Grafting of Moringa gum by Acrylamide with Microwave Irradiation Technique for Enhancement of Control Release Tablet

Authors: - Ayush Kumar Mahato*, Sandip Prasad Tiwari, Naimish Nanda

Address: - Faculty of Pharmacy, Kalinga University, Kotni, Near Mantralaya, Naya Raipur, Chhattisgarh- 492101, India.

Corresponding Author:

Mr. Ayush Kumar Mahato

M.Pharmacy,

Faculty of Pharmacy, Kalinga University, Naya Raipur. (C.G.). INDIA

PIN- 492101

Abstract

The present research focuses on the chemical modification of Moringa oleifera gum through graft copolymerization with acrylamide using a microwave irradiation technique, aimed at enhancing its performance in controlled-release drug delivery systems. Moringa gum, a natural polysaccharide known for its biodegradability, biocompatibility, and functional versatility, was selected as the base polymer. Grafting was initiated using ceric ammonium nitrate (CAN) as a redox initiator. The process parameters—such as reaction time, monomer concentration, and temperature—were systematically optimized to achieve efficient grafting. Microwave irradiation enabled uniform and rapid energy distribution, significantly improving the grafting efficiency, thermal stability, and swelling behaviour of the resulting polymer. The grafted moringa gum exhibited a grafting percentage of 257.23% and a grafting efficiency of 50.51%, confirming substantial incorporation of acrylamide into the gum matrix. The modified polymer was then utilized to formulate tablets using the wet granulation method, with starch serving as a binder and talcum powder as a lubricant. Uniform tablets were produced using a single-punch tablet press. Dissolution studies were conducted using USP Type II (paddle) apparatus in phosphate buffer (pH 6.8) at 37 ± 0.5°C. The tablets demonstrated 35% drug release in 15 minutes and 94% release within 60 minutes, indicating effective sustained-release behaviour. These findings demonstrate that grafted moringa gum, synthesized via microwave-assisted grafting, holds significant promise as a functional and sustainable excipient in advanced pharmaceutical formulations.

Keywords- Moringa Gum , Grafting, Microwave irradiation, Grafting Percentage, Grafting Efficiency

1. Introduction

Polymers are the fundamental building blocks of drug delivery systems, giving them their weight, consistency, and volume. In order to balance the health of all living things, nature has given humans a vast array of materials [(1)]. Oral matrix tablets keep on to be the most widely produced and utilized system, despite the development of several technologies to regulate the release of medications from a dosage form. The simplicity of their technology, affordability, ease of fabrication, and convenience are the reasons behind matrix systems' widespread use [(2)] Numerous naturally occurring plant-based excipients, including gums, mucilages, and supramolecular polysaccharides, have been thoroughly investigated and are used in the food, textile, pharmaceutical, cosmetic, and other industries for different goals [(3)]. The term "gum" refers to a family of naturally occurring polysaccharides that are known for their gel-forming or viscous solution properties. These polysaccharide gums are abundant raw materials extracted from plants and are extensively studied for their sustainable, biocompatible, and biodegradable properties. The molecular structure and chemical composition depend upon the source, extraction, and processing techniques. Plant gums are complex polysaccharides or carbohydrate polymers whose chemical makeup often derives from sugar monomers like starch, cellulose, hyaluronan, and alginate units. Natural gums usually come from plant tubers or seeds and seaweed, making them environmentally friendly natural polysaccharides.[(4-5)] Different sources result in gums with varying molecular structures and properties. These natural polymers are frequently employed in the production of paper[(6)], food[(7)] ,and cosmetics[(8)]. They also serve various functions, including as coagulants [(9)], thickening agents[(10-11)], drilling additives[(12-13)], suspending agents [(14)],matrices for nanomaterials[(15)], pharmaceutical adjuvants[(16)], and materials for textiles and dyeing[(17)]. Through extensive research on various plant-based gums, especially plant exudates and seed gums, valuable natural sources of complex polysaccharides have been identified. Due to their texture and stability, these gums are liked for various applications. Their molecular structure and chemical makeup affect their physical, chemical, and functional characteristics [(18)]. Natural gums have gained significant attention due to their numerous advantages in pharmaceutical and biomedical applications. They are biodegradable and renewable, making them environmentally friendly as they do not pose harm to either the environment or human health. Their biocompatible and non-toxic nature ensures safe interaction with biological systems, enhancing their suitability for various drug delivery systems. Additionally, natural gums are widely accessible and cost-effective, which further contributes to their popularity. Being composed of edible materials, they are well accepted by patients and enjoy a high level of public approval. However, the application of raw

gums is somewhat restricted due to their specific structural features and limited functional groups. They are prone to enzymatic degradation and microbial contamination, which can compromise their stability. During storage, the viscosity of their solutions or mucilage may decrease, and they often exhibit an uncontrolled rate of hydration, making them challenging to process and fabricate in their native form[19,20].

