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A REVIEW ON TECHNIQUES FOR GRAFTING OF NATURAL POLYMERS AND THEIR APPLICATIONS

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Abstract

Polymer science have gained the advancement for improving the range of applications. The grafting techniques of natural gums or polymers have been modified for monomers production to transport enhanced polymeric material. These modifications are important against various challenges. It provides enhanced stability, compatibility, flexibility, and rigidity for the synthesized/modified co-polymer. This review enumerates the grafting concept, grafting modification, grafting techniques and applications of grafted natural gums in drug delivery.

Keywords: Grafting, Natural gums, co-polymer, drug delivery

Introduction

The natures are gifted with the wide variety of materials to human beings for balancing the healthiness of all living things. Polymers form the core of drug delivery system for providing the weight, consistency and volume or the administration of the drug (Bhattacharya *et al.,*1998). Due to the complexity of polymer structure that needs to be arise for extensive understanding of the surface and bulk properties of polymers which can show the desired functionalities (Sah *et al.*, 2016). In drug delivery, both natural and synthetic polymers are used. Natural gums are derived from the seeds or tubers of plants & seaweed which consist of multiple sugar units linked together to form large molecules. Intensive research on natural polymers due to their sustainability, biodegradability & biosafety (Yvon, 2014).

Advantages of natural gum

- The gums should be biocompatible or non-toxic in nature.
- Naturally available biodegradable polymers are produced by living organisms. Gums are truly renewable source & no adverse impact or environmental health.
- They are low in cost & easily available. Their production cost is lowered than synthetic material.
- The collection of natural gums is easy in different season & in the large quantities due to simple production processes involved. In developing countries, governments promote the production of plants containing gums because of the wide application a variety of industries.
- As compared with synthetic gums the chances of adverse effect are fewer with natural material (Mukherjee *et al.,* 2008).

Disadvantages of natural gum

Microbial contamination- Microbial contamination accumulated natural material due to 10% or more moisture content present in the gums which can be prevented by proper handling & the use of preservatives.

Reduced viscosity on storage- Natural gums come in to contact with water then increased the viscosity of the formulations due to their complexity in the nature of gums and reduced the viscosity of formulation after storage (Mukherjee *et al.,* 2008).

Applications of Natural gum

Table 1: Natural gums gaining importance for grafting.

Common Name	Botanical name	Family	Plant parts	Pharm. application	Therapeutic application	Adverse effects	References
Acacia	*Acacia senegal*	Legumino-Sae	Bark	Osmotic drug delivery	Dental plaque, weight loss	Allergic reaction, respiratory problem, skin lesions	(www. Acacia gum. 2017)
Bhara gum	*Terminalia bellericaroxb*	Combret-aceae	Plant bark	Microen capsulation	Peptic ulcer disease	Diarrhea, dizziness, Headache, anorexia	(Nayak *et al.,* 2008).
Guar gum	*Cyamompsis tetragan-olobus*	Legumino-seae	Seed	Colon targeted drug delivery, micro spheres, stabilizing, thickening, disintegrants agent	Constipation, Diarrhea, Diabetes	Blockage of esophagus & Intestine, gas production, GI Obstruction	(Baveja *et al.*, 1997; Patel *et al.*, 2014; Chourassia *et al.,* 2004).

