DESDE 2013
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Pädi Boletín Científico de Ciencias Básicas e Ingenierías del ICBI

ISSN: 2007-6363

Publicación Semestral Pädi Vol. 13 No. Especial 4 (2025) 40-45

Gamma radiation effect in polycarbonate (with and without UV stabilizer)
Efectos de la radiación gamma en policarbonato (con y sin estabilizante UV)

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Resumen

Se investigó el efecto de la radiación gamma a dosis de 100 a 500 kGy sobre la estabilidad mecánica y térmica en dos grados de policarbonato con y sin estabilizador UV. La caracterización se realizó mediante análisis visual de color, análisis dinámico mecánico (DMA), y tratamientos de recocido térmico post-irradiación a temperaturas por debajo y por encima de la temperatura de transición vitrea (Tg). Ambos grados de policarbonato mostraron un amarillamiento dependiente de la dosis de radiación, mientras que los resultados de DMA muestran una disminución en la Tg en ambos materiales, indicativo de rompimiento de cadenas, observando un efecto menos pronunciado en el policarbonato con aditivo UV (3.21°C contra 4.24°C a una dosis de 500 kGy). Las pruebas de recocido muestran como el PC sin protección sufrió una mayor formación de burbujas y deformación debido a la expansión de subproductos, un fenómeno el cual se ve suprimido en el PC con estabilizador UV. El trabajo concluye que el estabilizador UV proporciona protección mitigando la reducción en el peso molecular al limitar el rompimiento de cadenas y mejora la estabilidad térmica post-irradiación al inhibir la formación de productos volátiles.

Palabras clave: Policarbonato, Radiación gamma, aditivos estabilizantes UV.

Abstract

The effect of gamma radiation doses (100 to 500 kGy) on the thermomechanical and thermal stability of two polycarbonate (PC) grades, one with and one without a UV stabilizer, was investigated. Characterization was performed using visual color analysis, dynamic mechanical analysis (DMA) to determine the glass transition temperature (Tg), and post-irradiation annealing treatments at temperatures below and above the Tg. Both PC's exhibited dose-dependent yellowing while DMA results revealed a decrease in the glass transition temperature for both materials, indicative of chain scission, but the effect was less pronounced in the UV stabilized PC (a drop of 3.21°C vs 4.24°C at 500 kGy). The annealing studies demonstrated that the unprotected PC suffered severe foaming and deformation due to the expansion of trapped byproducts, a phenomenon suppressed in the PC-UV specimens. The study concludes that the UV stabilizer provides a protective mechanism mitigating the reduction in molecular weight by limiting chain scission and enhancing post-irradiation thermal stability by inhibiting the formation of volatile degradation products.

Keywords: Polycarbonate, Gamma radiation, UV stabilizer.

1. Introduction

Polycarbonate (PC) is a thermoplastic form of engineering polymers widely used in high performance applications, such as automotive components, medical devices and consumables due to its exceptional combination of transparency, impact strength and dimensional stability. In many of these fields, PC components must undergo gamma radiation sterilization, an effective method that can induce degradation in the polymer. This degradation can be observed as yellowing, loss of ductility and the alteration of mechanical properties, thereby limiting material´s service life. (Gohil 2022) (Zhou 2023)(Osanloomehr 2023) (Grzelak 2023)

To mitigate these effects, the formulations with stabilizing additives into polycarbonate formulations is a common practice. UV stabilizers, primarily designed to protect against photolytic degradation, are a frequent addition. However, their efficacy in protecting against ionizing radiation, such as

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Historial del manuscrito: recibido el 22/09/2025, última versión-revisada recibida el 24/10/2025, aceptado el 03/11/2025, publicado el 12/12/2025. **DOI:** https://doi.org/10.29057/icbi.v13iEspecial4.16085

gamma radiation is not fully characterized. Therefore, it is crucial to understand whether these additives offer cross-protection or if specific formulations are required for high-radiation environments. (Yu 2021) (Lan 2022) (Rodríguez-Ramírez 2021)

The objective of this work is to systematically investigate and compare the effect of gamma radiation on the properties of two polycarbonate grades: one standard and one containing a UV stabilizer additive. The impact of doses from 100 to 500 kGy was evaluated on colour change, viscoelastic properties via Dynamic Mechanical Analysis (DMA), and post-irradiation thermal stability through annealing treatments. This study aims to determine the efficacy of the UV stabilizer in mitigating the different degradation mechanisms induced by gamma radiation.

