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Unraveling the Depth Dose Effects: Implications for Electron Beam Irradiation in Polymer Extrusion

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Abstract

Polymers play a critical role across many industries, from food packaging to healthcare and automotive sectors. However, certain polymers have specific drawbacks that limit their broader application. This necessitates enhanced processing techniques such as electron beam irradiation (EBI), which physically crosslinks polymers and polymer blends to form three dimensional networks. These crosslinked structures are generally considered beneficial because they improve the mechanical strength, thermal stability, durability and many other attributes of the irradiated materials. While EBI offers significant performance benefits, it also presents some challenges. One of these challenges is the so called "depth/dose effect", a phenomenon where electron dosage absorption decreases with depth, leading to non-uniform crosslinking within the material. This manuscript explores the complexities of the depth/dose effect and its impact on polymer properties, particularly in multilayer films and other high-performance applications. Key factors like material composition, geometric configuration, and process variables (e.g., beam voltage and current) are discussed in relation to their impact on the depth/dose effect. The author then presents several strategies to counteract this effect, including using multi pass irradiation, additive incorporation. Overall, the manuscript provides a detailed understanding of how the depth/dose effect works and what can be done to mitigate it so that polymer extrusion processes subjected to electron beam irradiation yield consistent, high-quality products.

Introduction

Polymeric materials play a vital role in numerous key industries, significantly enhancing both functionality and efficiency. In food packaging, for example, polymers are essential for preserving and even extending the shelf life of various products (Tajeddin et al., 2020). In the healthcare sector, these materials are integral to drug delivery systems, medical devices, and pharmaceutical packaging, where they contribute to improved safety and effectiveness (Maitz, 2015). The automotive industry also relies heavily on polymers, which enhance vehicle performance and fuel efficiency (Zhang et al., 2022). Meanwhile, the aerospace sector requires high-performance polymeric materials that are lightweight yet durable enough to endure extreme conditions. Additionally, agriculture, polymers are utilized for greenhouse coverings and crop protection (Sikder et al., 2021). In the construction industry, they provide insulation and structural support, both of which are essential for promoting energy efficiency and sustainability (Shen et al., 2020).

While polymers are utilized in many applications, most are effective only within specific temperature ranges (Brydson, 1999). Polyethylene, which accounts for more than 70% of the global plastics market, functions optimally between -100°C and 120°C. When subjected to elevated temperatures, it begins to soften and loses essential physical properties, which restricts its use in high-temperature scenarios (Samburski et al., 1996). A potential solution to this limitation is the crosslinking of polyethylene through electron beam irradiation (EBI). EBI, a form of ionizing radiation, produces highly reactive species upon interacting with materials, which can modify the molecular structure in various ways. In the case of polymers like polyethylene, EBI primarily promotes branching and crosslinking reactions that enhance the material's stability at higher temperatures. These crosslinking and branching processes increase the polymer's

molecular weight, resulting in an insoluble, threedimensional network, while degradation or scission leads to a reduction in the original molecular weight (Chapiro, 1962).

Crosslinking can generally be achieved through two main techniques: physical crosslinking and chemical crosslinking. Physical crosslinking is commonly carried out using electron beam irradiation, which transforms thermoplastic polymers into thermoset forms, creating a more durable and non-melting polymer network. Different types of polyethylene, including linear low-density polyethylene (LLDPE), low-density polyethylene (LDPE), high-density polyethylene (HDPE), ethylene vinyl acetate (EVA), and polyolefin elastomers (POE), are often crosslinked using this method (Singh et al., 2003). Additionally, thermoplastic elastomers like styrenic copolymers can also be crosslinked physically. These polymers exhibit both thermoplastic and elastomeric properties, forming stable structures through mechanisms like crystallization or phase separation (Maji et al., 2022). Hydrogels made from polyethylene glycol (PEG) and its derivatives are developed through hydrogen bonding, making them particularly useful in biomedical applications (Sun et al., 2023). Polyvinyl alcohol (PVA) similarly forms crosslinked networks in water-based environments via hydrogen bonding (Sau et al., 2021). Polyacrylic acid (PAA) can undergo physical crosslinking in aqueous systems, forming hydrogels that are promising for drug delivery applications (Thang et al., 2023). Overall, physical crosslinking enhances the strength, elasticity, and thermal stability of these polymers while maintaining their processability.

Chemical crosslinking, on the other hand, involves establishing covalent bonds between polymer chains, leading to a permanent network structure that boosts both strength and thermal stability, typically achieved through specific chemical agents or reactions (Tillet et al., 2011). In contrast, physical crosslinking depends on non-covalent interactions like hydrogen bonding, ionic interactions, or Van der Waals forces. These interactions are often reversible, allowing the material to return to its original form when conditions, such as temperature, change. While chemical crosslinking provides increased durability, physical crosslinking offers greater flexibility and ease of processing. This discussion will focus solely on physical crosslinking.

