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Modified Surface Polymer Prepared by using Radiation-Induced Grafting Co-polymerization Method and Its Application: A Mini Review

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Abstract. The use of radiation-induced grafting (RIG) polymerization techniques is an appealing way to create and develop polymerization of polymer. The method of copolymerization preparation was evaluated in this paper based on the types of radiation-induced grafting polymerization and their prospective applications. Based on the method of preparation, the optimum grafting yield of the monomer in the polymer backbone is described. The approach used to summarise this review was to go through polymer-related papers from the science direct online database from 2010 to 2021. Then, from those selected journals, the technique of preparation, the grafting yield, and the application were reviewed. Furthermore, the majority of researchers from reviewed journals employed gamma irradiation to prepare the modified polymer, followed by electron beam irradiation, plasma irradiation and UV irradiation. Gamma irradiation is popular because it has a better penetration rate and generates a purer result. As a result of this review paper, the radiation-induced graft polymer from the conducted study is ideal for use in biomedical applications, as a material for wastewater treatment fibre membranes, as an absorbent, and as a gas remover.

1. Introduction

Polymer is a key substance in today's culture that has been widely utilized in a variety of industries, including as clothes, packaging materials, home appliances, and wastewater treatment. Furthermore, by merging modern technology with polymer, a wide range of polymeric substrates can be created, resulting in a new material for artificial skin. The qualities required for skin tissue engineering, including as non-immunogenicity, biocompatibility, biodegradability, and hydrophilicity, can be created by modifying the polymeric properties. [1]. Thus, by applying RIG co-polymerization, the polymer modification can be changed.

The use of advanced modification techniques to modify polymeric materials has increased dramatically in recent decades. Based on their design structure, the changed materials can be applied in a wide variety of applications. RIG polymerization is a method of designing and developing additional functionality for polymer materials while maintaining the shape and physical qualities of the trunk polymer [2][3]. Apart from that, while utilizing RIG polymerization to transfer functional groups to polymers, there is no unnecessary use of chemicals or catalysts. As a result, this process can be regarded both cost-effective and environmentally friendly, as it does not involve the use of a catalyst or a hazardous chemical that could harm human skin or the environment [4]. Gamma rays, electron beams, plasma treatment, and ultraviolet treatment are the four basic methods for initiating graft polymerization.

Grafting yield is used as a criterion to represent the degree of polymerization to determine the appropriateness of the radiation-induced graft polymerization reaction. The grafting yield determines the degree of graft polymerization response. Because it demonstrates the presence of a significant number of monomer functions in the trunk polymer, a higher grafting yield suggested the superiority of the functional materials. Apart from being associated with its application, the grafted polymer with a high grafting yield is the most desired while producing the polymer [2].

2. Surface modification of polymer using radiation-induced grafting polymerization method

According to the Science Direct database, there are four common ways to change the polymer using RIG techniques. There is gamma irradiation, electron beam irradiation, plasma irradiation, and UV irradiation (Figure 1).

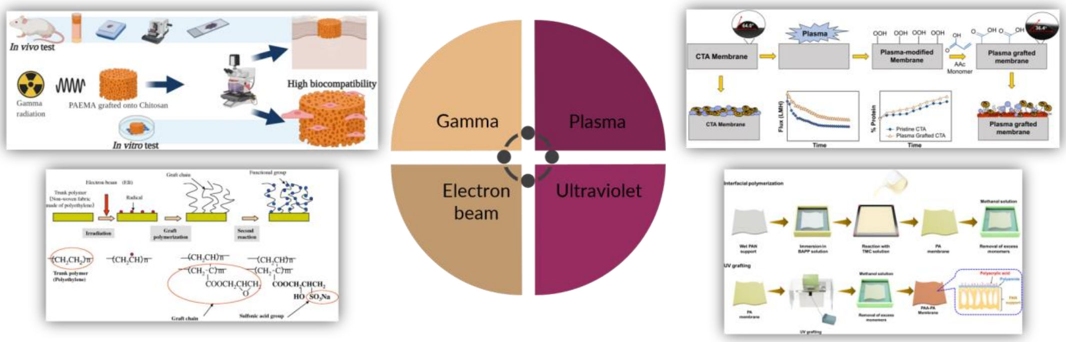

Figure 1. The types of commonly used radiation-induced grafting techniques.