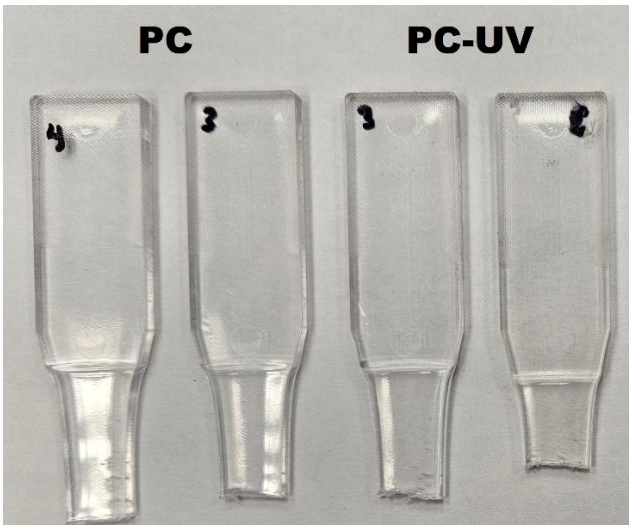

Figure 1: PC injected samples

2. Materials and methods

2.1 *Materials*

Two types of polycarbonates form the same commercial brand were used: one containing a proprietary ultraviolet stabilizing additive (PC UV) and the other without this additive (PC). Both materials were supplied in pellet form.

2.2 Preparation of specimens

Pellets of both materials were dried at 110 °C for 4 hours to reduce moisture below 0.2%. Tensile specimens were produced according to ISO 527 standard using a 100-ton injection molding machine equipped with a two-cavity steel mold. Typical molding conditions included a melt temperature of 270 °C (280°C in the nozzle), a mold temperature of 100°C, injection pressure of 100 MPa and packing pressure at 70% of the peak value. Cooling was maintained for 15 s to allow proper solidification. The parameters were optimized to obtain defect-free specimens with reproducible weight and geometry, suitable for irradiation.

2.3 *Gamma irradiation*

The tensile specimens were exposed to gamma radiation in a JS-6500 irradiator equipped with cobalt-60 sources at the National Institute of Nuclear Research of México (ININ). Irradiation was carried out at room temperature, under atmospheric conditions and an absorbed dose of 8 kGy/h. The selected doses were 100, 200, 300, 400 and 500 kGy. After irradiation, the specimens were stored at ambient conditions in sealed bags to avoid environmental degradation.

3. Experimental testing

3.1 Dynamic mechanical analysis (DMA)

DMA was carried out using a Q800 (TA instruments) in three-point bending mode. Tests were conducted from 30°C to 170°C at a heating rate of 5°C/min. The measured parameters included the storage modulus, loss modulus and damping factors. The glass transition temperature was determined from the peak of the tan delta (δ) curve.

3.2 Annealing treatment

Thermal annealing was performed to assess thermal stability of irradiated PC. Specimens were exposed in a convection oven at 135°C and 165°C for 2 hours under ambient atmosphere. After annealing, the samples were cooled to room temperature inside the oven to reduce residual stress. Macroscopic changes such as bubble formation, surface color variations and dimensional distortion were recorded as qualitative indicators of thermal degradation.

4. Results and discussion

After gamma irradiation, both PC specimens (UV-stabilized and non-stabilized) exhibited noticeable changes in color. At low doses (100-200 kGy), the variation was subtle, showing a slightly yellowish tone. However, as the radiation increased (300-500 kGy), both specimens developed a more intense color, ranging from light amber to light brown (figure 2).