Electron beam irradiation (EBI) plays an essential role in the processing of polymers, greatly improving the performance of various materials. For instance, blends of crosslinked HDPE-PU-EVA exhibit remarkable thermal stability, demonstrating four times the stability of non-crosslinked HDPE after one hour at 180 °C (Lee et al., 2021). By focusing a beam of high-energy electrons onto a substrate, EBI facilitates crosslinking within the polymer's structure. This crosslinking is vital for enhancing attributes such as mechanical strength, thermal stability, and barrier performance, especially in applications like packaging and medical devices (Garavand et al., 2017; Chmielewski et al., 2005). The extent of crosslinking can vary significantly based on several factors, including the type of polymer, the specific application, and the degree of branching present. Another important consideration is the "depth/dose effect," which describes the uneven absorption of electron dosage throughout the thickness of the irradiated material. Although this phenomenon is significant, it has not received much attention in the field of polymer processing. This manuscript intends to fill this gap by examining key aspects of the depth/dose effect and discussing how polymer processing can address its challenges.

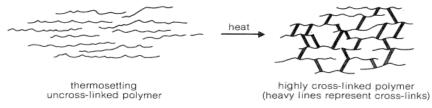


Figure 1: Schematic representation of the conversion of an uncross-linked thermosetting polymer to a highly cross-linked polymer. The cross-links are shown in a two-dimensional network, but in practice three-dimensional networks are formed (Roberts and Caserio (1977)).

The depth/dose effect leads to variations in electron dosage across different layers of a polymer product, resulting in unequal crosslinking throughout the material. As energetic electrons penetrate matter, they interact with atoms, losing kinetic energy due to Coulomb interactions and ultimately stopping at a certain depth known as the electron range. This variation can significantly affect the final properties of the product, such as its sealability, durability, and overall functionality (Singh et al., 2003). For instance, polymers with different chemical structures may display varying degrees of crosslinking, resulting in inconsistencies in product quality (Hirschl et al., 2013).

The depth/dose effect has long been recognized as a challenge in electron beam processing. To counteract its effects, manufacturers frequently employ multipass irradiation, where the polymer material is exposed to the electron beam multiple times. This technique aims to achieve a more uniform distribution of dosage, thus reducing the issues linked to uneven crosslinking (Chaudhary et al., 2017). By subjecting the material to multiple passes, manufacturers can better control the dose received by both surface and interior layers, which is crucial for thicker or more complex materials. For example, utilizing two-pass or four-pass configurations can enhance the stability of the irradiation process, improving control over the depth/dose profile and ultimately enhancing product consistency. However, there is a lack of literature discussing how these configurations can optimize the irradiation process, which this manuscript seeks to address.

In polymer extrusion, especially with multilayer films, the depth/dose effect has significant implications. The multilayer film structure adds complexity during irradiation since each layer may respond differently to the electron beam due to variations in thickness, polymer chemistry, and the presence of additives (Ashfaq et al., 2020). A thorough understanding of these interactions is essential for optimizing the EBI process and ensuring that the final product meets its specifications.

Additionally, the depth/dose effect has important implications for product design and process

development. For instance, excessive crosslinking in specific layers can compromise critical properties like sealability, which is especially important in packaging applications (Tamboli et al., 2017). Consequently, careful attention must be paid to polymer formulation and processing parameters to achieve the desired balance of properties.

Recent research has suggested various strategies for managing the depth/dose effect during electron beam irradiation. One such approach involves incorporating additives that can influence the crosslinking behavior of polymers. For example, antioxidants can help mitigate excessive crosslinking, preserving the essential properties of the material (Lai, W. F., 2021). Furthermore, modifying processing parameters such as beam current, voltage, and pass configuration can substantially affect the depth/dose profile, allowing for more customized product outcomes (Ashfaq et al., 2020).

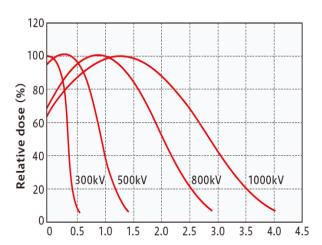
In summary, the depth/dose effect is a critical consideration in the electron beam irradiation of polymer materials. Its impact extends to multiple aspects of product performance, particularly in multilayer films and packaging. As the industry advances, gaining a deeper understanding of this phenomenon will enable manufacturers to optimize their processes and create high-performance materials that meet the requirements of various applications. This manuscript aims to provide valuable insights into the depth/dose effect, ultimately facilitating improved product design and process enhancements in polymer extrusion.

Overview of depth/dose effect

In the process of electron beam irradiation, the amount of electron dosage that materials absorb varies with depth. This variation is a key aspect of how electrons interact with different substances. As electrons move through a material, they lose energy primarily through ionization and scattering, resulting in a specific depth/dose profile (Chmielewski et al., 2005; Singh et al., 2003). Typically, the dosage is highest at the surface and decreases as you go deeper into the material. Several factors influence this decrease, including the energy of the electrons, the density and composition of the material, and the particular

conditions under which irradiation occurs (Ashfaq et al., 2020).

The depth/dose master curve (see Figure 2 below) demonstrates the correlation between penetration depth and the percentage of absorbed electrons, illustrating how various conditions can affect dosage distribution. The vertical axis of this curve represents the relative dose, which indicates the amount of energy absorbed by the material relative to other points, while the horizontal axis shows the penetration depth into a material with a specific gravity of 1. In general, higher energy electrons can penetrate further, but the dosage still declines with increased depth. This variation can lead to notable differences in the physical properties of the material throughout its thickness (Feng et al., 2020).