Moringa gum is derived from *Moringa oleifera* Lam., a plant belonging to the Moringaceae family, also known by various names such as the "kelor tree," "horse plant radish tree," and "drumstick tree"[21].The gum is collected from the exudate released at the site of injury on the plant's stem. Initially, this exudate appears white but gradually darkens to a reddish-brown or brownish-black color upon exposure to sunlight. Once dried, the gum is ground into fine particles and passed through a sieve of mesh size 80 to ensure uniformity[22]. To Isolate Moringa gum, a weighed quantity of the dried powder is stirred in distilled water at room temperature for about 6 to 8 hours. After stirring, the mixture is centrifuged to separate the supernatant from the residue. The residue is thoroughly washed, and the supernatants are pooled and diluted with distilled water up to a volume of 500 mL. Acetone, in approximately double the volume, is added slowly while stirring continuously with a magnetic stirrer to induce precipitation. This precipitate is repeatedly washed with distilled water and then dried under vacuum at a temperature of 50–60 °C[23]. Graft copolymerization of moringa gum involves the chemical modification of its backbone by attaching polymer side chains with distinct chemical characteristics. These side chains may consist of either a single type of monomer or a blend of two monomers. Grafting with a single monomer is generally a straightforward, one-step process, whereas dual monomer grafting requires multiple steps, including the sequential and continuous addition of both monomers.

Grafting percentage and grafting efficiency are two critical parameters used to evaluate the effectiveness of graft copolymerization. Grafting percentage (GP%) refers to the increase in weight of the polymer backbone due to the successful grafting of monomer chains. It indicates the extent of grafting and is calculated based on the weight gain of the polymer. On the other hand, grafting efficiency (GE%) represents the proportion of monomer that has successfully grafted onto the polymer backbone relative to the total monomer used in the reaction. It reflects how efficiently the monomer has been utilized without forming homopolymers or side products. While a high grafting percentage shows significant modification of the polymer, a high grafting efficiency indicates minimal monomer wastage and greater process economy. Both parameters together provide insight into the effectiveness and quality of the grafting process[24].

Percentage of Grafting (%GP):

$$\left(\frac{Weight\ of\ acrylamide\ in\ the\ grafted\ polymer}{\{Weight\ of\ original\ gum\}} \times 100\right)$$

Grafting Efficiency (%GE):

$$\left(\frac{Weight\ of\ grafted\ polymer}{Total\ weight\ of\ acrylamide\ used} \times 100\right)$$

2. Material And Methods

2.1. Materials

Table No. 1. List of materials

Materials	Category	Source
Moringa Gum Powder	Natural Polymer	Genius Nature Herbs Pvt Ltd , Theethipalayam, Tamil Nadu
Acrylamide	Monomer	Labogens Fine Chem Industry, Ludhiana , Punjab
Ceric Ammonium Nitrate (CAN)	Initiator	Universal Fine Chem, Howrah, West Bengal
Acetone	Solubilizer	Kalinga University, Naya Raipur
Distilled water	Solvent	Kalinga University, Naya Raipur

2.2 Method: For the grafting procedure, an accurate measurement of 17.66 g of Moringa Gum Powder was taken. To this, 17.67 g of Acrylamide was introduced as the grafting monomer. To initiate the polymerization process, 4.41 g of Ceric Ammonium Nitrate (CAN) was used as the initiator. Finally, 400 ml of distilled water was incorporated into the mixture to dissolve the components and support the grafting reaction.

2.2.1. Preparation of Grafted Moringa Gum

1. Preparation of Moringa Gum Solution- A carefully weighed amount of Moringa oleifera gum powder was dissolved in distilled water in a clean beaker. The suspension was heated at 40°C on a thermostatically controlled hot plate under continuous agitation for complete dissolution and formation of a uniform gum solution.

2. Addition of Initiator and Monomer-After complete solubilization of the gum solution, acrylamide (monomer) was added slowly under continuous stirring. Then, ceric ammonium nitrate (CAN) was added as a free radical initiator. The addition was carried out at 40°C temperature to ensure uniform reaction conditions.