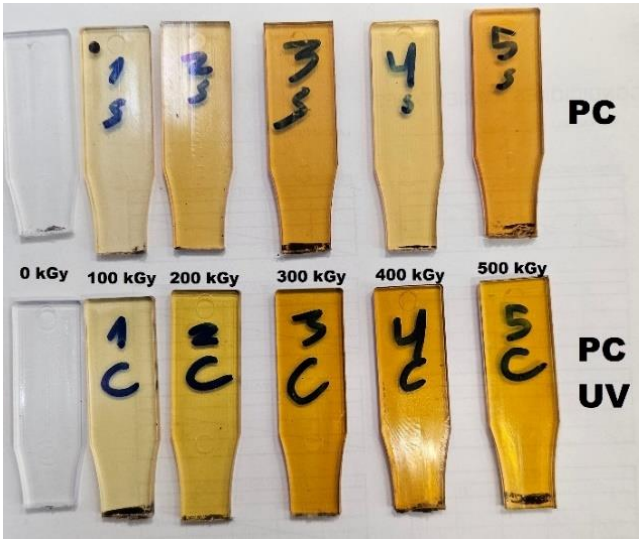

Figure 2: Color changes after irradiation.

The yellowing observed in PC specimens after gamma irradiation (100-500 kGy) is consistent with reported mechanisms of radiation induced color change. Gamma rays generate color centers (defects that absorb visible light) leading to a shift from transparent to amber or brown hues. This effect intensifies with dose as structural disorder increases, evidenced by a reduction of the optical bandgap, indicative of defect accumulation (Eggenhuisen 2022). At moderate doses, crosslinking can contribute to changes in refractive index, while at higher doses, degradation dominates, enhancing discoloration. Literature confirms that conventional PC exhibits a strong dose dependent yellowing, whereas stabilized formulations containing UV or color compensation additives show significantly milder changes, highlighting the protective role of stabilizers in delaying the buildup of chromophores responsible for visible color change. (De Brouwer 2022) (Ferreira 2009) (Nouh 2025) (Prajzler 2022).

4.2 DMA results

4.2.1 Polycarbonate without UV stabilizer

In the case of the PC without UV protection additive, the DMA results reveal a clear behavior dependent on the irradiation dose. Both the loss modulus (E¨) and the tan delta (than δ) show peaks associated with the transition temperature and relaxation of the material.

The loss modulus (Figure 3) indicates that, while relaxation mechanism is not drastically altered, a decreasing trend in the peak temperature can be observed as the dose increased. Still, the peaks tend to broaden, which suggests an increase in the heterogeneity of the polymeric network due to degradation. (Ramani 2003) (Silvia 2021).

This trend is confirmed in the tan δ curves (Figure 4), where the peak progressively shifts toward lower temperatures as the radiation increases. The glass transition values, extracted from the tan δ peaks (table 1) quantify this reduction, decreasing from 147.37°C for the 0 kGy sample to 143.13°C for the sample irradiated to 500 kGy. This behavior is an indication that the predominant mechanism is chain scission, which reduces the average molecular weight, thereby increasing the chain mobility and lowering the Tg. (Sinha 2004).

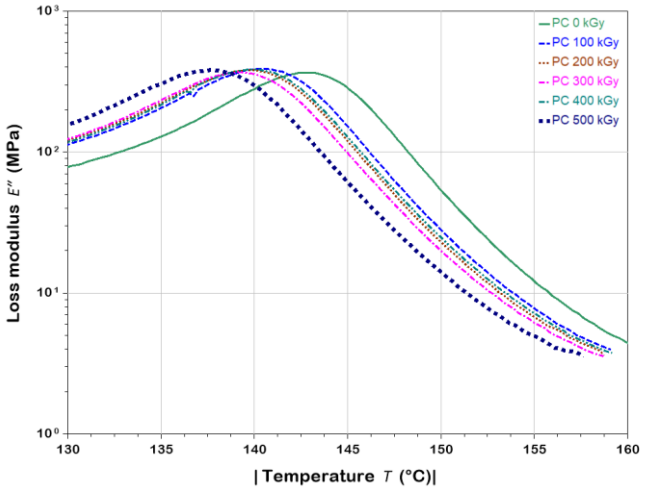

Figure 3: Loss modulus of PC samples at different gamma doses.

