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Advancement of Indian herbs knowledge in drug development

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Abstract
India has a rich heritage of traditional medicine. The knowledge of medicinal herbs has been passed down through generations, offering natural remedies for various ailments. With the progress of modern science, the integration of traditional wisdom with contemporary drug development approaches has gained increasing attention. Natural herbs are widely available in huge quantities. Natural herbs alternatively, are more appealing for pharmaceutical applications owing to their biocompatibility, low cost, biodegradability, chemical modification and necessary surface qualities but there are some drawbacks like microbial degradation, low solubility, and stability issues etc... Grafting of synthetic polymer onto natural polymers can introduce desired properties by the use of different techniques of grafting method. The yield of grafting increases in microwave assisted grafting method as compared to conventional grafting. The objective of this abstract is to spotlight the recent advances and modifications of natural herb polymer modification and its applications in pharmaceutical industries.

Keywords: Indian herbs, grafting, graft copolymer, herbs modification

1. Introduction

Making an environment free from chemical pollution the use of most toxic chemicals has been eliminated by the use of natural polymer. Polysaccharides are abundant natural polymers found in plants, animals and microorganisms. Their unique properties like biocompatibility, low-toxicity, and biodegradability make them attractive candidates for various applications across industries such as pharmaceuticals, foods, cosmetics and material science. The application of native natural polymer is however limited in drug delivery applications due to their inherent drawbacks like uncontrolled hydration, microbial contamination and drop in viscosity during storage· [1]. To overcome these challenges, researchers have turned to graft polymerization as a versatile technique for modifying polysaccharide, enabling the enhancement of their properties and expanding their application. Hybridization with synthetic polymer could be a potential alternative to resolve these issues without affecting its physical and chemical properties and they have varied application in drug delivery, flocculation, stabilizing agents etc. The graft copolymer can be assumed as a trunk polymer from which the branches of other polymer are attached and are radiating from different sites along the full length of polymeric backbone [2].

2. Natural polymer hybridization

This term focuses on the hybrid nature of combining natural (herbal) and synthetic (polymeric) components through graft copolymerization, emphasizing the fusion of both sources of materials

Indian herbal medicine, deeply rooted in centuries of tradition, has always been a source of significant pharmacological and therapeutic insights. The holistic approach of Ayurveda, India's ancient system of medicine has long relied on the use of plant-based remedies to treat a wide array of ailments. This treasure trove of knowledge has led to the identification of numerous bioactive compounds, which have inspired modern drug discovery and development.

In recent years, the growing interest in natural products has encouraged scientists to explore how traditional remedies can be harnessed for modern therapeutic applications. The integration of Indian herbal medicine into drug discovery is not limited to the direct isolation of active compounds but extends to the development of novel drug delivery systems and materials [3]. One such innovative approach is graft copolymerization, a method where natural polymers or herbal extracts are chemically bonded to synthetic polymer backbones to create hybrid materials with enhanced properties [4].

Graft copolymerization is particularly relevant when developing drug delivery systems, as it allows for the modification of polymers to improve drug solubility, stability, and controlled release. Indian herbs, with their diverse chemical constituents, offer a unique opportunity to synthesize graft copolymers with properties suited for specific therapeutic needs. For example, natural antioxidants, alkaloids, flavonoids, and terpenoids derived from Indian herbs could be grafted onto synthetic polymers, resulting in materials that enhance bioavailability and therapeutic efficacy.

This fusion of traditional herbal knowledge with modern scientific techniques like graft copolymerization has the potential to revolutionize drug discovery. By improving the effectiveness, safety, and controlled delivery of bioactive compounds, researchers can develop more targeted, efficient, and sustainable drug therapies, addressing unmet medical needs in a way that combines the best of both worlds: ancient wisdom and modern innovation [5].