Gellan gum	*Pseudomonas elodea*	Legumino-Seae	Non-pathogenic bacteria sphingo monas elodea from lactose or glucose	Ophthalmic drug delivery, sustaining agent, beads, hydrogels, thickening agent,	Constipation, low cholesterol	Abdominal bloating, flatulence, loose stools or diarrhea	Nutrients (Review. Com/Fibre gellan gum. 2016)
Karaya gum	*Sterculia urens*	Sterculiac-Eae	Exudates of sterculia tree	Muco-adhesive and Bucco adhesive, Emulsifier, stabilizing, thickening agent	Bulk forming laxative to treat constipation	Obstructs intestine, Diarrhea, reduce the absorption of other medicine	(Jagdev, 2014).
Locust bean gum	*Ceratonia siliqua*	Legumino-seae	Seed of plant	Controlled delivery, implants, beads, gels, Nanoparticle	anxiety	Increased gas low BP	(Kaity *et al.,* 2013)
Mucuna gum	*Mucuna flagillepes*	Papilliona-ceae	Seed	Micro spheres	Diabetes, analgesic, blood purifier, gout, gonorrhaea	Vomiting, Hallucination, insomnia, dementia, anorexia, diarrhea	(Deokar *et al.,* 2016)
Okra gum	*Hibiscus esculentus*	Malvaceae	Pods of plant	Binding agent, suspending agent, Hydrophilic matrix for controlled release drug delivery	Lungs & oral cavity cancer, improved digestion, vision, strengthen the bone	Kidney stone	(Mishra *et al.,* 2008)
Xanthan gum	*Xanthomonas lempestris*	-	Pulling bacteria	Pellets, controlled drug delivery system	Reducing cholesterol, lowering BP, Laxative, treating cancer	Intestinal inflammation, allergic reaction, ulcerative colitis	(Vterendruscolo *et al.,* 2005)

Grafting of polymers

Grafting: A graft is that unit attached to another by insertion or implantation so it becomes part of it. By graft techniques, chemical modification is obtained in natural and synthetic macromolecular moiety (e.g. carbohydrates, polyvinyl alcohol etc) that's novel research area of great interest. Graft polymerization is useful technique for modification in the properties of polymer and their modification by graft gums such as xanthan gum, Arabic gum, acacia gum etc (Vterendruscolo *et al.,* 2005).

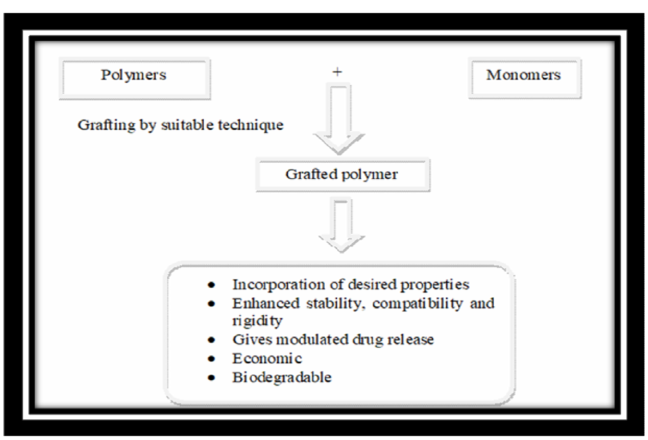

Fig. 1 : Steps of grafting technique and key benefits of grafting (Bhosale *et al.,* 2015).

Techniques/Method of grafting

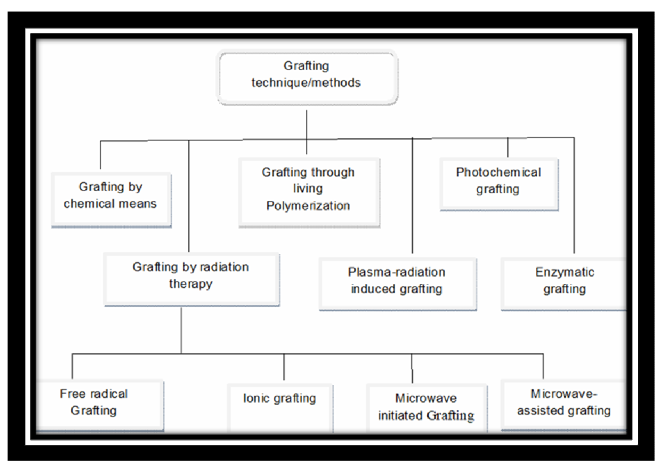

Fig. 2 : Techniques/Method of grafting (Bhosale *et al.,* 2015).