3. Graft Copolymerization Reaction-The reaction mixture was shaken well at 40°C for 2 hours to facilitate acrylamide grafting on the moringa gum backbone. The beaker was put in a refrigerator as soon as the reaction time was over, and the mixture was cooled for 1 hour.

4. Cyclic Heating and Cooling-The heating (2 hours at 40°C) and cooling (refrigeration for 1 hour) procedure was repeated six times to facilitate improved gel formation and grafting efficiency. The reaction mixture had reached gel-like consistency at the end of the sixth cycle and hence proved successful grafting and crosslinking.

5.Purification through Solvent Extraction -For the elimination of residual unreacted monomer and impurities, the gel-like reaction mass was treated with acetone. The reaction mass was stirred well, and the formed impurity-containing acetone phase was decanted. The grafted moringa gum was finally drained from the reaction vessel after being purified.

Fig. 2 Moringa Gum Washed in Acetone

6. Drying Process-The obtained grafted gum was dried in intervals under controlled conditions in the microwave four times a day for two days. The material was kept overnight in an airtight container between drying to prevent water absorption from the environment. After the reaction time, transfer the beaker to a refrigerator and cool the gum mixture for 1 hour.

7. Particle size reduction-The grafted gum was dried and hardened properly and then milled into fine powder with a mortar and pestle for analysis and formulation.

8. Storage-The grafted moringa gum powder was kept in a dry, clean, air-tight container at ambient conditions until further use in follow-up formulation and assessment studies.

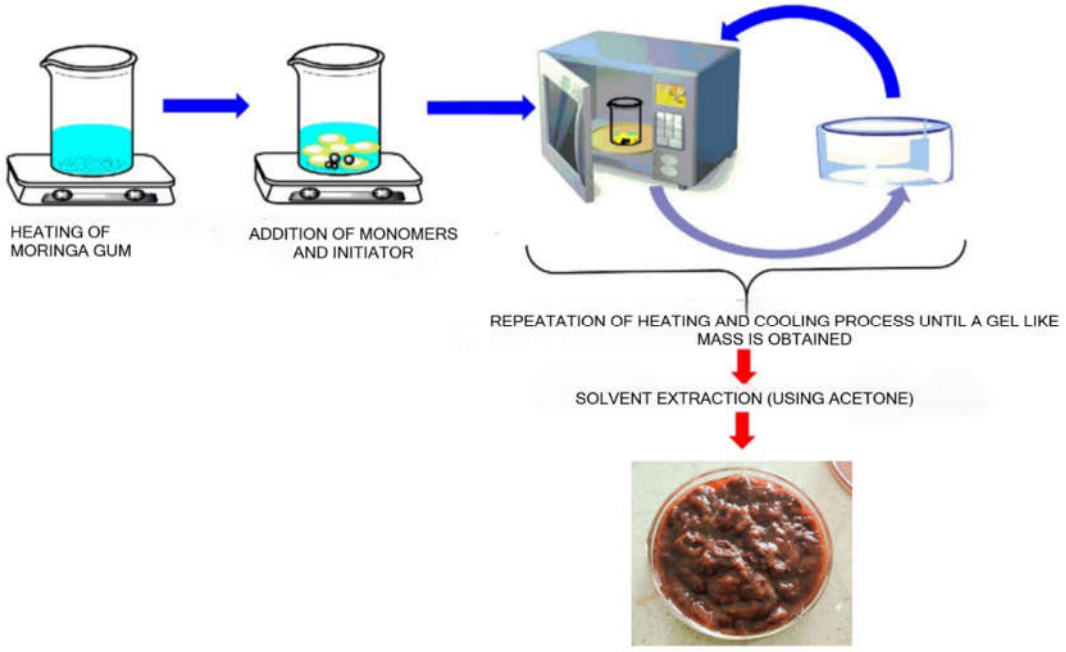


Fig. 1. Graphical Representation of Moringa Gum Preparation

2.2.2 Tablet Punching by Grafted Moringa Gum

Fig 4. Grafted Moringa Gum Tablet

The tablets were prepared by the wet granulation method, and grafted moringa gum was added to the formulation along with starch and talcum powder. Starch was added as a binder in the formulation because grafted moringa gum was not enough to provide adequate binding strength to facilitate proper tablet formation. Talcum powder was added as a lubricant and anti-sticking agent during compression.