3. Graft copolymerization

Graft copolymer is made up from the long sequence of one polymer with one or more branches of another polymer [6]. The synthesis of graft copolymer procedure begins when free radical sites will produce on preformed polymer with the help of redox initiator such as Ammonium persulphate, Cerric ammonium nitrate etc. [7]. On the polymer backbone, once the free radical sites are formed then the monomer is able to attach through the chain propogation step, leading to the formation of grafted chains. Graft copolymerization is an effective and cost efficient method for altering the properties of natural polysaccharides, making them suitable for use in drug delivery applications [8].

3.1Different techniques of grafting [9]

3.1.1 Chemical Grafting [10]

In this technique, initiator plays a vital role in grafting as it decides the rate of grafting process. Grafting can be initiated by chemical means through following two main paths such as free radical grafting and ionic grafting.

3.1.2 Free Radical grafting

Graft copolymers are formed when initiators produce free radicals during the chemical reaction which are transported to the substrate and mixed with the monomer. Potassium per sulphate (PPS), Ammonium per sulphate (APS), Ferrous Ammonium Persulphate (FAS), Cerric Ammonium Nitrate (CAN) etc. are the following initiators sustem utilized in the free radical grafting. Natural polysaccharides like Chitosan, starch, inulin and pectin are grafted with polyphenols through the action of free radicals. Different monomers such as acrylamide methacrylate, n-acrylonitrile have been grafted on polysaccharide through free radical method. Sonia Dhiman *et al*. reported that Moringa gum was modified using graft copolymerization based on free radical mechanism.

3.1.3 Ionic Grafting

Ionic grafting is of two types anionic or cationic with the help of using ions produced by high energy irradiation. Graft copolymer is formed when polymer is irradiated to produce the polymeric ionsand then reacts with the monomer.

- Anionic grafting
- Cationic Grafting

3.1.4 Grafting via living polymerization [11]

A "living" polymer refers to a type of polymer that maintains the ability to grow for an extended duration while chain transfer remains negligible. When light absorption is insufficient, the process can be enhanced by adding photosensitizers like benzoin ethyl ether [12].

3.1.5 Grafting through enzymes

In this method, enzymes facilitate the grafting process, which can be chemical or electrochemical. For instance, tyrosine can transform phenol into a reactive o-quinone, which then undergoes further non-enzymatic reactions with chitosan. Utilizing enzymes instead of reactive agents during grafting processes can be more environmentally friendly, cost-effective, safe, and successful. Chen *et al*. have investigated how tyrosinase starts the grafting of peptides onto the amine-containing polysaccharide chitosan.

3.1.6 Photochemical grafting [13]

The photochemical grafting process occurs when a chromophore on macromolecules absorbs light, leading to an excited state that produces reactive free radicals. This encompasses processes known as electron-induced excitation, ionization, and dissociation. Should light absorption be inadequate, photosensitizers like benzoin ethyl ether can facilitate the process.

3.1.7 Plasma- radiation induced grafting

In this approach, plasma conditions are achieved through a slow discharge that exerts a similar effect as ionizing radiation. Plasma polymerization employs electron-induced excitation, ionization, and dissociation. The accelerated electrons produced in the plasma contain enough energy to break chemical bonds in the polymer backbone, leading to the formation of macromolecule radicals that initiate grafting.