Chemical Means Grafting

In this technique, grafting can be initiated by chemical means through two main paths such as free radical grafting and ionic grafting and the initiator is extremely used in this grafting because it decides the rate of grafting process (Russell, 2002).

Grafting by living polymerization

The meaning of living polymer is that which holds its ability to proliferate for the extended period of time and growth occur to their highest size while chain transfer is still insignificant. When absorption of light is not sufficient the process can proceed by addition of photosensitizers such as benzoin ethyl ether (Coessens *et al.,* 2001).

Grafting through enzymes

In this technique, enzymes precede the grafting reaction (chemical or electrochemical). e.g tyrosine is able to convert the phenol into reactive o-quinone, which further undergoes successive non-enzymatic reaction with chitosen (Chen *et al.,* 2000).

Photochemical grafting

The photochemical grafting process is occurred with the formation of reactive free radical when chromophore on macromolecules absorbs light and undergoes an excitation state to form reactive free radical. If the absorption of light is not sufficient, the process may be accomplished by photosensitizers such as benzoin ethyl ether (Kubota *et al.,* 2001).

Plasma- radiation induced grafting

In this technique, plasma conditions can be obtained through sluggish discharge put forward the same potential as with ionizing radiation (Uchida *et al.,* 1990).

Grafting initiated by radiation technique

Free-radical grafting

In this grafting, polymer can be formed by irradiation of macromolecules which causes homolytic fission. The lifespan of free radicals depends on the nature of the polymer backbone. Grafting occurs by radiation technique in 3 different ways:

Pre-irradiation- Free radical 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.

Peroxidation- 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 grafting.

Mutual irradiation- In mutual Irradiation approach, polymers and monomers are irradiated at the same time to form free radicals (Coviello *et al.,* 1998).

Ionic grafting

This type of grafting can occur through an ionic mode using some initiators that's include alkali metal suspensions in a Lewis base liquid, organ metallic compounds, and sodium naphthalenide.

e.g alkyl aluminum (R3A1) and backbone polymer in halide form (AC1) act together forming carbonium ions along the polymer chain, leading to copolymerization. The reaction proceeds through cationic mechanisms (Mishra *et al.,* 1980).

$$ACl + R_3Al \rightarrow A + R_3Cl^-$$

$$A{+}{+}M \rightarrow AM{+}{-}M \rightarrow \text{graft copolymer} \qquad (1)$$

Cationic catalyst BF_3 can be implemented for this purpose. The anionic mechanism can also involved the grafting process. e.g sodium ammonia or methoxide of alkali metals form alkoxide of polymer (PO-, Na^+) which react with the monomer to form the graft copolymer,

$$P\text{-}OH + NaOR \rightarrow PO\text{-}Na{+} + ROH$$

PO^- + M → POM--M → graft co-polymer (Ng L-T *et al.,* 2001; Xie W *et al.,* 2002).

Microwave Initiated grafting

In this technique, no initiators can be used. A free radical grafting mechanism in which Hydroquinones are used as inhibitor to slow down the reactions of grafting under microwave irradiation (Pal *et al.,* 2011).

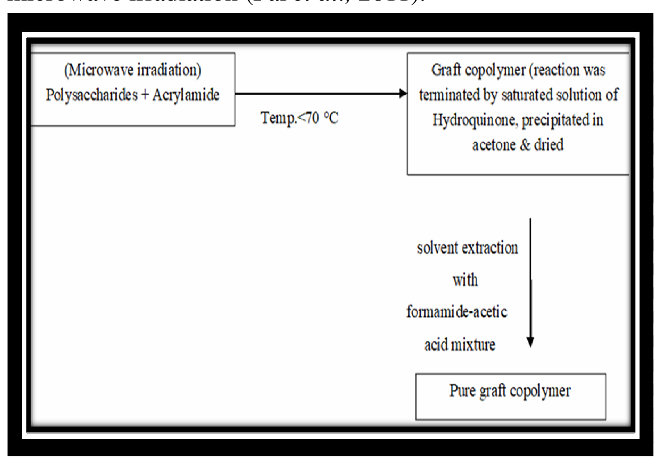

Fig. 3 : General method of Microwave initiated technique (Kaity *et al.,* 2013; Pal *et al.,* 2011).

