

IMPROVE THE NATIVE CHARACTERISTICS OF POLYSACCHARIDES BY GRAFTING THROUGH THE GAMMA RADIATION: A REVIEW

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Abstract

Polysaccharides are bio-degradable, inexpensive and easily available from consistent agricultural resources. Polysaccharides and their derivatives represent a group of polymer widely used in pharmaceutical and biomedical fields. The biodegradability of natural polymers reduces their shelf life. Grafting copolymerization technique is a most effective fascinating way for chemical modification of native characteristics of polysaccharides with maximum possibilities for improving the properties of polysaccharides and enhanced the range of exploitation. While the major difficulty facing us during synthesizing a graft copolymers reaction, is the lack of commercial methods of synthesis and lower percent graft yield. It is well known that, the most important methods of synthesis engage the employ of chemical free radical initiator i.e. conventional based methods. Graft copolymerization through -radiation method is a better method of grafting in comparison to chemical method and exhibits a great potential to synthesize the graft copolymers by virtue of its higher efficiency, low cost, higher thermal stability as well as enhanced the yield of graft copolymer. Future prospective of irradiation technique would be significant impacts to develop of polymerization.

Key words - Grafting, Polysaccharides, Gamma Radiation, Copolymer, Polymerization.

INTRODUCTION

Natural polymer mainly Polysaccharides are abundant in nature, universally found in almost all living organisms. They are present in numerous tissues of seeds, stems and leaves of plants, the body fluids of animals, shells of crustaceans and insects. They are also found in the cell walls and extra cellular fluids of bacteria, yeast, and fungi and are thus renewable reservoirs for synthesizing high performance materials [1,2]. Polysaccharides are polymers which are made by mono saccharides. Polysaccharide are cheap, have wide availability and available in a variety of structures with a variety of properties [3] and can be easily modified and are highly stable, safe, nontoxic, and hydrophilic and gel forming and in addition biodegradable, and widely used as targeted drug delivery systems. A problem encountered with the use of polysaccharides is their high water solubility. An ideal method is to adapt the solubility while still retaining their biodegradability. Many of the polysaccharides have already been used as colon specific drug carrier systems, such as Chitosan, chondroitin sulfate, pectin, Cyclodextrins, dextrin's, guar gum, X pectin, pectin, locust bean gum and amylose [4]. Most polysaccharides have been used as food and pharmaceutical and biomedical accidents because of their biodegradability, biocompatibility, easy availability and low cost. Seed polysaccharides, in native or derived forms from quince, flax, psyllium and guar [5] have been explored for commercial used in paper, cosmetics, textiles, and food and pharmaceutical industries. Adsorption, flame retardants, and corrosion inhibition are amongst the anticipated characteristic sought in a polysaccharide derivative for its actual commercial exploitation. Polysaccharides in their natural form are used as flocculants, and coagulants e.g., starch, sodium alginate, guar gum, amylopectin, xanthan gum, Chitosan, psyllium, and okra mucilage [6], while, in an adapted form they are used as water super Sorbent, e.g., guar-graft poly (sodium acrylate) [7]. Improvement of polysaccharide materials is often carried out through derivative of functional groups, grafting of polymeric chains, and by oxidation or hydrolytic degradation [8-12].

Grafting method is one of the greatest effective fascinating fields for chemical modification of polymers with unlimited possibilities for improving the characteristic of polymers and enhanced the range of exploitation. While the chief difficulty opposite us during synthesizing a graft copolymers reaction, is the lack of commercial methods of synthesis and lower present graft yield. It is well known that, the most significant methods of synthesis involve the employ of chemical free radical initiator i.e. conventional method [13-16] high energy radiations i.e. gamma and X-ray, UV- radiation based method, and recently the microwave based methods [17-20]. The conventional (physical method) process of grafting uses a chemical free radical initiator to generate a steady distribution of the free radical sites on the polymer backbone, where the monomer of the graft gets added up to form the graft chain. This process of synthesis has low reproducibility and is not very suitable for commercial scale synthesis.

A superior method of graft copolymerization synthesis is high energy radiation (gamma rays or electron beam) as the free radical generator [21-22]. But this method is not appropriate for grafting onto carbohydrate polymers due to it can reason damage to the polysaccharide backbone (i.e. Radiolysis) at the high energy radiation. While, the UV rays in the presence of proper photo sensitizer can also be used, but low penetration of UV- rays into the bulk of polymer makes it proper for surface grafting only. The significant



method of graft copolymer synthesis is through microwave radiation to generate the free radical sites on the natural polymer backbone. [23].

