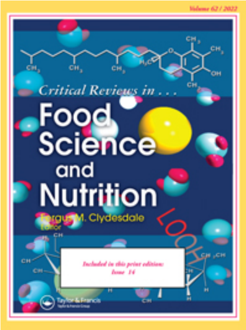

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REVIEW

Gums-based engineered bio-nanostructures for greening the 21st-century biotechnological settings

Muhammad Bilal[a], Hira Munir[b], Muhammad Imran Khan[c], Mohsin Khurshid[d], Tahir Rasheed[e], Komal Rizwan[f], Marcelo Franco[g], and Hafiz M. N. Iqbal[h]

[a]School of Life Science and Food Engineering, Huaiyin Institute of Technology, Huaian, China; [b]Department of Biochemistry and Biotechnology, University of Gujrat, Gujrat, Pakistan; [c]Hefei National Lab for Physical Sciences at the Microscale and the Centers for Biomedical Engineering, University of Science and Technology of China, Hefei, Anhui, China; [d]Department of Microbiology, Government College University Faisalabad, Faisalabad, Pakistan; [e]School of Chemistry & Chemical Engineering, Shanghai Jiao Tong University, Shanghai, China; [f]Department of Chemistry, University of Sahiwal, Sahiwal, Pakistan; [g]Department of Exact and Technological Sciences, State University of Santa Cruz, Ilhéus, Brazil; [h]Tecnologico de Monterrey, School of Engineering and Sciences, Monterrey, Mexico

ABSTRACT
Naturally occurring plant-based gums and their engineered bio-nanostructures have gained an immense essence of excellence in several industrial, biotechnological, and biomedical sectors of the modern world. Gums derived from bio-renewable resources that follow green chemistry principles are considered green macromolecules with unique structural and functional attributes. For instance, gum mostly obtained as exudates are bio-renewable, bio-degradable, bio-compatible, sustainable, overall cost-effective, and nontoxic. Gum exudates also offer tunable attributes that play a crucial role in engineering bio-nanostructures of interest for several bio- and non-bio applications, e.g., food-related items, therapeutic molecules, sustained and controlled delivery cues, bio-sensing constructs, and so on. With particular reference to plant gum exudates, this review focuses on applied perspectives of various gums, i.e., gum Arabic, gum albizzia, gum karaya, gum tragacanth, and gum kondagogu. After a brief introduction with problem statement and opportunities, structural and physicochemical attributes of plant-based natural gums are presented. Following that, considerable stress is given to green synthesis and stabilization of gum-based bio-nanostructures. The final part of the review focuses on the bio- and non-bio related applications of various types of gums polysaccharides-oriented bio-nanostructures.

KEYWORDS
Applications; biological macromolecules; biomedical; biosensor; gum exudates; metallic nanoparticles; nanostructures; tissue regeneration

Introduction

A wide range of nanostructures, i.e., metallic nanoparticles, carbon nanotubes (CNTs), and carbon nanofibers, have been successfully employed in numerous applications, such as biotechnology, biomedicine, energy storage/conversion, environmental management, water remediation, automobiles, pharmaceuticals, and health care system (Padil et al. 2018; Rasheed et al. 2019; Bilal, Nguyen, and Iqbal 2020; Rasheed et al. 2020). Nevertheless, most of the methods currently adopted to fabricate nanoparticles, nanotubes, or nanofibers depend on environmentally unfriendly materials and generate undesirable toxic waste. Green engineering nanotechnology applying sustainable chemistry concepts is a robust approach that facilitates the design and development of industrially compatible bioproducts and processes focusing on the utilization of bio-renewable chemicals, which resultantly reduce or circumvent the consumption and production of baleful substances (Bilal, Zhao, and Iqbal 2020). New green nanotechnology procedures often involved natural resources, nontoxic solvents, biodegradable, and bio-sustainable materials (Pereao et al. 2017; Bilal and Iqbal 2018; Bilal, Rasheed, et al. 2020; Liu et al. 2020; Ain et al. 2020).