Microwave Assisted Grafting

In this technique, redox initiator can be used. In the aqueous reaction mixture by the addition of initiators produces the ions to change microwave energy into heat energy. Under control of microwave dielectric heating, grafting reactions are helped in generation of free radicals from initiators (Pal *et al.,* 2011).

Analytical techniques utilized in characterization of grafted polymers

Table 2 : Analytical techniques utilized in characterization of grafted polymers

Analytical technique	Characterization	Detecting source	References
FTIR	Specific functional groups are added in to blend as a result of grafting. For characterization, 0.5 to 1 mm thick films of natural gum and graft copolymers can be prepared & analyzed by FTIR.	FTIR analyzer	(Kaity *et al.,* 2013)
13C -solid-state NMR	13C -solid-state NMR technique is used for hydrocarbon backbone characterization. Around 300 mg of sample is required to insert the ceramic rotor of NMR spectrophotometer for its characterization.	NMR spectrophotometer	(Vijan *et al.,* 2012)
DSC	DSC can be checked the purity and melting point of polymers & to obtain DSC thermograms of compounds. 3-5 mg samples are heated from 10 °C to 300 °C under nitrogen purge (50 ml/min) at a heating rate of 10 °C/min	DSC thermograms	(Vijan *et al.,* 2012)
Elemental analysis	PCHN 2400 microanalyzer is used for the elemental analysis. The samples of native gum and all the graft copolymer can be examined for contents. An elemental composition such as C, H, N content can be calculated	Microanalyzer	(Vijan *et al.,* 2012)
MW analysis	MW analysis can be used to determine the gain in molecular weight after grafting characterized.	MW analyzer	(Nishioka *et al.,* 1983)
Powder X-ray diffraction (PXRD)	X-ray diffractometer is used to record PXRD of the sample. The native gum and grafted gum can be studied.	X-ray diffractometer	(Kaity *et al.,* 2013)
Swelling Study	Two different media can be used for measurement of equilibrium swelling index. Weigh the amount of material (W1) is immersed in 50 ml of buffer (pH 1.2 to 6.8 and keep left for 2hr. After swollen quantity is recovered & excess water is removed with tissue paper and reweigh (W2). Swelling index (SI) = W2-1/W1×100	Swell test apparatus	(Vijan *et al.,* 2012)
Acute oral toxicity study	Acute oral toxicity study of grafted gum can be performed as per Organization of Economic Co-operation and Development guidelines using animals.	Animal used	(Vijan *et al.,* 2012)

Factor affecting the grafting:

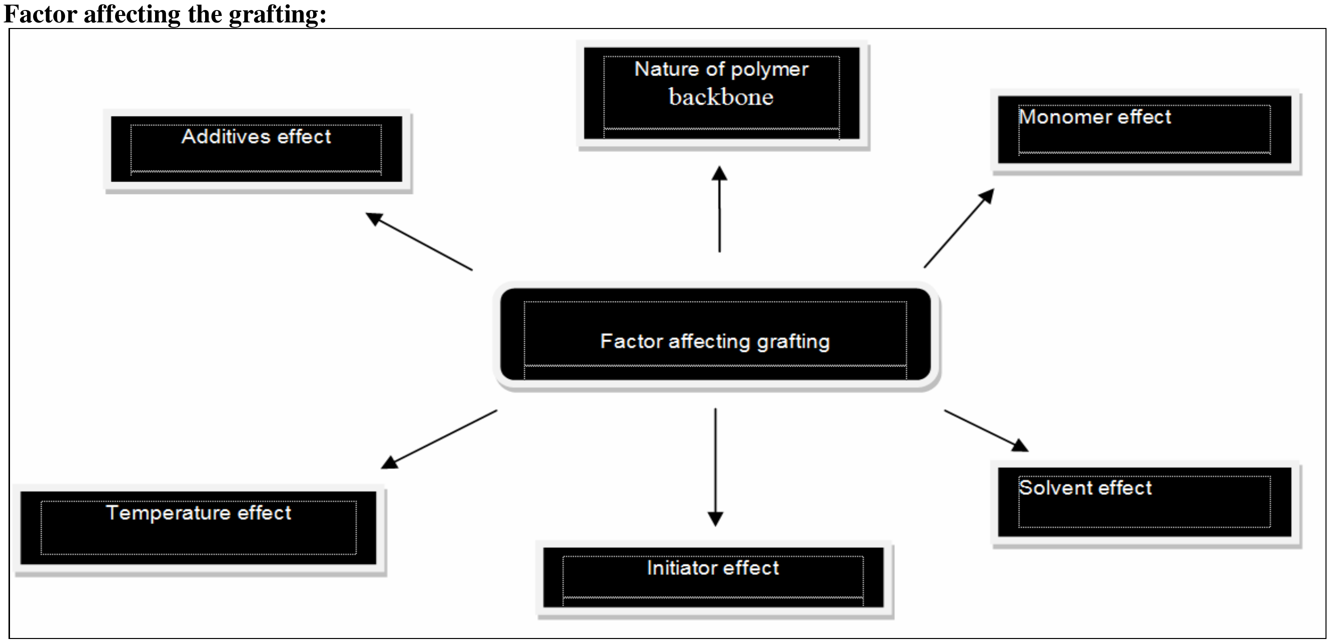

Fig. 4 : Factors affecting the grafting (Bhosale *et al.,* 2015).

Nature of Polymer Backbone

The nature of polymer backbone are 2 types physical nature and chemical composition which have important role in process of grafting that involves the covalent connection between monomer preformed polymeric backbone. Ng *et al.* have found that due to its insolubility, cellulose does not support the grafting reactions in water because the large size of polymeric chain bonding between amino residues in wool, cystine linkages, and H-bonding may be accountable for its setting characteristics (Ng *et al.,* 2001).

Monomers effect

The monomers reactivity is necessary for grafting process such as the nature of polymer backbone, which can be depends on various factors like polar and steric nature, swellability of backbone in the presence of monomers and concentration of monomers (Ng *et al.,* 2001).

Solvent effect

Solvent system is act as carrier through which monomers are passed to the surrounding area of backbone in grafting mechanism. The criteria for the selection of suitable solvent depends on numerous parameters such monomer solubility in the solvent, swelling properties of backbone, solvents miscibility (if more than one is used) and generation of free radical in presence of solvent (Coessens *et al.,* 2001).

Initiator effect

The natures of the initiator have a deep impact on the grafting process. Except for radiation technique, all other chemical grafting techniques required an initiator. Hence initiator nature, concentration, solubility, and function should be considered. e.g initiators like azobisisobutyronitrile, ceric ammonium nitrate and potassium persulfate (Nishioka *et al.,* 1983).

Temperature effect

Temperature is the important factor for controls the kinetics of graft copolymerization. With increasing temperature, grafting yield also increases until a limit is achieved due to faster monomeric diffusion in backbone which increases with the rising temperature (Sun *et al.,* 2003).

Additives effect

The presence of additives such as metal ions, acids, and inorganic salts affects the extent of graft copolymerization. Hence the reaction occur amongst the monomer and backbone has to fight with any reactions amongst monomer and additives (Chappas *et al.,* 1979).