In order to prevent powder sticking to punches when compressing, talcum powder was added to the mixture as a lubricant and anti-sticking agent. Sieve number 60 was used to sift all the ingredients and blend them together thoroughly to obtain a uniform powder mixture. Tablets were produced on a one-punch tablet machine using flat circular punches of 8 mm diameter. Compression was even, and no issue like sticking, capping, or breaking occurred.

Tablets were uniform in shape and possessed good hardness, which suggested starch was a good binder in the grafted moringa gum formula.

3. Result and Discussion

3.1. Moringa Gum Powder Solubility Test

To examine the solubility characteristics of moringa gum powder, a qualitative solubility test was conducted using three different solvents: distilled water, ethanol, and isopropyl alcohol. Approximately 1 gram of moringa gum powder was added to 100 mL of each solvent at room temperature (25°C) and stirred continuously. In distilled water, the gum powder began to swell and hydrate gradually, forming a thick, mucilaginous colloidal solution within 30 minutes. While the gum was only weakly soluble in water, it demonstrated excellent swelling and dispersion properties, indicating a strong hydration ability. when the gum powder was tested in ethanol and isopropyl alcohol, it remained largely insoluble, showing no signs of swelling or dissolution, even with vigorous stirring. The powder settled at the bottom, and no gel-like consistency was observed.

Fig.3 Solubility Test of Moringa Powder

Conclusion: The moringa gum powder was only dissolved in water, producing a hydrated gel with a thick texture, while it failed to demonstrate solubility in ethanol and isopropyl alcohol.

3.2. Grafting Percentage and Grafting Efficiency -Acrylamide was used as the monomer and a redox initiator system to perform the graft copolymerization of moringa gum. To start the grafting reaction, 17.67 grams of acrylamide were added to 17.61 grams of purified moringa gum, which served as the base polymer in this procedure. To get rid of any unreacted monomers or homopolymers, the product was carefully cleaned, filtered, and dried after the reaction was finished. The grafted polymer weighed 30.45 grams in the end.

The following formulas were used to determine the Grafting Percentage (GP) and Grafting Efficiency (GE):

Grafting Percentage (GP) =

$$\left(\frac{(Wg - W0)}{W0} \times 100\right)$$

Grafting Efficiency (GE) =

$$\left(\frac{(Wg - W0)}{Wm} \times 100\right)$$

Where:

$-$ W0 $=$ weight of moringa gum (17.61 g)

- Wg = weight of grafted polymer (62.92g)
- Wm = weight of monomer used (89.69 g)

Substituting the values:

GP =

$$\left(\frac{(62.92 - 17.61)}{17.61} \times 100\right)$$

=257.3%

GE =

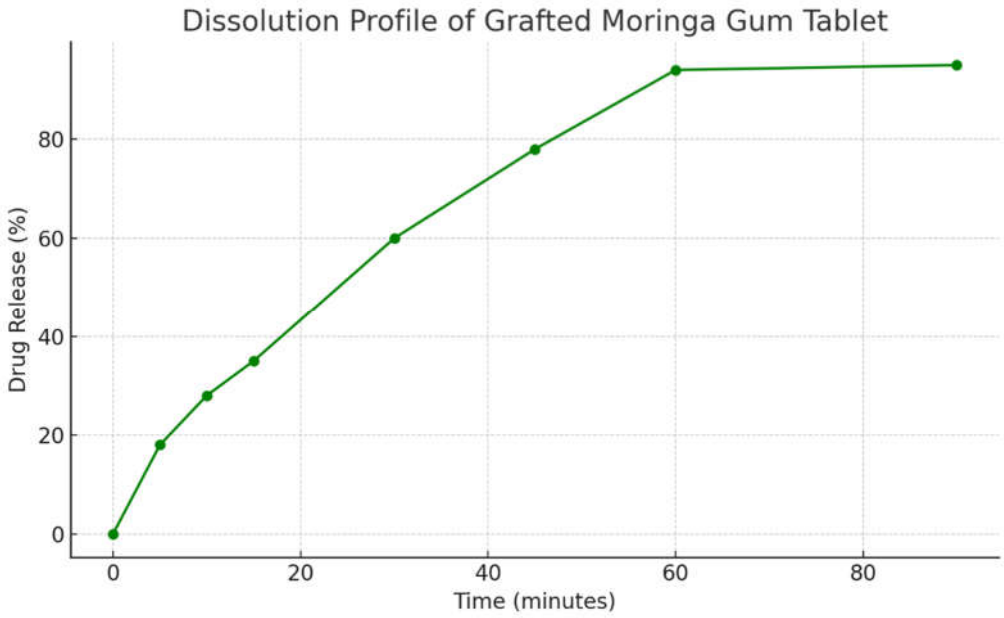
$$\left(\frac{(62.92 - 17.61)}{89.69} \times 100\right)$$

= 50.51%

The moderate grafting efficiency and percentage indicate that acrylamide was successfully grafted onto the backbone of moringa gum with little homopolymer formation. Because of its enhanced physicochemical characteristics, this modified gum may find application in regulated drug delivery systems.