First, utilizing cobalt-60 as a source of radiation, gamma radiation-induced grafting is more preferable and successful for altering and functionalizing polymers. As a result, gamma radiation-induced grafting can create instantaneous and uniform active radical sites, resulting in a high and clean grafting yield free of catalytic contamination and other harmful initiators. On a contrary, in the biomedical field, gamma is the preferred option [5]. However, because gamma radiation uses a radioactive isotope element, it is extremely damaging to one's health. Furthermore, waste management is challenging due to radioactive emissions, which may have a negative impact on human health and the environment [6].

The second most popular approach for polymer modification is electron beam radiation. On a polymeric surface, electron beam radiation can be utilized to create active sites or free radicals that can react with monomers to generate a graft copolymer, which is also known as a branched copolymer made of the main chain [7]. The gamma and electron beam methods have proven to generate a more stable product; nevertheless, gamma radiation is deemed notably superior to electron beam radiation since the approach achieved great grafting efficiency based on the copolymerization of natural rubber and polystyrene [8]. Then, another method to prepare the polymer is using plasma radiation techniques. Recently, Plasma irradiation has recently gained popularity as a surface modification technology that introduces desired functional groups of material structure that is ecologically friendly, does not use solvents, and takes less time [6]. According to Yang and colleagues, the plasma-induced grafting technique was used to modify carbon nanotubes (CNTs). The procedures were simple to use in revealing the active surface of CNTs and generating defects on the framework of prime CNTs without causing damage [9]. Furthermore, plasma irradiation can induce particular functional groups, such as carboxyl groups, to form on the membrane's surface, increasing the degree of hydrophilicity [10][11]. The membrane surface was coated with various energies and exposure times to activate gas plasma, and then the activated membrane was exposed to air to generate a peroxide group during plasma grafting polymerization. On the membrane surface, monomers such as hydroxyl, amine, or carboxyl groups were produced [11][10].

Finally, ultraviolet (UV) is one of the ways for polymer modification. UV radiation from sunlight was formerly thought to be a prevalent stressor on human skin. UVA (320-400 nm), UVB (290-320 nm), and UVC (320-400 nm) are the three forms of an ultraviolet radiation based on their spectrum [12]. The most damaging UV is UVC, which has the ability to harm human life due to its short wavelength and strong energy. UVB, on the other hand, is an excellent choice for breaking the polymer and causing photodegradation. UV irradiation grafting technologies have the advantages of milder reaction conditions, ease of operation, and the ability to be used on a larger industrial scale [13]. In addition, compared to alternative methods, UV treatment offers simplicity, speed, and inexpensive fabrication costs [14]. Table 1 summarises the research that was conducted based on the journal that was obtained.

3. Grafting yield

The fraction of monomer attached to the trunk polymer is referred to as grafting yield. The value of the percentage degree of grafting (DG) must be assessed in order to determine grafting yields. The types of solvents utilized, irradiation time or reaction time, temperature, absorbed dose, monomer concentration, and irradiation distance are all factors that influence the value of the DG during the polymerization process.