Gum polysaccharides are derived from bio-renewable resources. They are safe, cost-effective, ecologically acceptable, and abundantly present in nature (Padil et al. 2018; Nazarzadeh, Makvandi, and Tay 2019). Other advantageous features of plant-based gum polysaccharides are shown in Figure 1. Natural gums are water-soluble polymers that can increase the viscosity of aqueous solutions, even in minor quantities. They are widely used as thickeners and emulsifiers to control the texture and stability of the final product (Nazarzadeh, Makvandi, and Tay 2019). Gums and gums-derived bio-composites find diverse applications in the biomedical research domains compared to their synthetic counterparts (Seeli and Prabaharan 2017; Padil et al. 2018). In the recent decade, there has been incredible research progress in the fabrication and engineering of gum-based bio-nano composites. The current work spotlight various gums, i.e., gum Arabic, gum albizzia, gum karaya, gum tragacanth, gum kondagogu, and others (Figure 2), as candidates to construct nanocubes with therapeutic and functional values to confront the current challenges and drawbacks of in-practice counterparts.

CONTACT Muhammad Bilal bilaluaf@hotmail.com School of Life Science and Food Engineering, Huaiyin Institute of Technology, Huaian 223003, China; Hafiz M. N. Iqbal hafiz.iqbal@tec.mx Tecnologico de Monterrey, School of Engineering and Sciences, Monterrey, 64849, Mexico.
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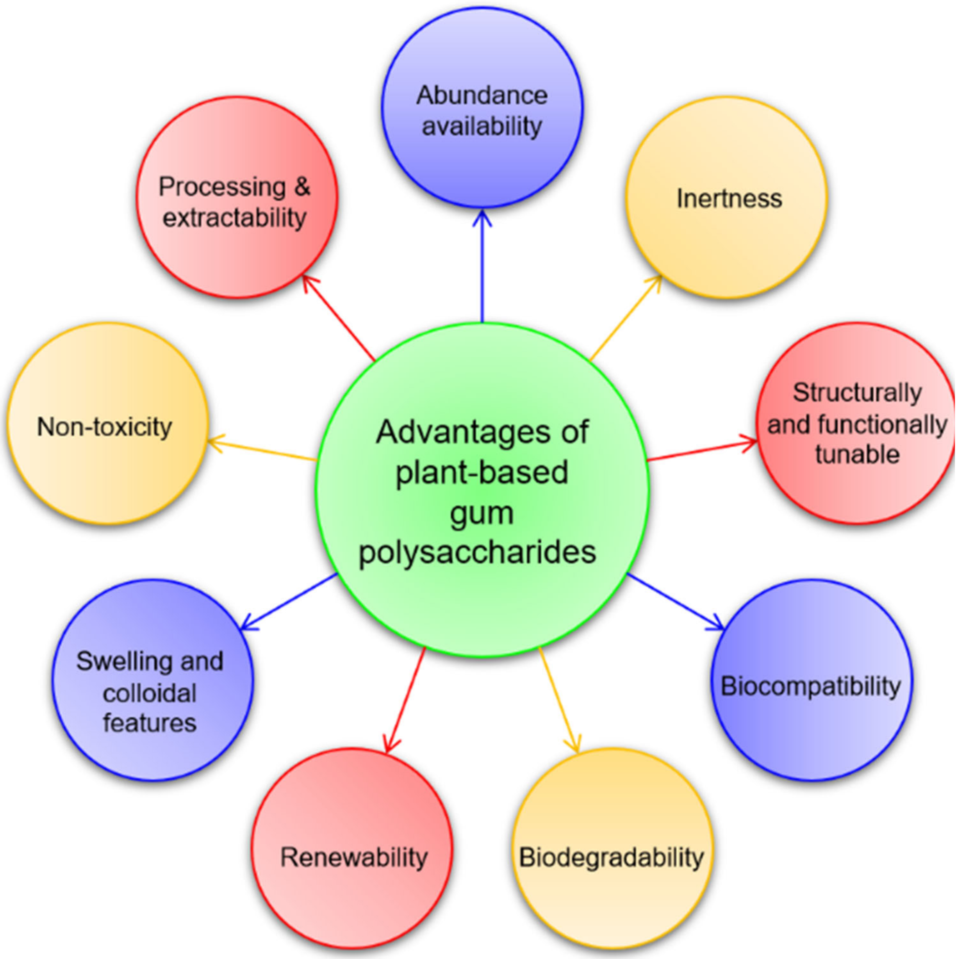

Figure 1. Advantageous features of plant-based gum polysaccharides.

Natural gums: structural and physicochemical properties

The widespread use of natural gum polysaccharides in multiple fields results in novel fabrication and development by modifying mechanical, structural, and biochemical properties. Unique biological and chemical characteristics of natural gums polysaccharides have been demonstrated to be highly desired in medicine, nutrition, agriculture, pharmacology, chemical and environmental engineering (Halake et al. 2016; Bilal and Iqbal 2019a, 2019b; Kausar et al. 2020; Zhang et al. 2021). Scientists are frequently endeavoring to use various gums to develop medical systems like drug delivery, wound curative bandages, and biomimetic tissue engineering scaffolds (de Oliveira Barud et al. 2016). Natural polysaccharides, such as xanthan gum, gum tragacanth, pectin, dextran, carrageenan, guar gum, etc., have been employed for medicinal purposes in the form of nanoparticles, nanofilms, and nanofibers (Azizi et al.2017).