Penetration depth into specific gravity 1 material (mm)

Figure 2 Depth Dose curve for a hypothetical material with specific gravity 1

For example, when a material with a specific gravity of 1 undergoes irradiation at an acceleration voltage of 300 kV, the surface dose (indicated at 0 mm on the horizontal axis) is nearly 100%. As the electrons penetrate deeper into the material, this dose decreases significantly, falling below 10% at a depth of 0.5 mm from the surface. Generally, the penetration capacity for a material with a specific gravity (ρ) is inversely related to its density, expressed as $1/\rho$. Thus, materials with higher specific gravities are less penetrable by electrons, while those with lower specific gravities allow for greater penetration.

For instance, if a material has a specific gravity of 2, the penetration depth indicated on the horizontal axis would be reduced by half. Conversely, a material with a specific gravity of 0.5 would effectively double the penetration capacity. Additionally, as the acceleration voltage increases, the Depth-Dose Curve usually shows a peak in relative dose at a slightly deeper level than the surface. This effect arises because higher-speed electrons interact with the material too quickly, passing through it before delivering sufficient energy to induce effective crosslinking.

The depth/dose effect has been acknowledged for many years, prompting the development of multi-pass irradiation techniques (Chmielewski et al., 2005). In these methods, materials are subjected to the electron beam multiple times, promoting a more uniform dosage distribution throughout their thickness. Multipass operations are particularly vital for applications that require consistent material properties, such as in medical device manufacturing, packaging films, and advanced polymer composites.

While the depth/dose effect can present challenges, it is not necessarily detrimental. Gaining a thorough understanding of this effect is crucial for optimizing product performance. It enables manufacturers to customize the irradiation process to enhance desired attributes while addressing potential issues, such as decreased mechanical strength or thermal stability.

Factors Influencing the Depth/Dose Effect: Various elements contribute to the complexity of the electron beam irradiation process and its related depth/dose effect:

1. **Material Composition:** Different polymers exhibit distinct responses to electron irradiation. For example, ethylene-vinyl acetate (EVA) tends to undergo crosslinking more easily than linear low-density polyethylene (LLDPE) when both are exposed to the same irradiation conditions (Azizi et al., 2019). Additionally, the presence of additives within the material can influence the extent of crosslinking, making it more challenging to achieve uniform dosage distribution.

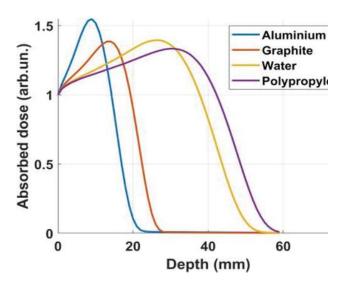


Figure 3 Depth dose distribution in aluminum, graphite, water, and polypropylene irradiated with 10 MeV electrons. (Bliznyuk et al., 2023)

2. Geometric Configuration: The shape of materials being irradiated—commonly in the form of tubular films or layers—presents various challenges. Ideal shapes, such as parallelepipeds, facilitate perpendicular electron penetration, resulting in a predictable dose distribution. In contrast, objects with more complex geometries, like spheres, ellipsoids, or cylinders, create irregular dose distributions due to non-perpendicular electron interactions. Simulations conducted with 10 MeV electrons indicate that parallelepiped shapes experience a sharp decline in dose after a certain depth, leading to underexposure at their edges. Conversely, spheres often show overexposure at their equatorial regions, while cylinders tend to exhibit overexposure on their lateral surfaces, although their depth-dose distribution remains comparable to that of parallelepipeds. Overall, factors such as electron energy, object shape, density, and chemical composition significantly influence the distribution of absorbed doses, necessitating tailored strategies for irradiating geometrically complex objects. Additionally, issues related to gas accumulation can arise; gases generated during irradiation may become trapped within these structures, potentially causing inconsistencies in dose delivery and resulting in defects in the material.

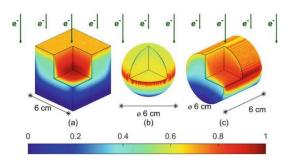


Figure 4 3D-color map of absorbed dose distribution throughout the water phantoms: (a) parallelepiped with the edge of 6 cm, (b) sphere with 6 cm diameter, and (c) cylinder with the height of 6 cm during unilateral 10 MeV electron irradiation (Bliznyuk et al., 2023).

3. **Process Variables:** The details of the irradiation setup, including factors such as the electron beam voltage, current (amperage), configuration of the irradiation unit, the material's path through the beam, material thickness, and overall dosage, all play critical roles in shaping the depth/dose profile. Changes in voltage can significantly affect both the depth of penetration and the uniformity of the dosage received. Moreover, the way the material is aligned as it moves through the electron beam can add further variability to the process. This manuscript will thoroughly explore how these process variables impact the depth/dose profile.

Grasping the depth/dose effect is crucial for effective product design and process development, as it can lead to variations in material properties such as mechanical flexibility. and thermal resistance. strength. Manufacturers need to take this effect into account when creating multilayer products, since different doses across layers can significantly influence overall performance. In critical applications such as packaging and medical devices, it is essential to mitigate the depth/dose effect. Possible strategies include refining material formulations, implementing multi-pass irradiation, and fine-tuning irradiation parameters to achieve more uniform dosages.

Current research focuses on incorporating additives that help stabilize material properties during and after irradiation, which is vital for maintaining product integrity over time and addressing the challenges associated with the depth/dose effect. Understanding

this phenomenon is fundamental to the process of electron beam irradiation, as it affects material performance. By strategically modifying processing parameters and material designs, manufacturers can capitalize on the advantages of electron beam irradiation while minimizing its drawbacks, ultimately resulting in higher-quality materials for a wide range of applications. Addressing the depth/dose effect will

remain a priority as technology advances and the demand for high-performance materials grows.

