactor. The irradiation, performed at 500 kW for 3 hours, involved one tube irradiated once and another tube irradiated twice. Observations from the images (see Figure 3) reveal

modifications to the threads of tubes 2 and 3, accompanied by deformations at their closure, as well as alterations at their base, compared to the control tube.

Figure 3 Shuttle behaviour before and after neutron irradiation using X-ray imaging (XRI) (see online version for colours)



The chemically observed changes in irradiated polyethylene can result from various processes induced by neutron irradiation. These processes typically involve degradation or modification reactions of the molecular structure of polyethylene due to exposure to reactor neutrons.

Possible chemical reactions include molecular bond cleavage, the formation of free radicals, changes in molecular conformation and potentially rearrangements or secondary reactions. These chemical modifications can lead to alterations in the mechanical and structural properties of the material, as observed in the thread connections, closure of tubes and other parts of the polyethylene.

3.2 *Infrared spectroscopy*

Infrared spectroscopy is an analytical method based on the interaction between infrared light and matter. It measures the absorption of specific molecular vibrations of atomic bonds, as demonstrated in our study on polyethylene. The sample is exposed to an infrared light source, and variations in light intensity are recorded by a detector to create a spectrum. This spectrum helps identify functional groups in the substance, providing information about its chemical composition. Infrared spectroscopy is widely applied in various scientific fields.

Figure 4 and Table 2 detail the results obtained by infrared techniques for three types of irradiation shuttles: non-irradiated (non-irradiated shuttle), irradiated once for 3 hours at 500 kW (shuttle irradiated once), and irradiated twice for 3 hours each at the same power (shuttle irradiated twice). The last two underwent irradiation in the Rotary Specimen Rack (RSR) position of the TRIGA Mark II reactor. The table organises the data into three columns: the first indicates the wavelength, the second represents the

percentage transmittance after a single irradiation, and the third shows the percentage transmittance after two successive irradiations of three hours each.

Figure 4 Analysis of the ageing of polyethylene using infrared technique (see online version for colours)

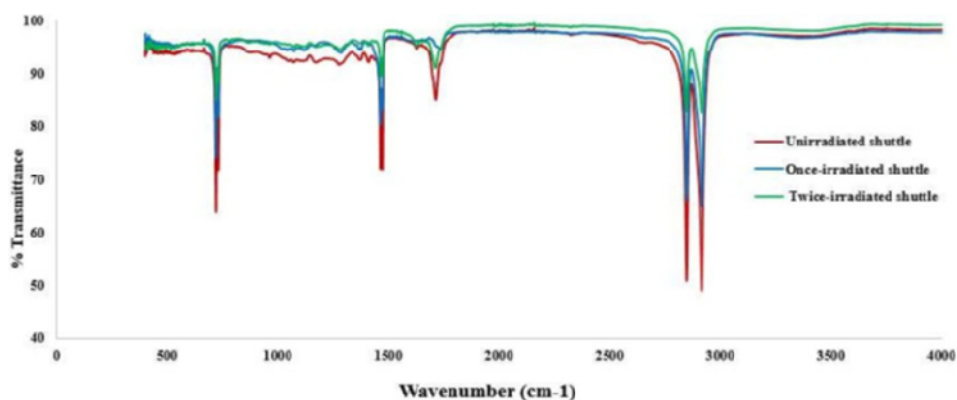


Table 2 Analysis of the ageing of polyethylene using infrared technique

Wavenumber (cm ⁻¹)	Unirradiated shuttle	% transmittance Once- irradiated shuttle	Twice-irradiated shuttle
2915.5	49.05	64.91	82.46
2848	50.86	66.28	82.75
719	64.4	74.29	85.27
720	64.92	75.34	85.41
730	71.8	81.76	88.13
729	73.94	83.42	88.36
1473.5	74.78	84.86	91.04
1471	78.78	86.64	91.66
1468.5	83.13	86.89	92.88
1468	83.37	86.74	92.93
1300.5	92.43	94.7	95.23

The discussion of the results reveals several notable trends. Overall, there is a progressive increase in transmittance percentage with the increase in wavelength for all shuttles, indicating better infrared transmission at higher wavelengths.