Gum tragacanth is a complex, anionic, acidic, and high molecular weight biopolymer that finds its uses in various areas due to ease of availability, nontoxicity, biodegradability, and increased durability, and stabilization against heat, acid, and microbial attacks. It is explicitly non-carcinogenic, non-allergenic, and exhibits no harmful activities (Buckley and O'Kelly 2004). In pharmacy, this heterogeneous and branched macrostructure is also utilized as a binder, suspending, and gelling agent in formulating pills and tablets. Moreover, it has shown the ability to prevent wound healing, cancer cell growth, and many other therapeutic applications (Fayazzadeh et al. 2014). The anticancerous potentialities of engineered cues derived from plant gum polysaccharides to kill malignant cells and impede metastasis by acting on tumor cells microenvironment through various mechanisms are shown in Figure 3. Gum tragacanth is extracted from the branches and stems of several *Astragalus* species including *A. gummifer*, *A. adscendens*, *A. tragacantha*, and *A. brachycalyx*. Characteristic attributes such as ionic nature, hydrophobicity, and the presence of biodegradable and pH-responsive moieties render gum tragacanth a promising candidate for the development of tissue engineering scaffolds and hydrogel-release control systems. Xanthan gum is a high molecular weight anionic polysaccharide that constitutes a viscous solution at low shear-rates. Xanthan gum secreted by a Gram-negative bacterium, Xanthomonas bacteria exhibits stability in a broader range of pH values (Rodríguez-Hernández and Tecante 1999). Its side chains consist of alternative residues of D-glucuronic acid and D-mannose in a ratio of 1:2 (Douglas et al. 2018). Gum polymer has been employed in various biomedical fields, including tissue regeneration, targeted release of drugs separately, or integration with other synthetic or natural polymers (Popa et al. 2010).