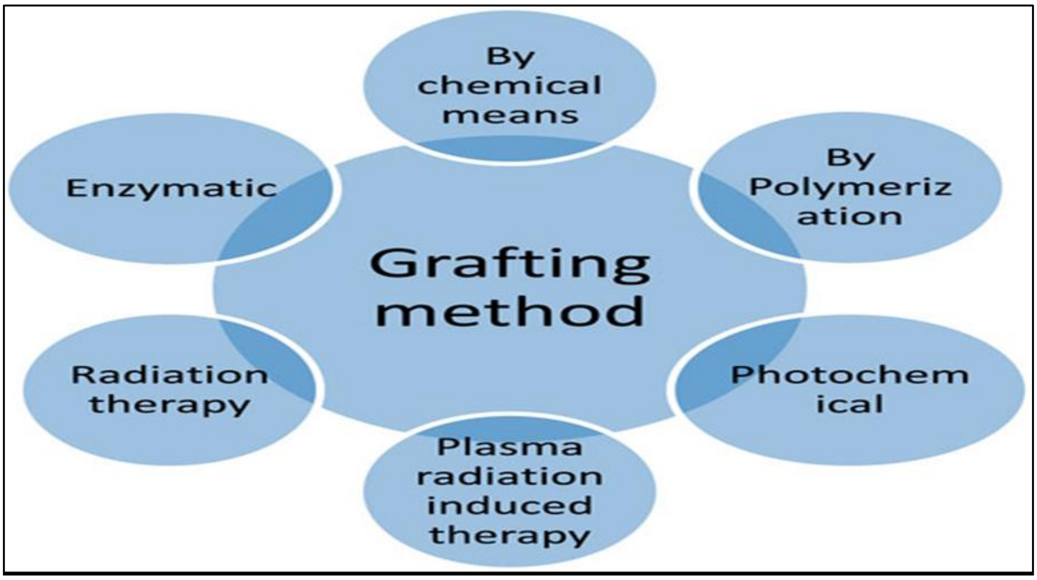

Fig 1: Methods of grafting

4. Microwave [7]

Microwaves are increasingly regarded as key tools in pharmaceutical research labs. Researchers have shown growing interest in microwave-assisted reactions due to their effective heating capabilities in laboratory settings. Microwave-assisted free radical polymerization is utilized to synthesize grafted natural copolymers, with or without initiators. Microwaves emit electromagnetic radiation in the frequency range from 300 MHz to 300 GHz. When subjected to microwaves, polar or charged particles align with the microwave's electric field, which reverses direction at a rate of 2.4 × 10^9/s at a frequency of 2.45 GHz. The inability of charged or polar particles to align quickly enough with the shifting electric field generates friction, producing heat in the medium. Microwave reactions can occur both in solution and in dry conditions. However, in dry conditions, reactant mixing can be impractical.

4.1 Advantages of Microwave Grafting

- It improves copolymer formation, resulting in increased yields.
- The power of microwaves and irradiation duration can effectively control grafting percentages.
- Uniform heating is achieved throughout the material.
- The process is rapid and efficient.
- The resulting product is pure.
- The occurrence of homopolymer reactions is minimized.
- There is potential for superheating.
- Reaction times are reduced.
- Yields are higher.
- Side reactions are diminished.

4.2 Objective of Microwave Heating

The primary aim of microwave heating is dielectric heating. Dielectric materials are essential for microwave-induced grafting, acting as insulators that become polarized in response to an applied electric field. This electric field separates positive and negative charges in atomic nuclei. When the field is applied, electrons move freely through conductors, while in dielectrics, it only slightly displaces electrons from their usual positions, creating an electron dipole, thus polarizing the material. Dielectric materials fall into two categories: polar and nonpolar.

4.3 Synthesis of Graft Copolymer-Two methods [14]

- Conventional grafting
- Microwave grafting
  - Microwave initiated grafting
  - Microwave assisted grafting

4.3.1 Conventional grafting

Conventional grafting for natural gums typically utilizes radical polymerization techniques [15]. This method generates active sites in the form of free radicals or adds functional groups onto the polymer backbone with the help of redox initiators like Ethylene-diamine tetraacetic acid (EDTA)/CAN, or ferrous ammonium sulfate/potassium persulfate. A major limitation of this technique is the risk of degrading the polysaccharide backbone, which also leads to limited control over the molecular weight distribution of the grafts. The process tends to be time-consuming, and it often results in products with high cross-linking density and fewer adsorption sites. The conventional grafting process can occur through three mechanisms: grafting through, grafting to, and grafting from.