The different types of solvents have different effects on grafting yields, as shown in Table 1. Poly [(R)-3-hydroxybutyric acid] (PHB) was grafted with the n-hydroxyethyl acrylamide (HEAA) monomer produced using ^{60}Co gamma radiation at a dosage rate of 1.5 kGy/h at a dose of 10 kGy, according to Ochoa-Segundo and colleagues. Several solvents, including ethanol (EtOH), hexane (Hex), ethyl acetate (EtOAc), chloroform ($CHCl_3$), acetone (Ace), sodium acetate (SA), silica nanoparticles (SN), and acetic acid, were used in this experiment (Ac). Based on the type of solvent used, it was discovered that ethyl acetate produced the highest grafting yield of 83.06% when compared to other solvents. The propensity of ethyl acetate to create radicals during polymerization is the reason behind this. It is because ethyl acetate has the ability to produce radicals during the polymerization that affects the result of the gravimetric grafting degree [1].

The reaction time and irradiation temperature, on the other hand, also play a crucial effect in grafting yields. According to Aoki and others, electron beam radiation-induced grafting is used to create an antimicrobial fabric from NVP grafted onto polyolefin nonwoven fabric and iodine adsorption. First, the polyolefin nonwoven fabric is irradiated with a total dose of less than 160 kGy in a nitrogen atmosphere. It is then dipped in a 30wt % aqueous solution of (NVP) monomer and immediately transferred to a glass vessel for heating. The grafted fabric is then washed and dried, yielding PVP fabric as a result. The grafting process was seen in this publication to be continuous and did not manipulate the types of solvents utilised; instead, it measured the grafting degree based on reaction time and heating polymerization temperature. As a result of the findings, it can be concluded that a high grafting yield may be obtained when the reaction time is 30 minutes and the heating temperature is 337 ℃, which resulting in a grafting yield of 98 percent [15]. As a corollary, the grafting yield is considered to be influenced by the reaction temperature. Selambakkannu and his colleagues suggest that this is because high temperatures accelerate the kinetics response, resulting in a larger degree of crosslinking. Furthermore, the temperature condition has an impact on crosslinker diffusion inside the solution. As a result, the crosslinker will have easy access to the active sites produced by irradiation on the polymer surface [38].

Table 1. Previous research conducted for the modification of polymer by using radiation-induced grafting techniques.

Polymer	Methods of Preparation	Optimum grafting yield	Application	References
N-Vinyl pyrrolidone irradiated onto polyolefin nonwoven fabric and subsequent adsorption of iodine	Primary process -RIG using electron beam under nitrogen atmosphere Secondary process - Adsorption of Iodine	The degree of grafting (DG) is 98%, 90% and 70% depend on the temperature which is 317 K, 327 K, and 337 K respectively	antibacterial fabric	[15]
Styrene and chloromethyl styrene are irradiated onto poly(ethylene-co-tetrafluoroethylene)	Irradiated with gamma rays	50 % monomer produce 68% DG 25% monomer produce 31%DG	The production of polymer electrolyte fuel cells to be used as a power source for portable electronic devices and automobiles.	[16]
Bio-adsorbent modified natural rubber (NR) was prepared by RIG polymerization of glycidyl methacrylate (GMA) onto NR seed particle in a latex	gamma irradiation	Grafting percentage - above 70% at 90% glycidyl methacrylate emulsion/10% NR seed particles in latex form at 5 kGy absorbed dose	Remove Th^{4+} ion from the aqueous solution	[17]
Poly [(R)-3-hydroxybutyric acid] (PHB) grafted with the n-hydroxyethyl acrylamide (HEAA) monomer	gamma radiation	Poly [(R)-3-hydroxybutyric acid] (PHB)-g-n-hydroxyethyl acrylamide (HEAA) - 83.06%	Used for potential skin tissue engineering applications	[1]
Methyl methacrylate (MMA), acrylic acid (AA) and styrene (St) were used as the monomers.	electron beam	N/A	Used for textile dyeing	[18]
Grafting of N, N-dimethyl aminoethyl methacrylate from the surface of polyethylene/polypropylene nonwoven fabric	gamma irradiation	40% N, N-dimethyl aminoethyl methacrylate (DMAEMA) with DMF as a solvent - 12 % DG 50% of DMAEMA with water/acetone - 5% GD	To act as antibacterial properties	[19]
Grafting the cationic and anionic monomers onto radiation crosslinked chitosan through mutual radiation grafting technique.	Gamma irradiation	[2-(methacryloxy)ethyl] trimethylammonium chloride-g- CR-chitosan (Grafting extent~ 12.44%)	Used for the removal of cationic and anionic dyes from an aqueous medium with fast kinetics.	[20]