3.3. Dissolution Testing- Dissolution was measured with a USP Type II (paddle) dissolution apparatus in 900 mL of 37 ± 0.5°C phosphate buffer, pH 6.8. The paddle speed was 50 rpm. Samples were collected at 5, 10, 15, 30, 45, 60, and 90 minutes, and an equal volume of fresh buffer was replaced every time.

The release of tablets was measured by a UV-Visible spectrophotometer. The tablets released 35% in 15 minutes and 94% in 60 minutes. The release profile showed that the formulation provided good sustained release of the drug. Grafted moringa gum may be the reason behind the controlled release pattern, and starch may have provided faster disintegration.



4. Conclusion

This effectively showcased the chemical alteration of Moringa oleifera gum through microwave-assisted graft copolymerization using acrylamide, with ceric ammonium nitrate serving as a potent redox initiator. The optimized grafting procedure achieved a remarkable grafting percentage of 257.23% and a grafting efficiency of 50.51%, signifying the successful integration of a synthetic monomer into the natural polymer framework. The modified gum exhibited enhanced physicochemical traits, particularly improved thermal stability and swelling properties, making it well-suited for controlled-release applications. The tablets formulated demonstrated outstanding mechanical strength and consistent drug release patterns, reaching 94% drug release within 60 minutes, affirming the capability of the grafted gum as a potential sustained-release matrix. Overall, this research not only highlights microwave-

assisted grafting as a rapid and effective method for polymer modification but also positions grafted Moringa oleifera gum as a promising, biodegradable, and sustainable excipient for innovative pharmaceutical dosage forms.

Graft copolymerization of Moringa oleifera gum using acrylamide and microwave irradiation proved to be an efficient method for enhancing the functional properties of this natural polymer. The application of microwave energy enabled effective and uniform heating, contributing to improved graft efficiency and substitution levels. The grafting percentage reaching 257.23% and a grafting efficiency of 50.51% not only demonstrate successful chemical modification but also suggest that this technique may be scalable for industrial applications. The selection of ceric ammonium nitrate (CAN) as the initiator was key to activating free radical sites on the polysaccharide structure, which allowed for effective grafting. The right combination of process parameters—such as irradiation duration, monomer concentration, and temperature—was essential for maximizing graft yield while maintaining the structural integrity of the polymer. The resulting grafted polymer exhibited improved thermal stability and swelling properties, which are advantageous for drug delivery systems, especially in hydrophilic matrix-type sustained-release tablets.

The preparation of tablets using the grafted gum encountered no processing issues, highlighting its favourable flow and compressibility. The dissolution characteristics showed a 35% release within the first 15 minutes, with nearly complete release (94%) after one hour, indicating the polymer's ability to control drug release over time. These results align with expected behaviour from grafted hydrogels, where swelling facilitates gradual drug diffusion. Compared to native natural gums, the grafted Moringa gum offered better release control, supporting the rationale for its chemical modification. Moreover, the biocompatibility and biodegradability of the base material ensure that the grafted excipient remains environmentally friendly and safe for pharmaceutical applications. These findings open up possibilities for developing innovative drug delivery systems using grafted natural polymers. Future research could explore in vitro performance, toxicity evaluations, and the polymer’s compatibility with a broader range of active pharmaceutical ingredients (APIs).

5. References

1. Bhattacharya A, Das A, De A. Structural influence on grafting of acrylamide based monomers on cellulose acetate**.**

2. Jani GK, Shah DP. Evaluation of mucilage of Hibiscus rosasinensis Linn as rate controlling matrix for sustained release of diclofenac. Drug development and industrial pharmacy. 2008 Jan 1;34(8):807-16.

3. George A, Shah PA, Shrivastav PS. Guar gum: Versatile natural polymer for drug delivery applications. European Polymer Journal. 2019 Mar 1;112:722-35.

4. Phillips GO, Williams PA, editors. Handbook of hydrocolloids. Elsevier; 2009 May 28.

5. Wang A, Wang W. Gum-g-copolymers: synthesis, properties, and applications. Polysaccharide based graft copolymers. 2013:149-203.