Applications of grafted natural gums in drug delivery

Table 3 : Few Examples of grafted gums with Application in drug delivery

Gums and Mucilage	Plant parts	Modification Technique	Application	References
Karaya gum	Exudate of sterculia tree	Heat Treatment	Disintegrating agent	(Jagdev 2014; Babu *et al.,* 2002)
Acacia gum	Plant bark	Chemical modification	Disintegrating agent	(Trivedi *et al.,* 1986; www. Acacia gum. 2017)
Starch	Leaves of the plant, stored in chloroplast in the form of granules	Physico-chemical treatment of starch for modification	Disintegrating agent, Binding agent	(Baveja *et al*., 1997; Okafor *et al*., 2001, www.britannica.com)
Sesbania gum	Seeds of Sesbania	Chemical modification	Gelling agent	(Stehling *et al.,* 1998).
Guar gum	Seed of plant	Chemical modification	Film coating, disintegrating agent, hydrogel	(Chaurasia *et al.,* 2006; Singh *et al.,* 2014)
Okra gum	Pod of plant	Chemical modification	Controlled drug delivery	(Sah *et al.,* 2016; Mishra *et al.,* 2008; Zaharuddin *et al.,* 2014)
Gellan gum	Non-pathogenic bacteria *Sphingomonas elodea*	Microwave-assisted grafting	Sustained drug delivery,	(Nutrients Review.Com/Fibre gellan gum. 2016; Deogade UM *et al.,* 2012)
Cashew gum	Bark exudate	Grafting by free radical polymerization	Sustained drug delivery	(Da Silva *et al.,* 2007; Kumar *et al.,* 2012)

In recent years, different verities of grafted gums have been used in the formulation of different types of drug delivery system. But some problems occur after modification of natural gums such as uncertain hydration rate, fall in viscosity, microbial contamination etc. In the field of Pharmaceutical technology, need arise for many efforts for alter the physical& chemical properties of polymeric materials. In recent years, researchers have done research works with respect to grafting modification.

Shruthi *et al.* (2016) by using microwave irradiation technique prepared acrylic acid grafted guar gum nano-composites using Silane-modified nano-clay. The grafting was confirmed via infra-red spectroscopy while XRD diffractograms suggested exfoliation of modified nano-clay in guar gum grafted acrylic acid. The reaction parameters, swelling characteristic, pH on swelling capabilities has been assessed (Shruthi SB *et al.,* 2016).

Zauro *et al.* (2016) formulated poly (diallyldimethylammonium chloride) grafted locust bean gum using ammonium peroxydisulfate as an initiator by microwave irradiation technique and N, N-methylene-bis-acrylamide as a crosslinker. The grafted gum was evaluated for the removal of anionic dye “indigo carmine” from aqueous solution. The adsorption data was observed to fit into Langmuir isotherm model and the maximum adsorption was found to be 35.12 mg/g gel.

Singh *et al.* (2011) studied the applicability of acrylamide grafted moth bean starch using Lamivudine was used as a model drug and its controlled release tablets were formulated using various concentration of grafted copolymer.

Mundargi *et al.* (2007) formulates the Metronidazole tablets using intrinsic developed graft copolymer of methacrylic acid with guar gum for colon targeted drug delivery. Drug release studies were performed in the simulated gastric fluid at pH 1.2 for 2 hr. and intestinal fluid at pH 7.4. Using Eudragit-L 100 (for enteric coating), the release of metronidazole was drastically reduced to 18-24%.

Varshosaz *et al.* (2006) have been developed sustained release matrix tablets of extremely water soluble tramadol HCl using xanthan gum and guar gum as hydrophilic matrices which reported as non-toxic, easily available, cheap and suitable hydrophilic matrix systems against broadly investigated hydrophilic matrices (i.e., hydroxypropyl methylcellulose or carboxymethyl cellulose).

Conclusion

In recent times there have been various techniques are used to modification of natural polymers to obtain new product materials. In controlled & sustained drug release systems generally, modified polymers can be used. Natural gums also are modified to get a more qualitative product for designing the formulation. In the present and upcoming future, enormous scope of natural gums for the research program and grafting modification for the development of novel drug delivery systems.

Declaration of interest

The author is responsible for the content and writing of this article.

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