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Table 1. Continued

Polymer	Methods of Preparation	Optimum grafting yield	Application	References
Polyolefin-based sheath/core type composite fibers such as polyethylene/polypropylene, polyethylene/polyethylenerephtarate, PE.PE(HDPE) grafted with glycidyl methacrylate	Gamma radiation	N/A	Used as filters for the purification of contaminants in the environment Fe oxide - better performance for arsenic removal	[21]
Nylon-6 and vinyl benzyl chloride	Electron beam	Optimum DG - 130 wt.% at 20% vol% of (mixtureof3-and4-isomers,97%contains700–1100ppm Nitromethane as an inhibitor,50–100ppm tert-butylcatecholasan inhibitor)	Adsorbent applications	[22]
Alicyclic polyimides (APIs) derived from 1,2,4,5-pyromellitic dianhydride (PMDA) bis (4-aminocyclo-hexyl) methane (DCHM) onto styrene	Gamma irradiation	N/A	Used for the polymer electrolyte membrane for fuel cells	[23]
Grafting the of 2,2,2-trifluoroethyl methacrylate (TFEM) onto polypropylene (PP)	Gamma irradiation	based on types of solvent used: Tetrahydrofuran – 124% Dimethyl sulfoxide – 108%	It has a hydrophobic monomer thus it is a good water repellence that can be used for face masks, protective clothing for medical/outdoor application.	[4]
Glycidyl methacrylate grafted onto vinyl modified sepiolite	Gamma radiation	Effect on the absorbed dose at 1% vinyl modified sepiolite and 5% glycidyl methacrylate – 350%	Used for metal ion adsorbent for the recovery of metals from wastewater	[24]
Glycidyl methacrylate (GMA) onto ultra-high molecular weight polyethylene (UHMWPE) fiber and further modifying with triethylenetetramine (TETA) and glycidyl trimethyl-ammonium chloride (GTA)	Gamma-ray pre-irradiation	447.5% DG at absorbed dose 10kGy, glycidyl methacrylate at 20% and temperature at 50 °C	Has the potential application for Cr (VI) removal from wastewater	[25]

Table 1. Continued

Polymer	Methods of preparation	Optimum grafting yield (%)	Application	References
Polysaccharides grafted onto poly(3-hydroxybutyrate)	Gamma-ray radiation	Ethyl acetate solvent – 32.11%	Application as biomaterials	[26]
N-vinylpyrrolidone onto segmented polyurethane samples based on isophorone diisocyanate	Gamma-ray radiation	N-vinylpyrrolidone – 53% DG at 1.2kGy	Modification of the polyurethane surface is used for biomedical application	[27]
N-vinyl-2-pyrrolidone is grafted onto polyethylene-coated polypropylene fiber	Electron beam	149% DG at 200kGy dose –	Adsorbent for volatile iodine adsorption from fission products in nuclear power plant reactors	[28]
PVDF (polyvinylidene fluoride) membrane modified with glycidyl methacrylate (GMA) and sodium sulfite	Gamma radiation	16% DG at concentration of glycidyl methacrylate 0.5wt%	Ammonia removal for membrane bioreactor	[29]
Acrylamide (AM) onto carboxymethyl cellulose (CMC)in the presence of a cross-linking agent, N, N-methylene bisacrylamide (MBA)	Gamma radiation	N/A	To control the release of agrochemicals by reducing irrigation water consumption and improve the physical properties of soil	[30]
Polystyrene (PS) grafted sepiolite nanohybrid (MS-g-PS) in the presence of dichloromethane (DCM) as a solvent	Gamma radiation	170% DG at 58% mmol/g of sepiolite nanohybrid with adsorbed dose at 70kGy	Used as a low cost and efficient ion exchange resins	[31]
Poly(N-iso-polyacrylamide) (PNIPAAm) onto an ethylene-tetrafluoroethylene copolymer (ETFE)	Electron beam	16% DG at 30wt% of NIPAAm solution -	Used to generate tissue and organ through lamination and transplantation	[32]
Grafting of zwitterionic 2-methacryloyloxyethyl phosphorylcholine (MPC) polymer brushes (5–8 nm) on the silicone (polydimethylsiloxane, PDMS)	Initiator free atmospheric plasma-induced polymerization	N/A	Used as silicone-based contact lenses, wound dressing, and artificial organs	[33]