The anomaly presented at 400 kGy, where the glass transition (144.90°C) is higher than that at 300 kGy (144.20°C). This phenomenon suggests the existence of competing mechanisms of chain scission and crosslinking. While scission seems to be the dominant process, it is possible that at 400 kGy, the effect of crosslinking (raising the glass transition temperature) may manifest transiently before chain scission becomes dominant again at higher doses.

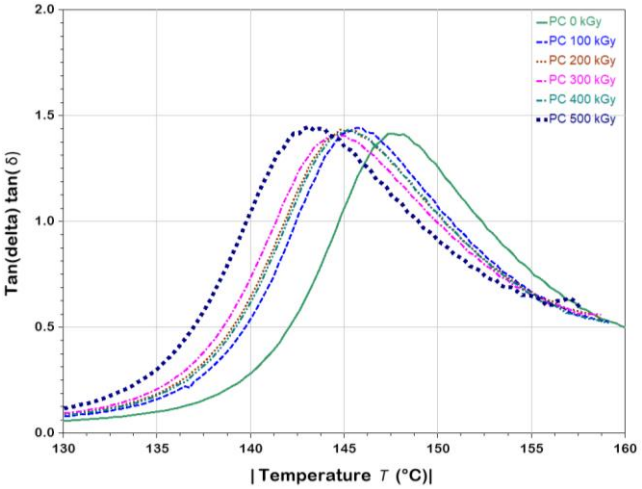

Figure 4: Tan δ curves of PC samples at different gamma doses.

Table 1: Glass transition temperatures of PC samples.

Specimen	Glass transition (°C)
PC 0 kGy	147.37
PC 100 kGy	145.67
PC 200 kGy	144.99
PC 300 kGy	144.20
PC 400 kGy	144.90
PC 500 kGy	143.13

4.2.2 Polycarbonate with UV stabilizer

The PC containing the UV stabilizer additive (PC-UV) exhibited a similar response to gamma radiation, although with

notable differences. The DMA results show that relaxation peak, identified from the loss modulus graphs (Figure 5), consistently shifts to lower temperatures as the radiation dose increases.

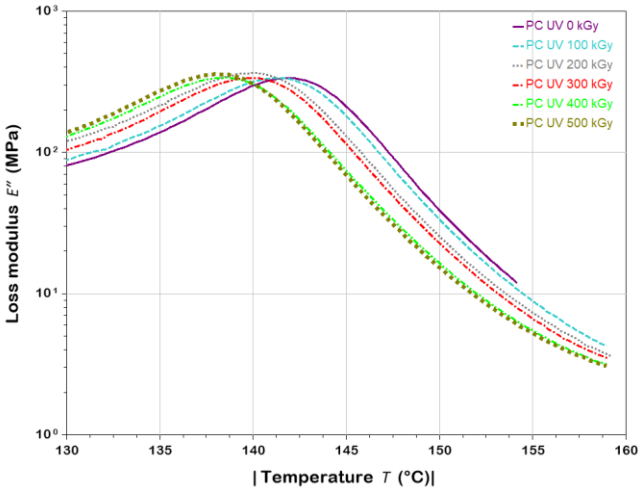

Figure 5: Loss modulus of PC-UV samples at different gamma doses.