Units, Conversion Factors, and Basic Physics of Irradiation: In the industry, several colloquial terms have emerged to describe the irradiation process. To clarify some key concepts, Tables 1 below present information and SI units for many relevant variables.

Parameter	Unit	Description	
Energy	MeV	Mega-electron Volts, a unit of energy commonly used for electrons.	
Dose	Gy	Gray, which represents the absorption of one joule of radiation energy per kilogram.	
Current	mA	Milliamperes, a measure of the electric current used in the electron beam.	
Voltage	kV	Kilovolts, the potential difference used to accelerate electrons.	
Time	S	Seconds, duration of irradiation exposure.	
Penetration Depth	mm	Millimeters, the depth to which electrons can penetrate a material.	

Table 1: Key Units and Conversion Factors in Electron Beam Irradiation

In the context of electron beam irradiation, the energy level (expressed in MeV) is a crucial factor as it affects both the depth of penetration and the distribution of dose within the material. Generally, higher electron energy allows for deeper penetration; however, this energy also influences how electrons interact with the material, ultimately impacting the extent of crosslinking achieved.

The dose, measured in grays (Gy), is another critical element, directly linked to the material's response during irradiation. This measurement indicates the amount of energy deposited within the material, which subsequently affects its physical and chemical characteristics.

Additionally, current (in milliamperes) and voltage (in kilovolts) are significant in determining the efficiency and properties of the electron beam. The current controls the rate at which electrons are emitted, while the voltage determines the energy level of the electrons, thereby influencing their ability to penetrate the material effectively.

A basic understanding of physics further clarifies the principles at play. The core concept behind electron beam irradiation involves the interaction of highenergy electrons with matter. When these electrons collide with the atoms in the polymer, they can ionize these atoms, breaking molecular bonds and generating free radicals. These free radicals can subsequently react with adjacent polymer chains, resulting in crosslinking.

The degree of crosslinking is contingent on both the electron dose and the composition of the material. Different polymers respond uniquely to irradiation, dictated by their chemical structure. For instance, polymers with higher unsaturation levels or specific functional groups may undergo reactions more readily, leading to increased crosslinking.

Comprehending these dynamics allows researchers and manufacturers to optimize the irradiation process to achieve the desired material properties while mitigating negative outcomes, such as brittleness or reduced flexibility. Table 2 provides various constants relevant to electron interactions.

Quantity	Symbol	Value
electron charge	e	1.6022 x 10 ⁻¹⁹ C
electron mass	m_e	9.1095 x 10 ⁻³¹ kg
electron-volt	eV	1.6022 x 10 ⁻¹⁹ J

Table 2: Basic physical constants for an electron

Incorporating electron beam irradiation technology into polymer processing presents considerable potential for improving material performance across a range of applications. A deep understanding of the intricacies of the depth/dose effect, operational parameters, and the fundamental physics of electron interactions is essential for stakeholders aiming to optimize processes and produce consistent, high-quality products. As the field continues to evolve, ongoing research and development efforts will be vital for overcoming challenges and maximizing the advantages of electron beam irradiation in various polymer applications.

This manuscript aims to provide a thorough overview of the impact of electron beam irradiation on polymer processing, serving as a valuable guide for future innovations and advancements in the industry.

Impact of Various Factors on Depth/Dose Profiles

In electron beam irradiation (EBI), the depth/dose profile is affected by several important factors that dictate the uniformity and effectiveness of crosslinking within the material. A comprehensive understanding of these factors is crucial for optimizing irradiation processes and attaining the desired properties in the final product. For the purpose of this discussion, we will focus on materials configured as tubular films or multilayers; however, these factors similarly impact other geometric configurations.

1. Beam Voltage

The voltage applied in the electron crosslinking unit (ECLU) is a critical factor that influences the energy

of the electrons generated during the process. The necessary energy levels for electron beam applications depend significantly on the thickness and density of the material being treated. Electron accelerators can be classified into three categories based on energy levels: low (80 to 300 keV), medium (300 to 1000 keV), and high (1 to 10 MeV).

Low-energy electron beam accelerators are generally utilized for the polymerization and crosslinking of thin films, plastic laminates, and single-strand wires. In contrast, medium-energy systems are often used for crosslinking wire insulation, heat-shrinkable products, and for achieving partial crosslinking in tire components.

Higher voltages result in increased kinetic energy of the electrons, allowing for deeper penetration into the material. However, this deeper penetration creates a steeper dosage gradient, where the surface layers receive a significantly higher dose compared to the interior, potentially leading to uneven crosslinking. Consequently, while the outer layers may demonstrate improved mechanical properties, the inner layers could remain inadequately crosslinked. Conversely, lower voltages can facilitate a more uniform crosslinking profile at shallower depths.

Thus, it is crucial to find a balance in voltage levels tailored to the material's thickness and the desired properties (Ashfaq et al., 2020). Manufacturers must judiciously select the ECLU voltage to optimize the irradiation effects based on specific application requirements.