Table 1. Continued

Polymer	Methods of preparation	Optimum grafting yield (%)	Application	References
Iodine grafted into poly (*N*-vinyl pyrrolidone) coatings (pPVP) with controllable thicknesses	Plasma induced grafting polymerization	N/A	Used as coating for the medical implants' devices and wound dressing	[34]
Glycidyl methacrylate (GMA) was grafted onto polypropylene (PP) fibers	Plasma induced grafting polymerization	N/A	Used as the selective adsorption of Cr (VI)	[35]
Acrylic acid (AA) monomer was grafted on the Low-Density Polyethylene (LDPE) film	UV radiation	AA onto LDPE films in different solvent solutions (water) – 550% under 4 min treatment AA onto LDPE films in various concentrations of water/acetone solutions – approximately 520%	Application in food packaging and food cling wrap by maintaining food quality with reduced additive	[36]
Polyethylene glycols (PEG) grafted onto the surface of Poly (ether ether ketone) (PEEK) membranes	UV radiation	Optimum grafting degree 12% DG at Irradiation time at 30 min and 60 min	Used as the antifouling for the hollow fiber membrane	[13]
Cellulose triacetate (CTA) membrane grafted with acrylic acid	Plasma grafting polymerization using Argon and carbon dioxide gases	13% DG at time 5s (Acrylic acid + CO_2) and 12% DG at15s (Acrylic acid + Ar)	Used as an anti-protein fouling properties in forward osmosis for protein recovery application.	[10]
(Poly (ethylene glycol) methyl ether methacrylate) (poly (PEGMA)) grafted onto polysulfone (PSf) substrates	UV/ozone induced graft polymerization	N/A	For CO_2 separation and remarkable for other membrane application.	[14]
Hydroxyethyl methacrylate (HEMA) was grafted onto the surface of PAN fiber fabric (PAN-g-HEMA)	UV radiation	80% DG at monomer concentration 40wt%. 75% DG at concentration of Benzophenone at 2wt%	Used as fire retardant	[37]

Other criteria that affect grafting yield are depend on the irradiation dose and monomer concentrations. N-vinyl-2-pyrrolidone (NVP) was grafted into polyethylene-coated polypropylene skin-core (PE/PP) by Ye and his team. NVP and ethanol were introduced to PE/PP in a PE bag before the irradiation procedure, with the mass ratio of the NVP/ethanol solution ranging from 50% to 100%. After the leftover monomer was removed, the PE bag was heat-sealed. The NVP-containing PE/PP was then grafted using an electron beam of 1.5 MeV energy with varied absorbed doses ranging from 25 to 200 kGy at ambient temperature and an irradiation time of 5 path – 40 path with an absorbed time of roughly 40s to 320s. It is determined that the highest grafting yield was obtained when the absorbed doses were at 100 kGy with 100% monomer concentrations. However, the grafting yield increased sharply with 100% NVP and reached 146% at the absorbed dose of 200 kGy. However, high energy consumption seems to make it an unsuitable condition for polymerization. Thus, it is determined that the materials with grafting yield of 100 to 200 % were considered as the best adsorbents and the adsorbents prepared from 70% NVP at the adsorbed doses of 100 kGy are appraise as the finest condition [28].