It proceeds by three ways:

- The "grafting through" method involves copolymerizing already prepared vinyl-functionalized polysaccharides with co-monomers.
- The "grafting from" method grows grafts directly from the polysaccharide backbones and is often considered the most effective technique.
- The "grafting to" method chemically bonds preformed polymer chains to a surface

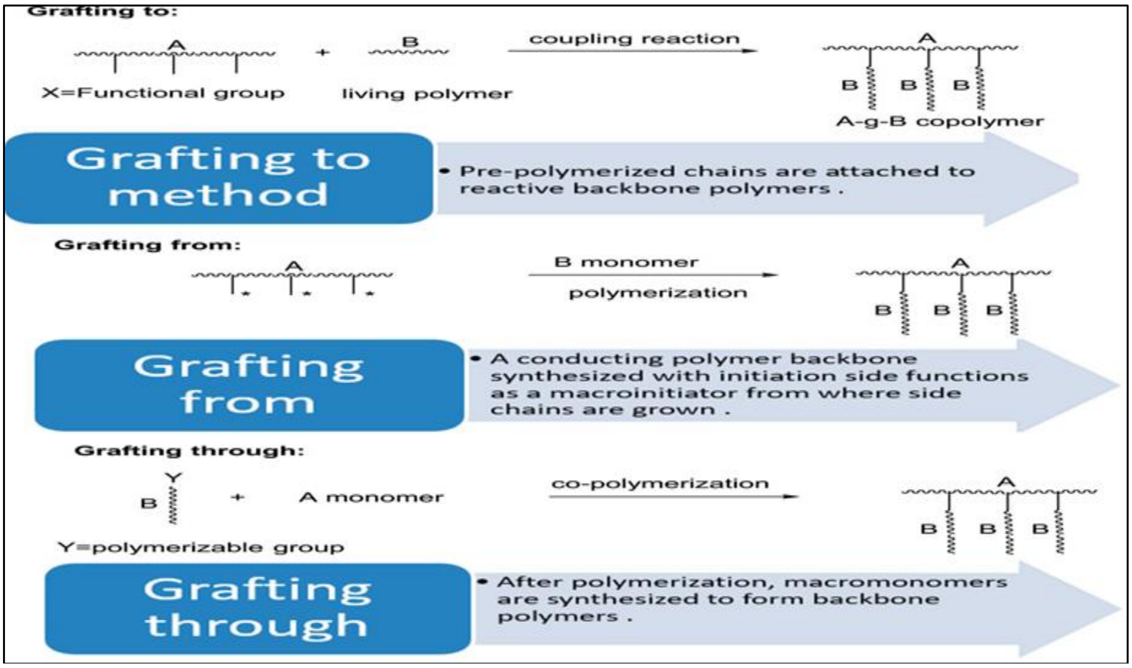

Fig 2: Techniques of polymer grafting

Disadvantages of conventionally grafting

- Requires an inert atmosphere.
- Yields lower grafting due to homopolymer formation.
- Prolonged reaction times.
- Limited to solution setups only.
- Requires an initiator.
- Slower heating rates.

To address these issues, the microwave grafting technique has proven to be an efficient alternative, showing increased application across various fields [16].
Based upon the reaction conditions, the grafting reactions can be classified into two categories

- Grafting in aqueous solution
- Grafting at solid support

4.3.2 Grafting in Aqueous Solution

- Two approaches are employed[17]
- Microwave initiated grafting
- Microwave assisted grafting

4.3.2.1 Microwave Initiated Grafting

In microwave-initiated grafting, no initiators are used, although hydroquinone may act as a radical inhibitor to prevent unwanted grafting reactions, despite modern instrumentation like ESR not confirming free radicals' presence in the mixture. The heating in this method arises from the dipolar relaxation of the solvent (water) and the localized rotation of polar functional groups of polysaccharides during microwave exposure [18] Microwave-initiated grafting has emerged as a promising technique in material science, offering a fast and efficient way to functionalize surfaces. This innovative method has attracted significant attention for its ability to facilitate grafting under mild conditions, reducing energy usage and minimizing reaction times while enhancing product yield.