Analysing the irradiated shuttles compared to the non-irradiated shuttle shows a general increase in transmittance after each irradiation session. This suggests that irradiation modifies the optical properties of the shuttle materials, leading to greater infrared transmission.

These results provide crucial insights into understanding the effects of irradiation on the properties of irradiation shuttles, which can have significant implications for the development and improvement of materials intended for similar environments. The

variation in transmittance with wavelength also offers avenues for further studies on the underlying mechanisms of these optical changes.

Figure 4 and Table 2 represent the percentage of transmittance in the case of no irradiation, single irradiation, and double irradiations of the polyethylene shuttle based on wavelength, along with the associated infrared peaks and transmittance percentages for specific values. These data highlight significant changes among the non-irradiated, once irradiated and twice irradiated samples, along with the corresponding chemical formulas:

- 2915.5 cm^{-1} (*C-H Groups*): Transmittance increases remarkably from 49.05% (non-irradiated) to 64.91% (once-irradiated) and then to 82.46% (twice-irradiated). These variations could indicate changes in the composition of C-H groups, suggesting a possible evolution of hydrocarbon chains.
- 2848 cm^{-1} (*Aliphatic C-H Bonds*): Transmittance significantly increases from 50.86% (non-irradiated) to 66.28% (once irradiated) and then to 82.75% (twice irradiated), emphasising potential modifications in aliphatic bonds, such as hydrocarbon chains.
- 719 cm^{-1} (*Alkyl Groups or Carbon-Carbon Bonds C-H*): A notable increase in transmittance from 64.4% (non-irradiated) to 74.29% (once-irradiated) and then to 85.27% (twice irradiated) is observed. These variations could indicate changes in C-H bonds, suggesting the evolution of alkyl groups or other carbon-carbon bonds.
- 720 cm^{-1} (*Out-of-Plane Vibrations of C-H Bonds*): Transmittance remarkably increases from 64.92% (non-irradiated) to 75.34% (once irradiated) and then to 85.41% (twice-irradiated), highlighting potential modifications in the out-of-plane vibrations of C-H bonds, indicating changes in alkyl groups or other carbon-carbon bonds.
- 730 cm^{-1} (*Hydrocarbon Chains C-H*): A significant increase in transmittance from 71.8% (non-irradiated) to 81.76% (once irradiated) and then to 88.13% (twice irradiated) suggests changes in the vibrations of C-H bonds, emphasising the possible evolution of hydrocarbon chains.
- 729 cm^{-1} (*Alkyl Groups*): Transmittance notably increases from 73.94% (non-irradiated) to 83.42% (once-irradiated) and then to 88.36% (twice-irradiated), highlighting variations in the vibrations of C-H bonds, indicating the possible evolution of alkyl groups.
- 1473.5 cm^{-1} (*Methyl C-H Groups*): A significant increase in transmittance from 74.78% (non-irradiated) to 84.86% (once-irradiated) and then to 91.04% (twice irradiated) suggests changes in the asymmetric vibrations of C-H bonds, indicating potential evolution of methyl groups.
- 1471 cm^{-1} (*Methyl C-H Groups*): Transmittance increases remarkably from 78.78% (non-irradiated) to 86.64% (once-irradiated) and then to 91.66% (twice irradiated), highlighting potential modifications in the asymmetric vibrations of C-H bonds, indicating changes in methyl groups.

- 1468.5 cm^{-1} (*Alkyl C-H Groups*): Transmittance notably increases from 83.13% (non-irradiated) to 86.89% (once irradiated) and then to 92.88% (twice-irradiated), suggesting changes in the asymmetric vibrations of C-H bonds, indicating possible evolution of alkyl groups.
- 1468 cm^{-1} (*Alkyl C-H Groups*): Transmittance increases remarkably from 83.37% (non-irradiated) to 86.74% (once-irradiated) and then to 92.93% (twice-irradiated), emphasising variations in the asymmetric vibrations of C-H bonds, indicating possible changes in alkyl groups.
- 1300.5 cm^{-1} (*Methyl or Other Functional Groups C-H*): Transmittance significantly increases from 92.43% (non-irradiated) to 94.7% (once irradiated) and then to 95.23% (twice irradiated), suggesting potential changes in the vibrations of methyl groups or other functional groups C-H.