Researchers are more specifically interested in grafting polymers to surfaces to modify the chemical, physical and bio-logical characteristic of biometrics using irradiation methods. The grafting of a monomer onto a natural polymer backbone by high-energy irradiation is an important technique for adapting the natural polymer. The radiation graft copolymerization can be performed by the direct / mutual method in which the monomer and the polymer (graft substrate) is irradiated together concurrently to create the radicals, which are initially the polymerization or by pre irradiation /indirect grafting in which the monomer is contacted with the polymer backbone irradiated alone before [24]. The radiation method seems an excellent method for the synthesis of hydrogen, because a polymer in an aqueous solution, with a monomer dissolved in it, undergoes crosslinking and graft copolymerization on irradiation to yield a hydrogel. Simple process control, no initiators, cross linkers, no waste, and relatively low operating costs make an irradiation method a proper choice for the synthesis of hydrogen [25]. Radiation induced grafting proposals some unique advantages over the conventional chemical grafting technique since it results in uniform grafting and it is room a temperature process [26]. Radiation grafted co-polymers have been wide applications in many fields like surface modification [27], separation purposes [28], biotechnology [29], electrochemical applications such as battery separator, electro-dialysis and solid polymer electrolyte in fuel cells [10]. Ion exchange membranes have been explored for their appropriateness as battery separators because of their durability, long life, high charge density and appreciable ion exchange capacity at optimum water content, which is the greatest desirable characteristic of the separator membranes. Radiation induced grafting of different monomers onto poly (propylene) (PP) by pre irradiation as well as post irradiation process has been investigated to improve/introduce certain desired characteristic [30-31].

In the past several techniques had been developed for radiation-induced grafting. In the pre-irradiation technique polymeric matrix is first irradiated in order to generate reactive sites either in an inert atmosphere or in vacuum. The radicals have to be stable at ambient temperature in order to initiate covalent bonds with monomers after irradiation [32-33]. The procedure results in removal of homo polymerization, but the yield of grafting is in this case relatively low. Another difference of such a way is an application of air atmosphere during exposure to ionizing radiation. Both methods are proper if only formed radicals are stable, sufficient to react with monomers in post-irradiation effect. In such a case, grafting, contingent on the system, might be performed after a long storage time. The most common technique used for grafting consists in irradiation of monomer solution in contact with the surface of natural polymers. Hypothetically, radicals produced both in natural polymer and in monomer might initiate covalent bonds between the reagents. However, the contribution of reactive sites situated in polymeric matrices in grafting procedures has not been determined yet and made an effort to determine the role of radicals produced in selected polymers on radiation-induced grafting [34].

Ionizing high energy radiation, like UV radiation, gamma rays, X ray, and electron beam, and, microwave radiation had been used as an initiator for the modification natural polysaccharides.

S.N.	Radiation	Energy	Energy Sources
1	Gamma Radiation	1 MV	Radioactive Material (60Co, 137Cs)
2	X – Rays	50 – 200 Kv	Generator Synchrotron
3	UV Radiation	4.13–6.20,	UV lamp
4	Microwave Radiation	300mhz-300ghz	Microwave Domestic Oven
5	Electron Beam	20 Kv – 10 MV	E-Beam Accelerators

The irradiation of aqueous natural polymer solution results in the formation of radicals on the polymer chains and irradiated of water molecules results in the generate the hydroxyl radicals, which also attack the polymer chains, resulting in the formation of macro radicals. Recombination of the macro radicals with various natural polymer chains by the covalent bonds, then cross-linked structure is made. Examples of polymers cross-linked through the radiation process are poly (vinyl alcohol), poly (ethylene glycol) and poly (acrylic acid). The chief benefit of the radiation initiation over the chemical initiation is the production of relatively pure, maximum yield and initiator-free SAPs [35].

The high energy irradiation category includes ion radiation, -irradiation (electron beam), and -radiation and widely used to achieve either ion establishment at the top surface layer or to deposit coatings. For this purpose, numerous ions like hydrogen, noble gases, gold, *etc.* are employed. High energy photons can bring surface radicals, which work as initiating sites for subsequent functionalization. For the action of polymers, additional immediate chemical effects are also likely, free radical recombination and crosslinking, and chain scission [36].