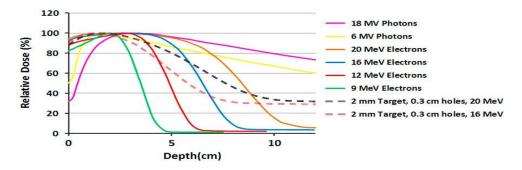


Figure 5 Typical depth-dose distribution curves for different particles and different beam energies (Khaledi et al., 2018)

To establish depth-dose curves, manufacturers and researchers typically employ dosimetry techniques. (Note that dosimetry is a complex field that will not be addressed in detail here.) A common practice involves applying a thin layer of standard polyethylene resin on top of the actual polymer as it passes through the electron crosslinking unit (ECLU). This approach allows for dose measurements to be taken from the standard polyethylene rather than the polymer itself.

The dose is usually assessed using Fourier Transform Infrared Spectroscopy (FTIR), which is regarded as a reliable and accurate method. The resulting depth-dose master curves, illustrated in Figure 3, can be further analyzed by fitting the data to a polynomial equation, with the coefficients determined through regression analysis of the experimental results.

$$D = N \cdot t^5 + M \cdot t^4 + I \cdot t^3 + J \cdot t^2 + K \cdot t + L$$

Equation 1: Polynomial fit for master curves where "D" represents the relative dose and "t" denotes the depth or thickness.

As indicated in Figure 3, higher voltages enable electrons to penetrate deeper before being fully absorbed. This increased penetration is a function of the electron energy, which dictates the maximum depth achievable. By utilizing the master curve across various voltage levels, it is possible to generate a graph that illustrates the relationship between extinguishing depth and voltage. The figure reveals that as voltage rises, the relative depth also increases, but at a diminishing rate. In other words, the rate at which relative depth changes in relation to voltage decreases as the voltage continues to increase.

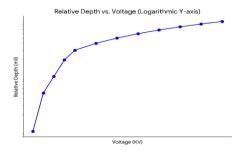


Figure 5 Curve showing the relationship of Depthdose distribution as a function of voltage

2. Number of Passes Through the Electron Beam Unit

The setup of the electron beam unit, especially the number of times the substrate passes through the irradiation zone, plays a crucial role in shaping the depth/dose profile. Multi-pass operations, such as twopass or four-pass configurations, are often utilized to improve the uniformity of dose distribution. Each pass exposes the material to additional electron dosage, helping to mitigate the variations caused by the depth/dose effect. For instance, a four-pass approach allows for adequate irradiation to penetrate deeper layers, reducing the risk of under-crosslinking and ensuring all layers develop the required mechanical properties. However, a key challenge is to prevent over-crosslinking in the surface layers, which can adversely affect properties like flexibility and sealability (Chmielewski et al., 2005). Thus, manufacturers must carefully weigh the trade-offs between the number of passes and the resultant material characteristics to fine-tune their processes for specific applications.

Increasing the number of passes through the electron crosslinking unit (ECLU) adds complexity to tracking the material, but it generally results in a more uniform dose profile, ensuring that all layers of the material (especially in multilayer films) receive a consistent absorbed dose. Moreover, multiple passes can necessitate lower beam current to achieve the same dosage compared to a single pass, making this approach advantageous despite its intricacies.

A fundamental decision regarding the depth/dose effect during the ECLU process is whether to employ a fewer-pass or multiple-pass mode. While the fewer-pass mode offers increased stability, it often leads to a non-uniform depth/dose profile, where higher doses in inner layers can compromise attributes like heat sealability. Several strategies can help address this issue. For example, incorporating antioxidants in the inner layer can bolster its resistance to crosslinking, allowing it to tolerate higher doses more effectively. Alternatively, increasing the melt index (MI) of the inner layer can enhance its flowability after irradiation. Another potential solution involves adding a crosslink enhancer to the outer layers to compensate

for the reduced doses they receive, ensuring effective crosslinking even at lower overall doses; unsaturated polymers like EPDMs are particularly well-suited for this purpose. Additionally, adjusting the ECLU voltage can also affect the depth/dose profile—higher voltage typically yields better uniformity, while lower voltage can prevent deep penetration into the inner layers. Each of these approaches provides a means to optimize the ECLU process while addressing the challenges associated with the depth/dose effect.

3. Thickness (t) and density

Material thickness and density critical determinants affecting the depth/dose profile during electron beam irradiation. When polymer samples of varying thicknesses are exposed to the same dose and dose rate in air, they demonstrate different behaviors regarding crosslinking and oxidation degradation. Thicker or denser materials often present challenges for achieving uniform electron penetration, as the outer layers tend to absorb more energy from the electrons than the inner layers. This discrepancy results in a non-uniform crosslinking profile, where the surface layers may exhibit properties that significantly differ from those of the core.

In the case of multilayer films, the interaction between thickness and the distinct chemistries of different polymers can further complicate the irradiation process. For instance, if an outer layer is considerably thicker than the inner layers, it may dominate the overall crosslinking characteristics, thereby affecting the performance of the entire material structure. Therefore, understanding how the thickness of each layer interacts with the electron beam is essential for achieving an optimal balance of mechanical, thermal, and barrier properties. In this report, the effects of density are simplified for clarity. While this may lead to some quantitative inaccuracies, the qualitative conclusions will still hold true.

Figure 4 illustrates the results from infrared (IR) analysis and solubility tests conducted on linear low-density polyethylene (LLDPE) irradiated in air. Uniform sheets of the polymer were microtomed, allowing for the assessment of oxidation reactions through the measurement of carbonyl group absorption peaks. The study also evaluated changes in

molecular weight via gel extraction tests, analyzing both carbonyl concentration and gel fraction profiles in relation to the distance from the polymer's surface (Sun et al., 2017).