4.3.2.2 Microwave Assisted Grafting

In this process, the ions will produce by addition of external redox initiators to the reaction mixture, and their presence enhances the ability of the aqueous reaction mixture to convert the microwave energy to heat energy.Under the influence of microwave dielectric heating, the generation of free radicals from the initiators facilitates the grafting reaction and there are various methods of graft copolymer formation depends on the different methods of free radicals generation on the backbone of polysaccharide.

The methods are:

- **Pre-irradiation:** Free radicals are generated through the polymer backbone irradiation in the vacuum or in presence of inert gas that followed by treating polymer substrate with the monomers in the liquid or vapor state or as a solution in a suitable solvent.
- **Per oxidation:** Based on the nature of polymeric backbone and irradiation circumstances, large amount of polymer is subjected to high-energy radiation in the presence of air or oxygen to form hydro-peroxides or di-peroxides At higher temperature, stable peroxy products are treated with monomer then decomposition of peroxides to radicals initiate approach, polymers and monomers are irradiated at the same time t grafting.form free radicals.
- **Mutual irradiation:** The polysaccharide and the monomer are irradiated simultaneously to form free radicals and grafting. The product created is a combination of homopolymer and graft copolymer.

4.3.3 Grafting on solid support

Some type of reactions have been established out between supported reagents which are transparent in nature like Silica, alumina etc.in dry media or solvent free procedures which gives clean, easy to perform, cheap, safe and environmental friendly conditions. Due to adsorption of reactant on solid support, the reagents are reacted neat and clean or are pre-absorbes on more or less microwave transparent technique like silica, alumina etc. Some of the difficulties like non uniform heating, mixing of the reaction temperature remain unsolved.

Advantages of microwave over conventionally methods

- Higher grafting yield is obtained
- Less reaction times
- It is possible in solid as well as in solution
- Initiator may or may not required
- Uniform heating
- Higher efficiency and process speed
- Product is pure
- Reduction of side reactions

Table 1: Analytical techniques utilized in characterization of grafted polymers

Analytical Technique	Characterization	Methodology	Applications
Fourier-Transform Infrared Spectroscopy (FTIR)	Identifies chemical functional groups introduced by grafting	Measures absorption of infrared light by molecular bonds	Confirmation of grafting by detecting functional groups; comparison of grafted and original materials.Qualitative analysis using FTIR revealed several stretching peaks where Cassava starch showed the peaks of oh stretching vibration whereas in grafted Cassava the OH is replaced by polysaccharide chain.[19]
Differential Scanning Calorimetry (DSC)	Examines thermal transitions (e.g., Tg, Tm)	Measures heat flow during heating/cooling of a material	Analyzes changes in thermal behavior and polymer transitions after grafting.[20] O Sreekanth Reddy *et al* reported that thermogram of both SA and KG showed two peaks of exothermic and endothermic whereas graft copolymer placebo microbeads showed the same two peaks in addition to these peaks one more peak is observed which indicated the successful formation of graft copolymer
X-ray Diffractometer (XRD)	Determines surface composition and chemical states	Analyzes the energy of photoelectrons emitted from the material surface	Surface chemistry analysis; quantification of grafting efficiency Sharma *et al* reported that XRD curve of ungrafted chitosan showed high relative intensities due to the crystalline region but in grafted XRD samples no such sharp peaks were observed which confirmed the amorphous nature of polymer due to grafting.[21]
Scanning electron microscope (SEM)	Studies surface morphology and topography	Uses electron beams to generate high-resolution surface images	Observes surface roughness, distribution, and homogeneity of grafts [22]. The surface of grafted cassava showed a rough surface and presence of large number of pores indicating aporousstructure.As radiation power increases more free radicals are generated increase the graft sites. More polyacrylamide will be grafted on cassava starch causing more rough and porous surfaces on the morphological appearance.
Transmission electron microscope (TEM)	Provides high-resolution imaging of internal structure	Transmits electrons through thin samples for detailed imaging	Studies internal morphology and nanoscale distribution of grafted chains
Nuclear Magnetic Resonance (NMR) Spectroscopy	Determines molecular structure and composition	Detects nuclear spins in an external magnetic field to assess chemical structure	Confirms the molecular structure of grafted polymers; identifies grafted chains.[23]
Swelling study	Assesses the degree of swelling and polymer network interactions	Measures the change in dimensions or mass of a polymer when exposed to a solvent	Evaluates the crosslinking density, network structure, and swelling behavior of grafted materials[24]
Elemental Analysis (CHN Analysis)	Quantifies elemental composition	Measures the content of carbon, hydrogen, and nitrogen in a sample	Used to determine the elemental composition of the grafted material and verify grafting efficiency. Gumghatti has no nitrogen.However, all the grades of grafted gum ghatti have nitrogen. The presence of nitrogen is the proof of grafting
Molecular Weight (MW) Analysis	Determines the molecular weight distribution	Uses techniques like GPC, light scattering, or viscometry to measure the molecular weight of the polymer	Analyzes the distribution and average molecular weight of grafted polymers