Ionizing radiation used for understanding mechanism of graft copolymerization reaction and for initiation of the polymerization procedure. Some of the benefits of the radiation initiated graft copolymerization over conventional methods are: (i) nonappearance of



external matter, like initiator, catalyst, etc., (ii) polymerization at minimum temperature or in solid state, (iii) rate of the initiation step can simply be controlled by varying dose rate and (iv) the initiating radicals can be created regularly by -irradiation. The gamma ray induced radiation polymerization, approximately speaking, in addition to the narrow sense "radiation polymerization (also known as radiation initiated polymerization i.e. Using high energy ionizing radiation to irradiate monomers, and produces ions or radicals, so that form the active site and the graft copolymerization reaction and also includes the and radiation crosslinking polymerization, etc. [37-40].

RADIATION TECHNIQUES

Radiation techniques have many benefits over conventional methods. When using radiation for materials processing, neither use the catalysts nor are additives vital in order to initiate the reaction, considering the energy source being UV radiation, gamma-ray, or electrons [41]. The chief advantage of these radiation-assisted techniques is that they are relatively simple. Surface graft copolymerization of polymers has attracted great interest in the past few years as it allows the tailoring of surface characteristics of several polymer materials and thus, specific functionalities useful for wide applications may be successfully produced [42]. When polymers are exposed to ionizing irradiation, surrounded radicals and peroxides or hydro peroxides are formed as a result. These radicals are talented of initiating polymerization reactions. Several polymerization methods have been advanced, including direct graft polymerization of vinyl monomers that already comprise necessary functional groups [43].

High-energy radiation, such as UV radiation, gamma-irradiation and electron beam lead to initiation of polymers in a technique that proper monomers may be grafted. For the amount of modification, *i.e.*, the grafting grade may be easily measured by careful difference of the radiation exposure and reaction conditions. The grafting method by gamma- irradiation is tremendously attractive considering that the improved biomaterial is obtained in the purest form possible, no chemical initiators are essential and it may be appropriate for almost all polymer-monomer mixtures and thus allowing vast potentials to generate materials with modified physical-chemical and biological properties [44]. Another benefit of gamma-irradiation is that it bids much higher saturation into the polymer if compared to the electron beam [45].

RADIATION INDUCED GRAFT COPOLYMERIZATION

Radiation-induced grafting is a well-established technology dating back almost 60 years. If the polymer substrate is bare to ionizing radiation (for instance, electron beams, gamma-rays, X-rays) the reactive sites arbitrarily produced along a natural polymer chain may work as a macro-initiators and initiate free radical polymerization of the monomer as a significant. This technique is appropriate for a lot of polymer/monomer combinations and unlike chemically initiated grafting; there is no adulteration from initiators. The extent of polymerization of monomers on substrates is articulated in terms of grafting yield, also known as graft copolymerization degree [1].

GAMMA () RAY-ASSISTED GRAFTING COPOLYMERIZATION

Radiation is a term which disguises the electromagnetic and particulate radiation from gamma radiation with a wavelength of Pico meters to radio waves with wavelength of hundreds of meters. High energy ray can be understood to mean the ray which can cause ionization in chemical compounds, which requires 10eV corresponding to 1000 kJ per mile and a wavelength of 100 nm. Gamma radiation is electromagnetic radiation of high occurrence and therefore energy. Gamma rays are ionizing radiation and are classically produced by the decay from high-energy states of atomic nuclei (gamma radiation decay) but also in many other methods. Currently, gamma irradiation-initiated graft polymerization was advanced as a preferred method for commercial synthesis and shows a great potential to synthesize the graft copolymers by the advantage of its higher efficiency, minimum cost, and convenient to improve the polymer [46, 3]. The graft copolymerization through -radiation method is a better method of grafting in comparison to chemical method in leading to higher thermal stability. Kinetic study of graft copolymerization of AAc and methacrylate onto starch using a ceric ion initiator has been studied by Taghizadeh et al. [4]. The utilization of gamma irradiation for the synthesis of gum-gcopolymers has been intensively concerned. For example, Lokhande et al. [5] And Biswal et al. [57] prepared for gum-g-copolymer by using -radiation as the initiation approach. It was found that ideal graft efficiency was obtained, and the graft efficiency is dependent on the dose of radiation. The increase of radiation dose can enhance the graft ratio and decrease the content of homo polymers, but the viscosity or molecular weight was decreased due to the depolymerisation effect of radiation to gum. Besides, the graft copolymer of other gums such as XG [48] and KG [8] was reported. However, the problems associated with radiation initiation involve lack of distinction between the different bonds of the backbone polymer because there is always a strong probability of radiation damage (radiolysis) to the gum backbone, and undesirable breakage of bonds may occur under the action of strong radiation [3]. In addition, this method has required a complex instrument and equipment and exists with safety problem resulting from the radiation. This limited the extensive application of radiation technology in the synthesis of gum-g-copolymer.