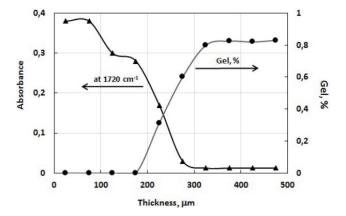


Figure 6 Carbonyl concentration and gel fractions profiles for a LLDPE sample irradiated (Sun et al., 2017)

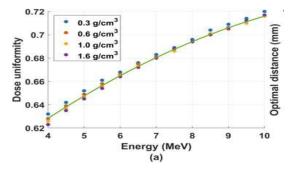


Figure 7 Dependency of the dose uniformity in water parallelepiped with the density ranging from 0.3 g/cm3 to 1.6 g/cm3 on the electron energy and the function approximating the calculated dependency (Bliznyuk et al., 2023).

4. Beam Current (i)

The beam current, which refers to the flow of electrons during the irradiation process, plays a crucial role in shaping the depth/dose profile. Increased beam current raises the radiation intensity, which can improve the efficiency of crosslinking. However, if the current is set too high, it can result in localized overheating, which poses a risk of thermal degradation for sensitive polymers. This degradation not only alters the material's immediate properties but can also create inconsistencies in the depth/dose profile. Specifically,

surface layers might absorb excessively high doses, while deeper sections may not receive sufficient irradiation. Therefore, it is vital for manufacturers to carefully calibrate the beam current to enhance crosslinking while safeguarding the material's integrity (Garavand et al., 2017).

5. Absorbed Dose (D)

The absorbed dose, expressed in Grays (Gy), quantifies the energy delivered to a material by electrons during irradiation, effectively measuring the interaction of electrons with the material. To control the irradiation process, it is important to establish a clear relationship between source parameters—including beam voltage, current, scanning width, uniformity, and conveyor speed—and the absorbed dose experienced by the material. Accurate measurements of absorbed dose and its distribution are typically obtained through a reliable dosimetry system, which must provide a certain level of precision.

This absorbed dose is a vital factor, as it directly affects the extent of crosslinking and the resulting properties of the polymer. By adjusting the absorbed dose, manufacturers can optimize the characteristics of the material to meet specific processing needs, enhancing attributes such as mechanical strength, thermal stability, or barrier performance.

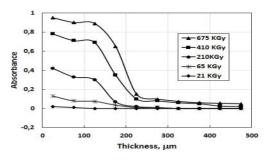


Figure 6 Carbonyl concentration profiles for samples of LLDPE irradiated in air at the constant dose rate of 1 kGy/h and different total absorbed doses (Sun et al., 2017).

6. Dose rate

While absorbed dose measures the total energy that a material takes in, dose rate refers to how quickly this energy is delivered, mathematically represented as the change in absorbed dose over time (dD/dt). Understanding dose rate is essential in materials processing. When dose rates are high, the abundance of free radicals generated can deplete the oxygen in the immediate vicinity of the surface quickly, minimizing oxidative degradation to just the outer layers. On the other hand, lower dose rates facilitate more oxygen diffusion into the material, resulting in a thicker oxidized layer.

As a result, the effects of irradiation on a given polymer can vary considerably based on the dose rate used. High dose rates primarily enhance crosslinking reactions, similar to what is seen in vacuum or inert environments. Conversely, lower dose rates tend to lead to degradation effects that penetrate more deeply into the irradiated material.

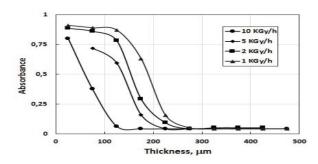


Figure 7 Carbonyl concentration profiles for samples of LLDPE irradiated in air at the total absorbed dose of 675 kGy and different dose rates (Sun et al., 2017)

7. Speed of Processing (v)

The speed at which materials traverse the electron beam unit plays a crucial role in determining the depth/dose profile. When materials move quickly through the beam, the overall exposure time is reduced, which can lead to lower doses throughout the thickness of the material. This reduced exposure may result in inadequate crosslinking, particularly in thicker sections that require prolonged exposure to achieve the desired crosslinking level. On the other hand, slower processing speeds can lead to more consistent irradiation but may also cause overheating or excessive crosslinking in surface layers, adversely affecting important properties such as flexibility and sealability. Therefore, finding the optimal processing speed is vital for balancing efficiency and effective crosslinking, making it a key factor in the design of

electron beam irradiation (EBI) processes (Tamboli et al., 2017).

Empirical relationship: Numerous factors influence the dose, as outlined above. However, developing a comprehensive empirical equation that encompasses all variables would result in a complicated formula necessitating a "correction factor," which would offer limited practical use. Fortunately, advancements in simulation and modeling techniques are enhancing the optimization of the electron beam irradiation process. Computational methods can now predict depth/dose profiles and evaluate how various processing conditions might affect final product properties (Nordlund et al., 2019). By utilizing these tools, manufacturers gain valuable insights into the complexities of the EBI process, allowing for more informed decisions regarding product design and process refinement.