Here's a table summarizing the analytical techniques used for the characterization of grafting techniques:

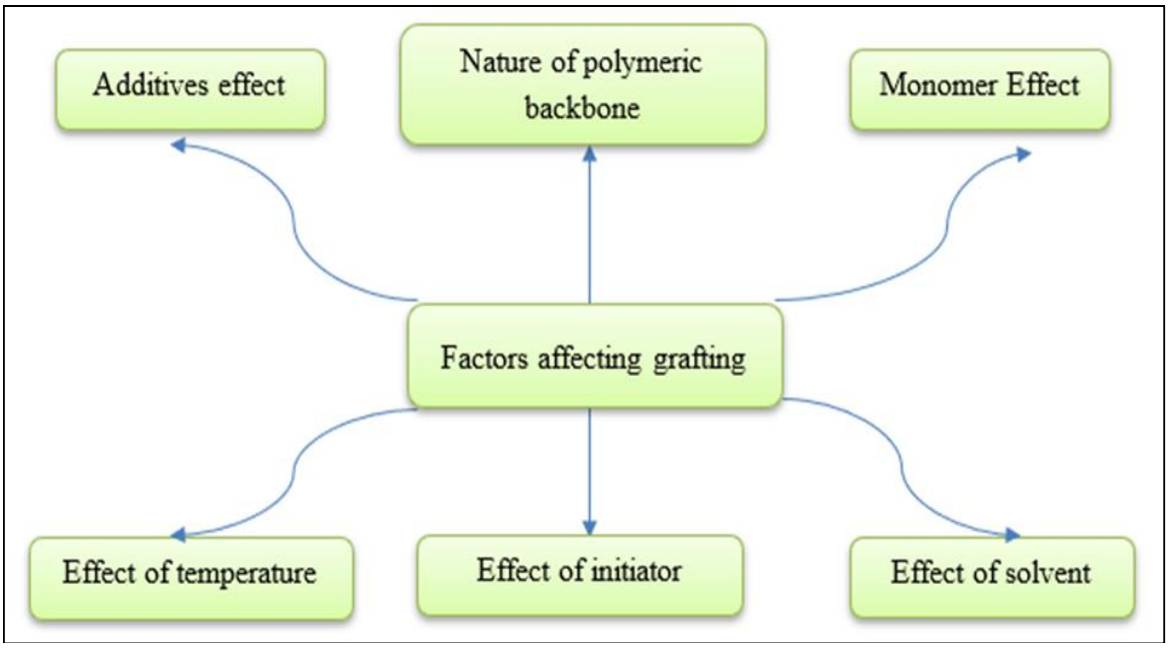

Fig 3: Factors affecting grafting

Factors affecting Grafting

5.1 Nature of backbone: Nature of backbone plays an important role in grafting. There are two types of natural polymeric backbone i.e. physical nature and chemical composition both have important role in the process of grafting that means covalent joining of monomer to a preformed polymeric backbone. NG *et al*. concluded that whereas cellulose is resistant to grafting reactions in water owing to its insolubility, due to the immense size of the polymeric chain bonding between the amino residues, the cystine linkages and intra-molecular H-bonding in wool are responsible for shaping and setting characteristics. In the presence of UV light, oxidative reactions are initiated and free radicals are formed, leading ultimately to grafting if monomers are present.

5.2 Effect of monomer: The reactivity of monomer is very important in grafting and it depends upon various factors like polar and steric nature and swell ability of backbone in the presence of monomer and concentration of monomer. Sharma *et al* reported that with the increase in the concentration of Acrylic acid grafting and grafting efficiency was observed to be increased but with the further increase in the concentration of Acrylic acid decrease in grafting was observed. This is predicted as at higher concentration of monomer chances of termination of polymeric radicals to form homopolymer increases.

5.3 Effect of solvent: The solvent used can influence the grafting process by affecting the solubility of the monomer and the mobility of reactive species. The chemical nature of the solvent and its interactions with the monomer and backbone polymer are important considerations [25]. Lele *et al* reports that natural polymer is soluble in water but with the decrease in the polarity and increase in the molecular weight of alcohol and increase in the dielectric constant the solubility of grafted polymer gets decreases.