Table 2: Glass transition temperatures of PC-UV samples

Specimen	Glass transition (°C)
PC-UV 0 kGy	146.94
PC-UV 100 kGy	146.62
PC-UV 200 kGy	145.72
PC-UV 300 kGy	145.30
PC-UV 400 kGy	144.36
PC-UV 500 kGy	143.73

A direct comparison between the two polycarbonate grades reveals the protective role of the UV stabilizer additive against gamma radiation induced degradation. First, at the base line (0 kGy), the initial value of the glass transition of the PC-UV (146.94 °C) is slightly lower than that of the standard PC (147.37 °C). This may suggest that the additive itself has a minor plasticizing effect, increasing the free volume within the polymer matrix.

The most significant finding lies in the magnitude of the glass transition reduction. After exposure to a 500 kGy dose, the standard PC experiment a total Tg drop of 4.24°C, whereas in the stabilized PC-UV a smaller drop of 3.21°C can be seen. This demonstrates that the stabilizer mitigates the extent of chain scission, preserving the molecular weight of the polymer to a greater degree. Additionally, the absence of the 400 kGy anomaly in the PC-UV sample suggests that the additive may also inhibit secondary reactions, such as crosslinking, leading to a more predictable degradation pathway.

Following the dynamic mechanical analysis, a significant post-test phenomenon concerning the material´s thermal stability was observed. Upon competition of the thermal ramp to 160°C, many of the irradiated specimens exhibited severe internal bubble formation and a notable loss of dimensional integrity, as shown in Figure 7. This effect was most pronounced in the non-stabilized PC samples at higher radiation doses. To further explore and systematically characterize this post-irradiation thermal instability, a series of annealing treatments were performed.

Figure 7: Post-DMA testing specimens

4.3 Annealing treatments

To evaluate the post-irradiation stability, the polycarbonate specimens were subjected to two distinct annealing treatments in a convection oven, each for a duration of 2 hours. The first annealing cycle was performed at 135 °C, a temperature approximately 10°C below the glass transition temperature, to assess sub-Tg structural relaxations. A second set of specimens was subjected to an annealing cycle at 155°C, 10°C above the glass transition, to investigate the material's stability in the rubbery state. After each treatment, the samples were allowed to cool slowly to room temperature inside the oven to minimize residual stress.

The first annealing treatment (below the glass transition) revealed significant differences in the thermal stability of the irradiated samples (Figure 8). The PC specimens without UV stabilizer displayed a clear and progressive increase in the internal bubble formation starting from 100 kGy, intensifying at 200 and 300 kGy. Beyond these doses, at 400 and 500 kGy, the samples exhibited severe deformation and bulging, indicating a substantial loss of dimensional integrity. This pronounced foaming and deformation suggest that even at a sub-Tg temperature, the accumulation of volatile degradation by products from irradiation was sufficient to exert internal pressure, leading to structural damage in the highly degraded matrix. In contrast, the PC-UV stabilized samples maintained dimensional stability across all radiation doses. Less bubble formation and no macroscopic deformation were observed, even at the higher dose of 500 kGy, although a persistent yellowing was still evident. This highlights the strong protective action of the UV stabilizer, effectively suppresses the formation of byproducts and preserves the polymer's structural integrity even under post-irradiation thermal stress.

The annealing treatment at 165°C, a temperature above the material's glass transition, induced severe thermal degradation in all specimens (Figure 9). Where the non-stabilized PC samples suffered a greater loss of structural integrity across all doses. All specimens exhibited foaming, swelling and wrapping, becoming completely distorted. Even the non-irradiated sample showed deformation, highlighting the material's inherent instability at this temperature.