Other than that, according to Taimur and teams, polystyrene (PS) was grafted with sepiolite nanohybrid (MS) using gamma rays' radiation grafting from the ^{60}Co irradiator in the presence of dichloromethane (DCM) as the solvent [31]. To get the highest grafting yield and kinetics, researchers are investigating the effects of varied irradiation dosages and monomer concentrations [39] [40]. Increases in grafting yield were observed as the absorbed dosages were increased. The increase in grafting yield is attributed to a rise in the number of radicals generated by radiation on monomers [41] [42]. Nonetheless, when grafting yield reaches particular doses, it can stop increasing because it has attained homo-polymerization, which causes the system's viscosity to increase, making it difficult to approach the monomer radicals to the grafting chain on MS [31] [43]. In terms of monomer concentration, the higher the monomer concentration, the higher the grafting yield. This is because the monomers are capable of reacting with the active sites of the irradiation materials, resulting in a corresponding increase in grafting yield [44]. In addition, according to Madrid and co-workers, the increasing of grafting yield along with the increasing of the monomer concentration is due to the higher diffusivity of monomers into the polymer matrix which amplifies the grafting yield [42].The irradiation distance is the last parameter that affects the value of the grafting yield. According to Ren and colleagues' research, UV induced photo graft polymerization was used to graft hydroxyethyl methacrylate (HEMA) onto the surface of a flame retardant polyacrylonitrile (PAN) fibre fabric. The grafting yield was affected by the irradiation distance. As the radiation distance reduces, the intensity of the radiation light on an object increases. Distance plays such an important role in establishing radical spots on the fabric. However, if the radiation is too close, the energy emitted by the radiation becomes too strong, causing the fabric to yellow and become brittle. [37].

4. Application of polymer prepared from RIG method.

According to the data in Table 1, biomedical and pharmaceutical applications are some of the applications. In this case, the polymer is used to make an antibacterial fabric that can be used as a wound dressing. It's also utilized for new skin tissue engineering, which involves laminating and transplanting new tissue and artificial organs. Aside from that, polymer modification to increase hydrophobicity and waterproof characteristics can be used to make face masks and contact lenses. Then, the other application that can be produced by using radiation-induced grafting technique is for electrical devices and automobiles where the trunk polymer was grafted to form polymer electrolyte fuel cells that act as power sources for the electrical devices and automobiles. An adsorbent and gas remover is another application. Hazardous gases such as TH^{4+} and ammonia may be easily removed, and it has the potential to be used as an adsorbent in water treatment for the removal of Cr (VI) and dyes from the water body. Apart from that, radiation-induced grafting polymer has commercial potential, as it may be utilized for textile dyeing. In food packaging application, it allows the reduction of food additives while retaining food quality. The modified polymer can also be used as an antifouling agent for hollow fiber membranes and as a fire retardant to inhibit fire spread.

5. Conclusion

Finally, the gamma, electron beam, plasma, and ultraviolet methods for preparing polymer via radiation-induced grafting are identified. For the synthesis of polymer utilising radiation induced grafting, most researchers prefer gamma. Furthermore, electron beam radiation is gaining popularity because it produces and contains no radioactive elements such as gamma. Plasma and UV are also used by some researchers. Furthermore, the most significant aspect in determining the optimum degree of grafting during polymerization is grafting yield. The types of solvent, irradiation time, irradiation temperature, irradiation dosages, monomer concentration, and irradiation distance are all parameters that affect grafting yield. These are the most critical elements to consider in order to maximize the grafting yield. Furthermore, it has been discovered that the polymer created by radiation-induced grafting can be used in a variety of applications, including biomedical, electrical applications, adsorbent for gas removal, commercialization as a waterproof face mask, antifouling for hollow fibre membranes, and fire retardant.

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