These transmittance percentage variations indicate alterations in the molecular structure of polyethylene following neutron irradiation. The notable changes between non-irradiated, once-irradiated, and twice-irradiated samples suggest significant chemical modifications. The specific values of 1468, 1468.5 and 1471 cm^{-1} , associated with alkyl, methyl and other C-H bonds clearly illustrate these changes, highlighting the significant impact of neutron irradiation on the chemical composition of polyethylene.

In summary, the detailed analysis of infrared peaks demonstrates that polyethylene undergoes notable chemical modifications during neutron irradiation, with significant variations in alkyl, methyl and other C-H bonds. These changes underscore the importance of understanding the effects of neutron irradiation on the composition and properties of polyethylene, which can have crucial implications for its use in nuclear environments.

4 Discussion

The results of this study reveal significant effects of neutron irradiation, with implications for the safety and durability of materials in a nuclear environment. Both techniques used, namely infrared and X-ray imaging, have shown important findings regarding the behaviour of polyethylene under neutron radiation.

Polyethylene demonstrates some resistance to neutron and gamma radiation due to its simple molecular structure and low density, limiting interactions with ionising radiation. However, the resistance of polyethylene depends on various factors such as radiation dose, exposure duration, temperature, and the presence of other chemical or physical agents that may affect its structure. Although polyethylene is less sensitive to the effects of ionising radiation, high doses or prolonged exposure can lead to degradation, reducing its physical and mechanical properties.

Performance tests under irradiation may be necessary to assess the lifespan and long-term resistance of polyethylene in applications involving neutron and gamma radiation. The results of this study indicate significant effects of neutron irradiation on the properties of polyethylene, with potential implications for the safety and durability of materials used in nuclear reactor environments. Further research may be needed to evaluate effects on a larger scale and determine appropriate measures to minimise damage caused by neutron irradiation.

At the molecular level, neutron irradiation can induce various reactions and modifications in polyethylene. These include molecular bond cleavage, the formation of free radicals, changes in molecular conformation, molecular rearrangements and secondary reactions. These chemical modifications can impact the mechanical, thermal and structural properties of polyethylene, as observed in the study, where changes were detected in thread connections and tube closure, as well as through infrared technique.

The thermal effect of neutron irradiation on polyethylene includes temperature elevation, changes in thermal conductivity, variations in thermal capacity, alterations of thermoplastic properties and modifications of phase transitions. These changes depend on factors such as irradiation dose, exposure duration and the initial characteristics of polyethylene.

From a mechanical perspective, neutron irradiation can lead to alterations in mechanical strength, ductility, toughness, stiffness and rupture toughness of polyethylene. These mechanical changes are also influenced by parameters such as irradiation dose, exposure duration and the initial characteristics of the material. In summary, a thorough understanding of the effects of neutron irradiation on polyethylene is crucial to ensure the reliability and durability of this material in nuclear environments.

5 Conclusion

In conclusion, neutron irradiation has significant effects on the properties of polyethylene, emphasising the importance of understanding the effects of neutron irradiation on materials in nuclear reactor environments to ensure their safety and durability. Further studies may be necessary to assess effects on a larger scale and to determine appropriate measures to minimise damage caused by neutron irradiation on materials used in sample irradiation in the reactor core. Measures can be taken to minimise damage from neutron irradiation, such as using materials more resistant to radiation or materials with a low neutron capture cross-section. The results of this study can help inform the selection and use of materials in nuclear reactor environments to ensure their long-term safety and durability. Additionally, further studies could explore the use of post-irradiation treatments to improve the properties of irradiated materials.

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