For practical purposes, we will focus on three primary variables that significantly impact dosage (D): current (i), line speed (v), and thickness (or a function of thickness). Their interrelationships can be summarized as follows:

$$D \propto i \times \frac{1}{v} \times \frac{1}{f(t)}$$

Equation 2: Factors affecting average dose

Current directly correlates with dose, while line speed has an inverse relationship. This understanding can be synthesized into a more effective primary variable for dosage control: the ratio of current to line speed, known as the ECLU algorithm coefficient, represented by the symbol ' θ ' (theta) (= i/v). Some newer ECLU units also display a "product factor," defined as $\theta \times 100$.

$$\mathbf{D} \propto \mathbf{\theta} \times \mathbf{f}(\mathbf{t})$$

Equation 3: Average Dose vs. ECLU algorithm coefficient (θ)

Over time, dosage data can be collected alongside beam current and line speed (which yields θ) and utilized through linear regression to establish a calibration curve of dose versus θ . This process assumes constant thickness, enabling the extraction of

the slope (m) and intercept (b) from the regression analysis. It's important to note that achieving a strong correlation requires a wide range of values for $\theta.$ For instance, in units of {mA/fpm}, a suitable range might be $0.05 \leq \theta \leq 0.20$ for a four-pass setup and $0.10 \leq \theta \leq 0.30$ for a two-pass configuration. A narrow range can result in excessive vertical scatter compared to the horizontal scale, leading to a lower regression 'R value.'

Conclusion

In summary, the depth/dose profile in electron beam irradiation (EBI) is affected by a variety of interrelated factors, including beam voltage, number of passes through the irradiation zone, material thickness and density, beam current, absorbed dose, dose rate, and processing speed. A thorough understanding of these elements is essential for optimizing the irradiation process to achieve consistent crosslinking and the desired properties of materials.

Beam voltage is crucial as it influences electron penetration and the distribution of dose, making its careful selection essential for balancing the characteristics of surface and internal layers. Utilizing multi-pass setups can improve the uniformity of dose distribution; however, this approach also introduces challenges that must be managed to avoid excessive crosslinking. Additionally, the thickness and density of materials complicate the situation, as thicker or denser samples may lead to uneven crosslinking profiles.

The efficiency of crosslinking is also directly impacted by beam current and absorbed dose, while the rate of dose delivery affects oxidative degradation within the material. Furthermore, the speed of processing is a key factor in determining exposure time, with slower speeds contributing to uniformity but risking overheating or over-crosslinking in surface layers.

The relevance of electron beam irradiation in polymer processing highlights its role as a versatile technology that enhances material performance across diverse applications. As industries pursue innovative approaches to improve product quality and durability, EBI continues to lead advancements in materials science and engineering. Ongoing exploration of its potential applications is likely to yield new

breakthroughs, reinforcing its significance in the future of polymer processing.

To assist manufacturers in managing these complexities, the use of computational modeling and simulation techniques is becoming more prevalent, enabling the prediction of depth/dose profiles and the evaluation of various processing conditions. By concentrating on essential variables like current, line speed, and thickness, and employing the derived ECLU algorithm coefficient, manufacturers can refine dosage control for specific applications, ultimately enhancing the performance and quality of end products. A holistic understanding of these factors will facilitate the creation of customized EBI processes that address a range of industrial requirements while ensuring material integrity and performance.

References

- Tajeddin, B., & Arabkhedri, M. (2020). Polymers and food packaging. In M. A. AlMaadeed, D. Ponnamma, & M. A. Carignano (Eds.), Polymer Science and Innovative Applications (pp. 525-543). Elsevier. https://doi.org/10.1016/B978-0-12-816808-0.00016-0
- Maitz, M. F. (2015). Applications of synthetic polymers in clinical medicine. Biosurface and Biotribology, 1(3), 161-176. https://doi.org/10.1016/j.bsbt.2015.08.002
- Shen, J., Liang, J., Lin, X., Lin, H., Yu, J., & Yang, Z. (2020). Recent progress in polymer-based building materials. International Journal of Polymer Science, 2020, 1-15. https://doi.org/10.1155/2020/8838160
- Zhang, W., & Xu, J. (2022). Advanced lightweight materials for automobiles: A review. Materials & Design, 221, 110994. https://doi.org/10.1016/j.matdes.2022.110994
- Parveez, B., Kittur, M. I., Badruddin, I. A., Kamangar, S., Hussien, M., & Umarfarooq, M. A. (2022). Scientific advancements in composite materials for aircraft applications: A review. Polymers, 14(22), 5007. https://doi.org/10.3390/polym14225007
- Sikder, A., Pearce, A. K., Parkinson, S. J., Napier, R., & O'Reilly, R. K. (2021). Recent trends in advanced polymer materials in agriculture-

- related applications. ACS Applied Polymer Materials, 3(3), 1203-1217. https://doi.org/10.1021/acsapm.0c00982
- 7. Brydson, J.A. (1999). *Plastics Materials* (7th ed.). Butterworth-Heinemann.
- 8. Samburski, G., Narkis, M., & Siegmann, A. (1996). Structure and properties of peroxide crosslinked polyethylene tubing after drawing. *Journal of Macromolecular Science, Part B*, 35, 843-862.