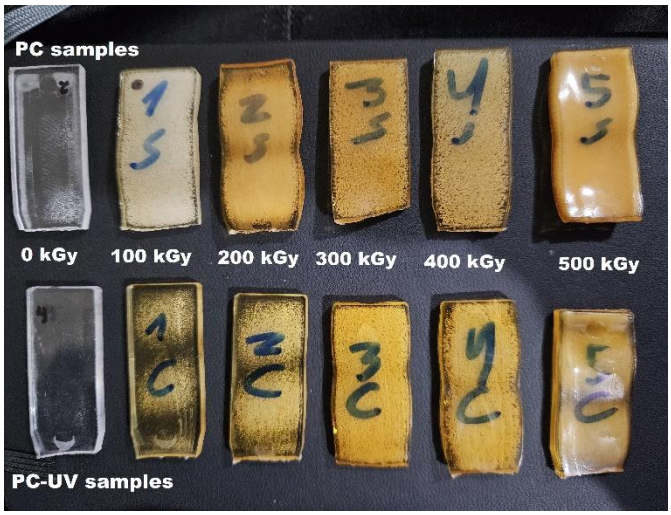

Figure 8: Below glass transition annealing results.

The UV stabilized PC specimens also displayed considerable thermal damage, still, the degradation was less severe. A clear dose-dependent trend of increasing deformation and bubbling was observed. When compared to their non-stabilized counterparts, the PC-UV samples were less swollen and retained more of their original geometry. This finding suggests that the stabilizer provides a residual protective effect, mitigating a little foaming behavior even when the polymer matrix is in a rubbery state.

Figure 9: Above glass transition annealing results.

The annealing experiments provide a visual confirmation of the degradation mechanisms and the stabilizer protective role. Observing foaming and dimensional instability are attributed to the thermal expansion of volatile byproducts, primarily CO and CO_2, which are generated during the radiolytic degradation of the polycarbonate's carbonate group and became trapped within the polymer matrix. (Araújo 1998) (Antonakou 2013) (Nishitsuji 2022) (Wong 2007).

Below the glass transition, the internal pressure caused by these gases was sufficient to overcome the matrix rigidity in the degraded, non-stabilized PC, leading to bubble nucleation and plastic deformation. When heated above the glass transition, the polymer entered a rubbery state with reduced viscosity, offering minimal resistance to the violent expansion of these gases, resulting in greater foaming.

The remarkable stability seen in the PC-UV specimens directly correlates with the DMA findings. By mitigating the initial radiolytic degradation (as evidenced by the smaller reduction in the glass transition), the UV stabilizer suppressed the formation of these gaseous byproducts. With a lower concentration of trapped volatiles, the internal pressure generated during annealing was reduced, thus preserving the material's structural integrity. Therefore, the annealing confirms that the stabilizer's function extends beyond preserving macromolecular weight: it critically enhances post-irradiation thermal stability by inhibiting the generation of volatile degradation products.

5. Conclusions

Based on the results, the main conclusions of this study are:

- Gamma irradiation at doses from 100 to 500 kGy induces significant degradation in both standard and UV-stabilized polycarbonate, evidenced by a progressive yellowing and a reduction in the glass transition temperature due to chain scission.
- The UV stabilizer additive provides a clear protective effect against macromolecular damage. The stabilized PC exhibited a smaller reduction in Tg (a drop of 3.21°C) compared to the standard PC (a drop of 4.24 °C) after a 500 kGy dose, indicating superior preservation of the molecular weight.
- Post-irradiation annealing studies revealed that the degradation mechanism involves the formation of trapped volatiles byproducts, where the unprotected PC suffered severe foaming and catastrophic loss of dimensional integrity upon heating, even below its Tg.
- The UV stabilizer enhances the post-irradiation thermal stability by suppressing the formation of gaseous byproducts, thus preserving structural deformation during subsequent thermal treatments. The protective mechanism of the additive is therefore mitigating chain scission and inhibiting the generation of volatile compounds.

Acknowledgements

The lead author wishes to express his gratitude to the Secretaría de Ciencia, Humanidades, Tecnología e Innovación (SECITHI) for the scholarship awarded for his doctoral studies in the Ph.D. in Materials Science program at the Universidad Autónoma del Estado de México (UAEMex). Furthermore, deep appreciation is extended to the Instituto Nacional de Investigaciones Nucleares (ININ) and the Centro de Tecnología Avanzada (CIATEQ) for their support during research stays and for their assistance with the experimental tests that were fundamental to the completion of this work.

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