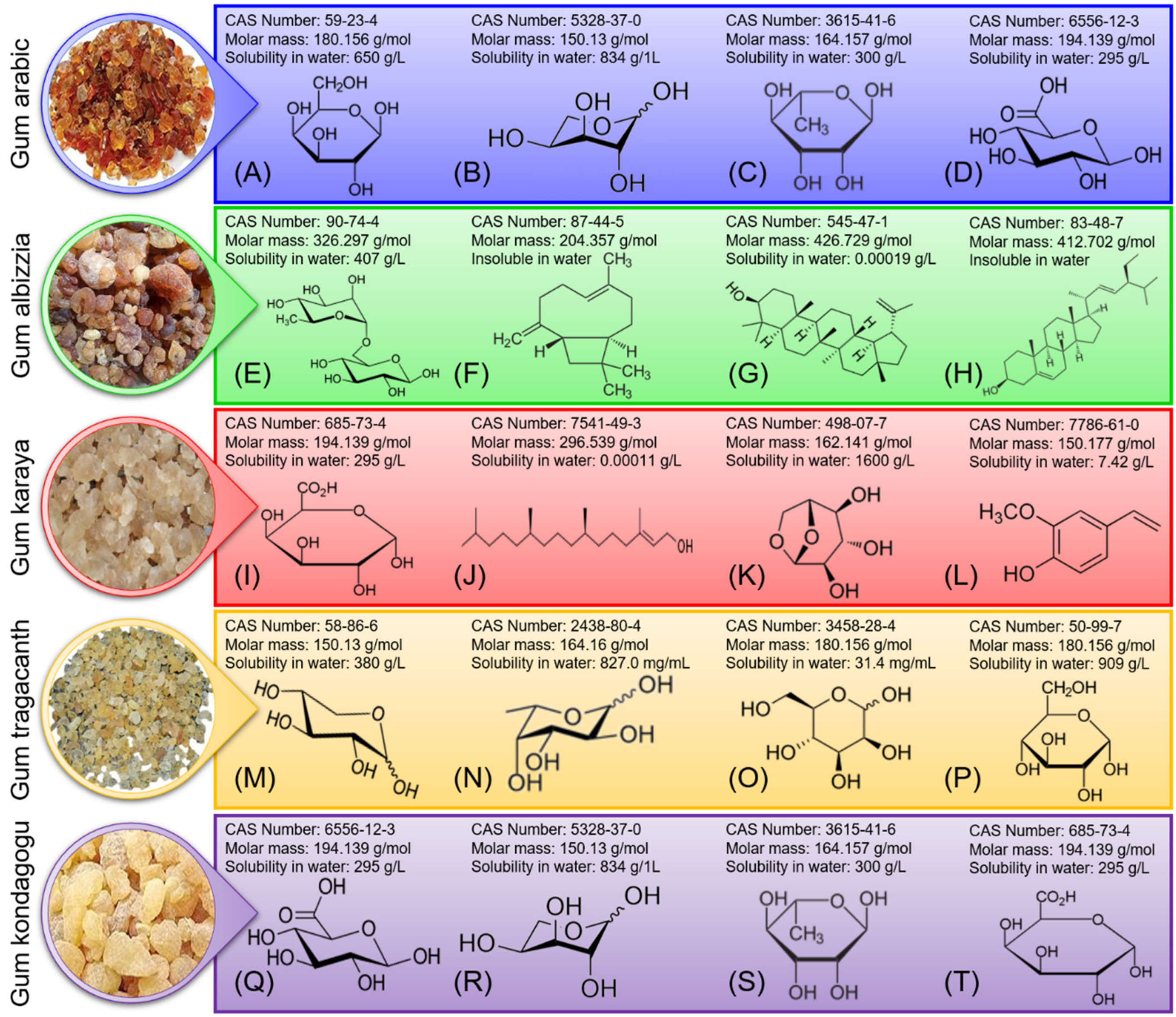

Figure 2. Morphological, structural and functional attributes of various plant-based gums, i.e., gum Arabic, gum albizzia, gum karaya, gum tragacanth, gum kondagogu. (A) D-galactose, (B) L-arabinose, (C) L-rhamnose, (D) D-glucuronic acid, (E) rutinose, (F) caryophyllene, (G) lupeol, (H) stigmasterol, (I) D-galacturonic acid, (J) phytol, (K) levoglucosan, (L) 2-methoxy-4-vinyl phenol, (M) D-xylose, (N) L-fucose, (O) D-mannopyranose (D-mannose), (P) α-D-glucopyranose (Q) D-glucuronic acid, (R) L-arabinose, (S) L-rhamnose, (T) D-galacturonic acid.

Dextran is a biocompatible, nontoxic, and hydrophilic homopolysaccharide that can be obtained bacterially from sucrose using dextransucrase (Sun and Mao 2012). It has been widely used in pharmaceutical and theranostic applications to ensure skin rehabilitation, reduce inflammatory responses, and promote wound curing due to biocompatibility and biodegradability attributes (Sun and Mao 2012). Impressing alginate features, an anionic biopolymer of mannuronic acid and glucuronic acid units (Wróblewska-Krepsztul et al. 2019), have made it an alluring candidate for extensive tissue engineering and nanomedicine applications. Chitosan is a linear polysaccharide obtained from the deacetylated chitin that has revealed significant potentiality in targeted drug delivery, nanomedicine, and tissue engineering (Sultankulov et al. 2019). Hyaluronic acid is a non-sulfated glycosaminoglycan that is widely distributed in epithelial, connective, and neural tissues. Due to several fascinating properties, such as the availability of numerous carboxylic acid and hydroxyl groups, hyaluronan is considered a robust polymer for tissue engineering and drug delivery (Zhang, Zheng, et al. 2017; Kim et al. 2019). Carrageenans isolated from red edible seaweeds are linear sulfated polysaccharides that can be utilized in several industrial compounds (Yegappan et al. 2018). It is currently considered a preferred modality in regenerative medicine and tissue engineering due to desired physicochemical properties such as high-water absorption capability, gelation mechanism, and the presence of a vast number of functional groups (Yegappan et al. 2019). The potential applicability of guar gum is attributed to its cytocompatibility, rheological and biodegradability features. It is a nonionic, water-soluble, and high-molecular-weight plant polysaccharide, mainly isolated from the seeds of *Cyampsis tetragonolobus* (Torres, Hallmark, and Wilson 2014).