The mechanism of gamma-radiation-initiated graft copolymerization [104]

Kaith et al prepared psyllium and acrylic acid based hydrogels by using -radiations and characterized using SEMs, FTIR. The swelling behavior of the final product (hydrogel) was investigated in distilled water as functions of time, temperature, and pH and hydrogel showed maximum swelling at 25 °C for 24 h. Hydrogen has been found to follow the non-Fickian type mechanism for the fungicide release [10]. Singh and Kumar, synthesized psyllium and N vinylpyrrolidone (NVP) based hydrogels by radiation induced crosslinking and hydogel were characterized with SEMs, FTIR and swelling studies. Swelling of the hydrogels was studied as a function of monomer concentration, total radiation dose, temperature, pH and [NaCl] of the swelling medium. The swelling kinetics of the hydrogels and release dynamics of anticancer model drug (5-fluorouracil) from the hydrogels has been carried out for the evaluation of swelling and drug release mechanism in the following steps [11].

Step I – Initiation



Step II - Propagation

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PSYLLIUM CROSSLINKED POLY (N-VINYL PYRROLIDONE) NETWORK [PSY- CL- POLY(NVP)]

Naggar et.al has developed the Tara gum and acrylic acid, hydrogen by using gamma irradiation, in the presence of N, N_methylene bis acrylamide (MBAAm) as a crosslinking agent. The synthesized hydrogels were characterized by FT-IR and evaluated by swelling studied as a function of MBAAm concentration, temperature and the nature of the swelling medium. The swelling kinetics of the hydrogen were studied in terms of the diffusion exponent "n". Hydrogels have non-Fickian type. Mechanical measurements (stress-strain) curves of hydrogels were evaluated to calculate the shear modulus values and the average molecular weight between crosslinks (Mc). Moreover, the absorption under load at 37 ^oC of water and urea aqueous solutions (as a major component of urine) by TG/AAc hydrogels was determined. [49]. Khalek and mahmaud have developed a novel polymeric flocculent by graft copolymerization of acrylamide (AAm) with acrylic acid (AAc) using gamma irradiation technique. The synthesized product ware characterized using Fourier-transform infrared spectroscopy (FTIR), and thermo-gravimetric analysis (TGA). They studied the effects of reaction parameters, such as total absorbed dose, and monomer concentration on grafting yield and the flocculation performance of the graft copolymer P (AAm/AAc) in coal suspension. They were observed that the grafting ratio was one of the key factors for the flocculating effects. The copolymers with various grafting ratios showed different flocculating properties and grafting ratio increased, the flocculating effect also increased [50].

Iskandar S. Studied the Graft Copolymerization of Methyl Methacrylate Monomer onto Starch and Natural Rubber Latex Initiated by Gamma Irradiation. He synthesized the biodegradable plastic. He was observed that the grafted copolymer of starch-poly methyl methacrylate and the grafted copolymer of natural rubber-poly methyl methacrylate were mixed in the blender, and dried it in the oven. The dried grafted copolymer mixture was then molded using a hydraulic press machine. He was found that the tensile strength of grafted copolymer mixture increased by -ray irradiation. Increasing of the grafted copolymer of natural rubber-poly methyl methacrylate content, the gel fraction and tensile strength of the grafted copolymer mixture was increased from 18 MPa to 23 MPa after recycled (film forming reprocessed) 3 times. The grafted copolymer mixture was degraded completely after soil buried for 6 months [51].