5.4 Effect of initiator: The choice of initiator and its concentration are crucial in controlling the grafting process. Initiators decompose to form free radicals, which initiate the grafting reaction. Both the type of initiator and its concentration can affect the degree of grafting [26]. Sheetal Jha *et al* reported that initially there is increase in the efficiency with increase in the initiator but after certain time initiator concentration has been achieved an increase in the initiator level will decrease the conversion of grafted monomer. It showed that further increase in the initiator concentration leads to deacrease in grafting as cerric ion is agood terminator and cerric ion at higher concentration causes the termination of grafted polymeric chains.

5.5 Role of additives on grafting: The presence of additives such as metal ions, acids and inorganic salts can modify the grafting process. These additives can influence the molecular weight of the grafted chains and the overall architecture of the copolymer. Even though some additives may enhance the monomer or backbone reaction to increase the grafting efficiency, the reverse will be happen if the reaction between the monomer and the additive is existing.

5.6 Effect of temperature: Temperature affects the kinetic energy of molecules, influencing the rate of grafting reactions. Studies have shown that increasing the temperature up to a certain point can enhance grafting efficiency, likely due to increased mobility of reactive sites. Sharma *et al* reported that temperature plays an important role as with the increase of temperature i.e. above 70 ºC it might deteriorate the product by accelerating various hydrogen abstraction and chain transfer reactions and lead to decrease in grafting.

6. Applications of grafted polymer in drug delivery

In the realm of modern medicine, drug delivery system plays a pivotal role in ensuring the efficacy, safety and targeted action of pharmaceutical compounds within the body.

6.1Controlled Release Systems

- **Polymer Grafting for Controlled Release**: Various grafted natural polysaccharide had been utilized to synthesize different types of drug delivery system. The advantage of controlled drug delivery is to maintain optimum concentration. Grafted polymers can be used to create drug delivery systems that release drugs at a controlled rate over an extended period. The polymer grafts can be designed to degrade or swell in response to specific environmental conditions (e.g., pH, temperature), enabling sustained drug.Bhagwat Durgacharan *et al* reported that propranololHCL drug was released from modified neem gum showed significant swelling and water retention capacity. Significant swelling of the matrix caused slower diffusion of drug [27].