6. Nakauma M, Funami T, Noda S, Ishihara S, Al-Assaf S, Nishinari K, Phillips GO. Comparison of sugar beet pectin, soybean soluble polysaccharide, and gum arabic as food emulsifiers. 1. Effect of concentration, pH, and salts on the emulsifying properties. Food Hydrocolloids. 2008 Oct 1;22(7):1254-67.

7. Fabre B, Fiorini-Puybaret C, inventors; Pierre Fabre Dermo Cosmetique SA, assignee. Cosmetic composition containing a locust bean gum hydrolysate. United States patent application US 13/141,286. 2011 Oct 20.

8. Al-Hamadani YA, Yusoff MS, Umar M, Bashir MJ, Adlan MN. Application of psyllium husk as coagulant and coagulant aid in semi-aerobic landfill leachate treatment. Journal of hazardous materials. 2011 Jun 15;190(1-3):582-7.

9. Koocheki A, Mortazavi SA, Shahidi F, Razavi SM, Taherian AR. Rheological properties of mucilage extracted from Alyssum homolocarpum seed as a new source of thickening agent. Journal of food engineering. 2009 Apr 1;91(3):490-6.

10. Ibrahim NA, Abo-Shosha MH, Allam EA, El-Zairy EM. New thickening agents based on tamarind seed gum and karaya gum polysaccharides. Carbohydrate Polymers. 2010 Jun 11;81(2):402-8.

11. Hamed SB, Belhadri M. Rheological properties of biopolymers drilling fluids. Journal of Petroleum Science and Engineering. 2009 Aug 1;67(3-4):84-90.

12. Khalil M, Mohamed Jan B. Herschel-Bulkley rheological parameters of a novel environmentally friendly lightweight biopolymer drilling fluid from xanthan gum and starch. Journal of Applied Polymer Science. 2012 Apr 5;124(1):595-606.

13. Mahmud HS, Oyi AR, Allagh T. Evaluation of the suspending properties of khaya senegalensis gum in paracetamol suspensions. Nigerian Journal of Pharmaceutical Sciences. 2009 Mar;8(1):128-34.

14. Sundar S, Kundu J, Kundu SC. Biopolymeric nanoparticles. Science and technology of advanced materials. 2010 Feb 1.

15. Jian H, Zhu L, Zhang W, Sun D, Jiang J. Galactomannan (from Gleditsia sinensis Lam.) and xanthan gum matrix tablets for controlled delivery of theophylline: In vitro drug release and swelling behaviour. Carbohydrate Polymers. 2012 Feb 14;87(3):2176-82.

16. Gong H, Liu M, Zhang B, Cui D, Gao C, Ni B, Chen J. Synthesis of oxidized guar gum by dry method and its application in reactive dye printing. International Journal of Biological Macromolecules. 2011 Dec 1;49(5):1083-91.

17. R. Badwaik H, Kumar Giri T, T. Nakhate K, Kashyap P, Krishna Tripathi D. Xanthan gum and its derivatives as a potential bio-polymeric carrier for drug delivery system. Current Drug Delivery. 2013 Oct 1;10(5):587-600.

18. Sharma G, Naushad M, Ala'a H, Kumar A, Khan MR, Kalia S, Bala M, Sharma A. Fabrication and characterization of chitosan-crosslinked-poly (alginic acid) nanohydrogel for adsorptive removal of Cr (VI) metal ion from aqueous medium. International journal of biological macromolecules. 2017 Feb 1;95:484-93

19. Goswami S, Naik S. Natural gums and its pharmaceutical application. Journal of Scientific and Innovative Research. 2014;3(1):112-21.

20. Anwar F, Bhanger MI. Analytical characterization of Moringa oleifera seed oil grown in temperate regions of Pakistan. Journal of Agricultural and food Chemistry. 2003 Oct 22;51(22):6558-63.

21. Panda D, Si S, Swain S, Kanungo SK, Gupta R. Preparation and evaluation of gels from gum of Moringa oleifera. Indian journal of pharmaceutical sciences. 2006 Nov 1;68(6):777.

22. Attri A, Kumar R, Rozera R, Ck S, Vyas M, Wadhwa P, Verma S. A systematic review on grafting techniques and their applications with reference to natural gums and mucilage. Plant Arch. 2019;19:979-82.

23. Athawale VD, Lele V. Graft copolymerization onto starch. II. Grafting of acrylic acid and preparation of it's hydrogels. Carbohydrate Polymers. 1998 Jan 1;35(1-2):21-7.