https://doi.org/10.1080/00222349608220411

- Chapiro, A. (1962). Radiation chemistry of polymeric systems. New York: Interscience Publishers.
- Tamboli, S. M., Mhaske, S. T., & Kale, D. D. (2004). Crosslinked polyethylene. *Indian Journal of Chemical Technology*. https://doi.org/10.1142/9781783267170
- Garavand, F., Rouhi, M., Razavi, S. H., Cacciotti, I., & Mohammadi, R. (2017). Improving the integrity of natural biopolymer films used in food packaging by crosslinking approach: A review. *International Journal of Biological Macromolecules*, 104(A), 687-707. https://doi.org/10.1016/j.ijbiomac.2017.06.093
- 12. Chmielewski, A. G., Haji-Saeid, M., & Ahmed, S. (2005). Progress in radiation processing of polymers. *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, 236(1–4), 44-54. https://doi.org/10.1016/j.nimb.2005.03.247
- Bliznyuk, U., Chernyaev, A., Ipatova, V., Nikitchenko, A., Studenikin, F., & Zolotov, S. (2023). Electron Beam Processing of Biological Objects and Materials. IntechOpen. https://doi.org/10.5772/intechopen.112699
- Singh, A., & Bahari, K. (2003). Use of highenergy radiation in polymer blends technology. In L. A. Utracki (Ed.), *Polymer blends handbook* (pp. 1-12). Springer, Dordrecht. https://doi.org/10.1007/0-306-48244-4 11
- Maji, P., & Naskar, K. (2022). Styrenic block copolymer-based thermoplastic elastomers in smart applications: Advances in synthesis, microstructure, and structure-property relationships—A review. *Journal of Applied*

Polymer Science, 139, 10.1002/app.52942. https://doi.org/10.1002/app.52942

- Sun, S., Cui, Y., Yuan, B., Dou, M., Wang, G., Xu, H., Wang, J., Yin, W., Wu, D., & Peng, C. (2023). Drug delivery systems based on polyethylene glycol hydrogels for enhanced bone regeneration. *Frontiers in Bioengineering and Biotechnology*, 11, 1117647. https://doi.org/10.3389/fbioe.2023.1117647. PMID: 36793443; PMCID: PMC9923112.
- Sau, S., Pandit, S., & Kundu, S. (2021). Crosslinked poly(vinyl alcohol): Structural, optical, and mechanical properties. *Surfaces and Interfaces*, 25, 101198. https://doi.org/10.1016/j.surfin.2021.101198.
- 18. Thang, N. H., Chien, T. B., & Cuong, D. X. (2023). Polymer-based hydrogels applied in drug delivery: An overview. *Gels*, 9(7), 523. https://doi.org/10.3390/gels9070523.
- 19. Tillet, G., Boutevin, B., & Ameduri, B. (2011). Chemical reactions of polymer crosslinking and post-crosslinking at room and medium temperature. *Progress in Polymer Science*, 36(2), 191-217. https://doi.org/10.1016/j.progpolymsci.2010.08. 003.
- Lee, J.G., Jeong, J.O., Jeong, S.I., & Park, J.S. (2021). Radiation-based crosslinking technique for enhanced thermal and mechanical properties of HDPE/EVA/PU blends. *Polymers (Basel)*, 13(16), 2832. https://doi.org/10.3390/polym13162832
- Khaledi, N., Sardari, D., Mohammadi, M., Ameri, A., & Reynaert, N. (2018). Dosimetric evaluation of a novel electron-photon mixed beam produced by a medical linear accelerator. *Journal of Radiotherapy in Practice*, 17(3), 319-331.

https://doi.org/10.1017/S1460396917000711

- Sun, Y., & Chmielewski, A. G. (Eds.). (2017).
 Applications of ionizing radiation in materials processing (Vol. 2). Institute of Nuclear Chemistry and Technology. Warszawa.
- Hirschl, C., Biebl-Rydlo, M., DeBiasio, M., Mühleisen, W., Neumaier, L., Scherf, W., Oreski, G., Eder, G., Chernev, B., Schwab, W., & Kraft, M. (2013). Determining the degree of

- crosslinking of ethylene vinyl acetate photovoltaic module encapsulants—A comparative study. Solar Energy Materials and Solar Cells, 116, 203-218. https://doi.org/10.1016/j.solmat.2013.04.022
- Chaudhary, N., Koiry, S. P., Singh, A., Tillu, A. R., Jha, P., Samanta, S., Debnath, A. K., Aswal, D. K., Mondal, R. K., Acharya, S., & Mittal, K. C. (2017). Electron beam induced modifications in flexible biaxially oriented polyethylene terephthalate sheets: Improved mechanical and electrical properties. *Materials Chemistry and Physics*, 189, 237-244. https://doi.org/10.1016/j.matchemphys.2016.12.054
- Ashfaq, A., Clochard, M. C., Coqueret, X., Dispenza, C., Driscoll, M. S., Ulański, P., & Al-Sheikhly, M. (2020). Polymerization reactions and modifications of polymers by ionizing radiation. *Polymers (Basel)*, 12(12), 2877. https://doi.org/10.3390/polym12122877
- Lai, W. F. (2021). Design of polymeric films for antioxidant active food packaging. *International Journal of Molecular Sciences*, 23(1), 12. https://doi.org/10.3390/ijms23010012
- Nordlund, K. (2019). Historical review of computer simulation of radiation effects in materials. *Journal of Nuclear Materials*, 520, 273-295.

https://doi.org/10.1016/j.jnucmat.2019.04.028

28. Azizi, S., Ouellet-Plamondon, C. M., Nguyen-Tri, P., Fréchette, M., & David, E. (2019). Electrical, thermal and rheological properties of low-density polyethylene/ethylene vinyl acetate/graphene-like composite. *Composites Part B: Engineering, 177*, 107288. https://doi.org/10.1016/j.compositesb.2019.1072