6.2 Buccal Drug Delivery [28]

- Buccal drug delivery has become an efficient and effective substitute to other traditional paths. This route of administration is beneficial for pediatric and geriatric patients [29]. According to Dhiman *et al* some natural polymers are used as mucoadhesive buccal drug delivery. Modified polymer was used to enable controlled release of ZLP via buccal route which avoid extensive hepatic metabolism, increasing bioavalibility, and avoiding early morning awakening without the use of middle –of-the-night dose. Grafted *Moringa* gum disks demonstrated a longer *mucoadhesion* time when compared with ungrafted Moringa gum.[30]

6.3 Nanoparticle-Based Drug Delivery

- **Polymer Nanoparticles**: Grafted polymers are commonly used in the synthesis of nanoparticles, which can encapsulate drugs and protect them from degradation in the body.[31] The grafted polymer chains can influence the stability, release rate, and targeting properties of the nanoparticles.[32]
- **Example**: Polymeric micelles, liposomes, and dendritic polymers can be designed using grafted polymers to encapsulate hydrophobic drugs, improving their solubility and bioavailability.

6.4 Gastro retentive drug delivery

- Patil *et al* reported that Acrylamide grafted neem gum was prepared for delivery of ciprofloxacin hydrochloride for the development of tablet GFDDS from neem gum by using microwave assisted method. Swelling character maintains the gastric fluid allowing gastric retention of tablets for longer duration of time.[33] The release data had shown the drug release was by swelling diffusion controlled system. Singh *et al* developed the matrix tablet of metoclopramide hydrochloride for increasing its bioavailability by extending gastric residence time.

6.5 Sustained drug delivery system
- Kaur *et al* concluded that grafted karaya gum proved to be successful and better polymer as compared to ungraftedkaraya gum in the formulation of sustained drug delivery system.(Jadach, B *et al*, 2022)

6.6 Waste water treatment
- Water is polluted chemically from wide range of toxic pollutants clay metals, aromatic molecules and dyes. These materials create severe environment problems for humans and other species.Therefore it requires recent technology that can reduce the toxic pollutants in water. Water pollution has also become the major source of concern for most of industries.Mervette *et al* reported that Biodegradability low cost and sustainable natural gum (GG) polymer make it favoured to formulate grafted hydrogel as an efficient alternative to biodegradable adsorbent for a sustainable system of waste water treatment. Mahto *et al* grafted guar gum with Itaconic acid using microwave irradiation technique (Mahto, A. *et al*, 2021)

7. Future trends
There is large number of polymers available in nature. Natural polymers as 'green approach' has been utilized as various dosage form in various drug delivery system. Different novel and conventional dosage forms have been prepared by using natural polymer. But there is need to explore the scope of grafted natural polymer gums and to improve their applications in industries. It has been evaluated from the studies that natural polymer gums and their derivatives after grafting enhances its properties by overcome their drawbacks. Hence there is need to reveal their applications in biomedical fields. In future various attempts are expected to synthesize more natural polymer gum based novel drug delivery system.

8. Conclusion
Natural polysaccharide owes to be better to synthetic polysaccharide-based drug delivery. The different types of dosage forms have been synthesized and put to use by employing natural polymers as a "green approach" for diverse drug delivery applications. Grafted polymers offer significant potential in enhancing drug delivery systems by providing controlled release, improving stability, and enabling targeted delivery. Their flexibility allows for the development of sophisticated delivery platforms that can be fine-tuned for specific therapeutic needs, improving the efficacy and safety of various drugs. In this review, it can be concluded that graft copolymerization is a novel technique and there are various applications in the field of pharmaceutics, drug delivery, microspheres, nanoparticles, etc.In this review, we discuss the grafting techniques and applications of polysaccharide grafted copolymers. From the findings, it can be concluded that polysaccharide grafted copolymers are an effective and innovative approach with diverse applications, including drug delivery, adsorption, textile and tannery waste treatment, domestic and sewage wastewater management, as well as agricultural uses, all benefiting humanity.

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