Lokhande et al. Prepared water super Sorbent guar-modified polymers by grafting of acrylonitrile onto GG through radiation [5]. The change in rayon fibers properties of their surface grafting with us by chemical reactions and -radiation method has been reported by Kaur et al. [52]. Pande et al. 15 studied the functionalization of poly (vinyl chloride) through irradiation in air with -rays [53]. Fanta G. Et al have studied the use of a mixture of acrylamide and 2-acrylamido – 2- methyl propane sulfonic acid for grafting on starch by gamma irradiation. It was found that the conversion of monomers to graft copolymer was nearly quantitative when pregelatinized wheat starch was irradiated along with the monomer [54]. Bhuiyan, et al., studied the Hydrogels based on Poly (vinyl pyrrolidone) (PVP) networks grafted with acrylic acid (AAc) was synthesized by the application of gamma radiation of 25 kGy dose from the Co-60 gamma source at room temperature (27°C). PVP concentration was kept constant (10%) and acrylic acid concentrations were varied from 0.0 to 2.0% wt. The hydrofoil was characterized using FTIR, XRD, DSC and TGA. The FTIR spectra



and XRD data indicate, the formation of copolymer networks, whereas the DSC and TGA study showed that the PVP/each blend hydrogels possess higher thermal stability than PVP hydrogels [55].



Casimiro et. al studied, 2-hydroxyethyl methacrylate has been grafted onto Chitosan by using either chemical initiation, or photoinduction or gamma radiation-induced polymerization, all under heterogeneous conditions. The evidence of grafting was provided by Fourier transform infrared spectroscopy and thermal analysis [56]. Pourjavadi et.al synthesized of an environmental-sensitive, super absorbent hydrogel using rays are described. Grafted poly acrylonitrile onto a homogeneous solution of starch and kappa -Carrageenan hybrid backbones created a smart material with reversible behavior. The reaction parameters (i.e. Acrylonitrile, starch and kappa-Carrageenan concentration, as well as -irradiation time) affecting the water absorbency of the hydrogen were optimized using the Taguchi method, in order to achieve a hydrogen with high swelling capacity. FTIR spectroscopy was used for confirming the structure of the product and the morphology of the synthesized hydrogel was examined by a scanning electron microscope [25]

Wang et.al studied the Surface treatment of poly (ethylene terephthalate) by gamma-ray induced graft copolymerization of methyl acrylate and its toughening effect of poly (ethylene terephthalate) /elastomer blend [57]. Madrid et.al, prepared the Modified microcrystalline cellulose (MCC) through gamma radiation-induced graft polymerization of glycidylmethacrylate(GMA). Simultaneous grafting was employed where in MCC with GMA in methanol was irradiated with gamma radiation in nitrogen atmosphere. They were studying the effect, so different experimental factors such as monomer concentration, type of solvent and absorbed dose for the degree of grafting, Dg, The amount of grafted GMA, expressed as Dg, was determined gravimetrically. Information from grafted samples subjected to Fourier transformed infrared spectroscopy (FTIR) in attenuated total reflectance (ATR) mode showed peaks corresponding to GMA which indicates successful grafting. The X-ray diffraction (XRD) analysis revealed that the crystalline region of MCC was not adversely affected after grafting with GMA. The thermo gravimetric analysis (TGA) data showed that the decomposition of grafted MCC occurred at higher temperature compared to the base MCC polymer [58]. Burillo et.al, studied the Synthesis and characterization of N-vinylcaprolactam/N, N-dimethylacrylamide grafted onto chitosan networks by gamma radiation and they were investigated that the binary graft systems were synthesized in one and two steps to evaluate the influences of architecture on the properties of the polymeric material. Maximum grafting percentages were obtained by the direct method. The different system so obtained were characterized by FTIR and TGA [59].

CONCLUSION

Polysaccharides, are biodegradable, fairly, shear stable cheap and easily available from reproducible natural resources. The biodegradability of polysaccharides reduces their shelf life. Modified polysaccharides have been the wide area of scientific research. These polysaccharides are cost effective, biodegradable and quite efficient towards various technological processes, provides a better option for the artificial synthetic materials. Grafting is a unique method among the techniques to add advanced properties to the natural polymer with a small loss of natural properties. One of the most successful paths of modification of polysaccharides is by graft copolymerization with proper monomers. The properties of the final product (grafted polysaccharide) can be properly modulated in



terms of percentage (%) grafting. The final product copolymer is thus planned at the molecular level for desired applications. The most current technique in graft copolymerization involves the use of radiations to initiate the graft copolymerization reactions

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