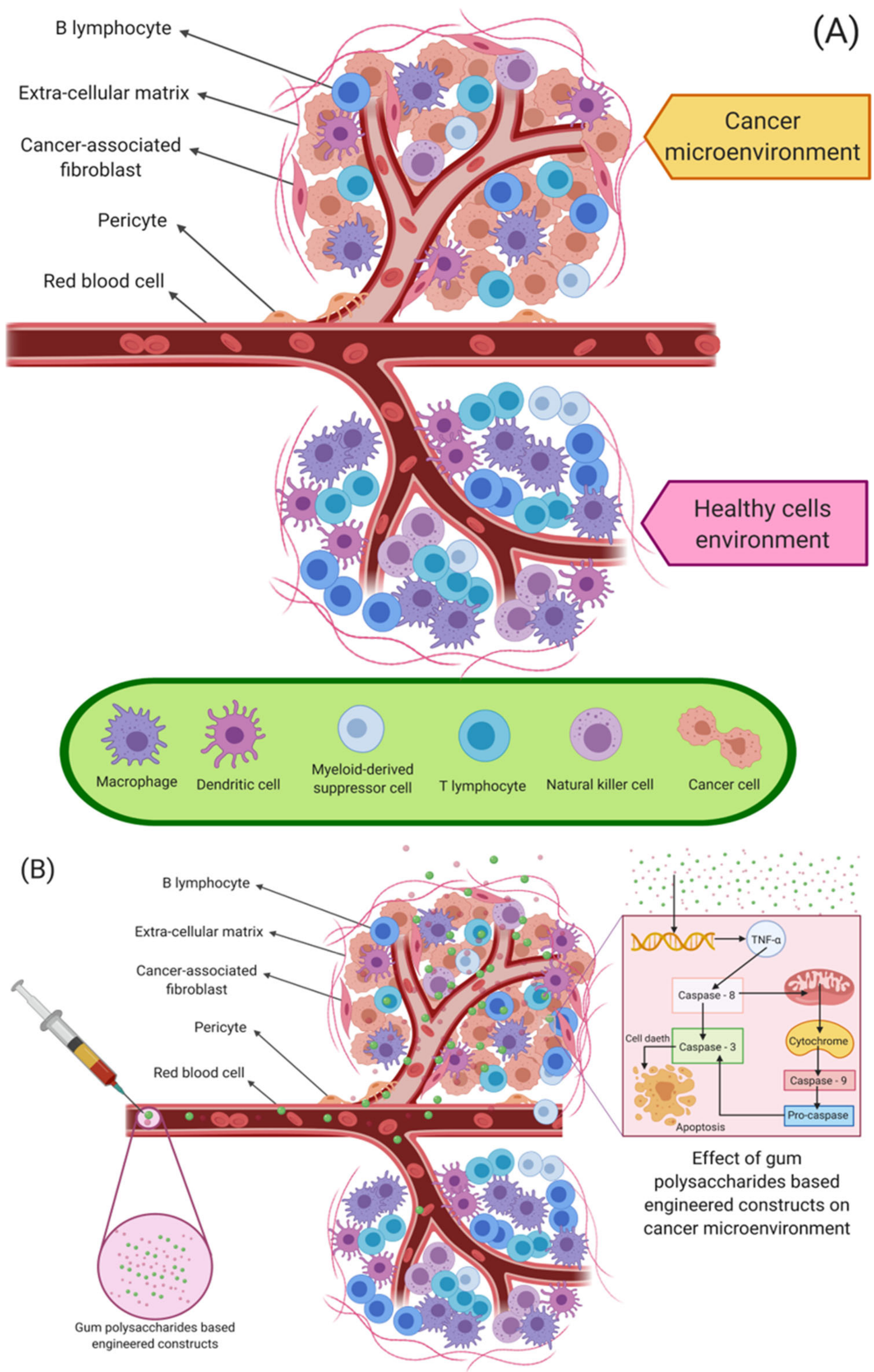

Figure 3. Various plant-based gums and their exploitation to engineer bio-nanostructures.

Gums-based bio-nanostructures

Nanotechnology implicates the manipulation, construction, and utilization of nanoscale materials with controlled shape and composition. Figure 4 illustrates various plant-based gums and their exploitation to engineer bio-nanostructures. A nanostructure is a structure of intermediate size between microscopic and molecular levels. Nanostructures are very tiny material configurations exhibiting size within the range of 1–100 nanometers. Nanotechnology deals with creating, designing, and engineering materials whose basic measure unit is in nanometers. This term also includes pharmaceutical and biomedical sciences, chemistry, advanced nanomaterials, and physics. These materials possess enormous applications as drug delivery nanocarriers, imaging agents, and radiosensitizers in radiation and photodynamic therapy (Hamblin et al. 2015; Brun and Sicard-Roselli 2016).

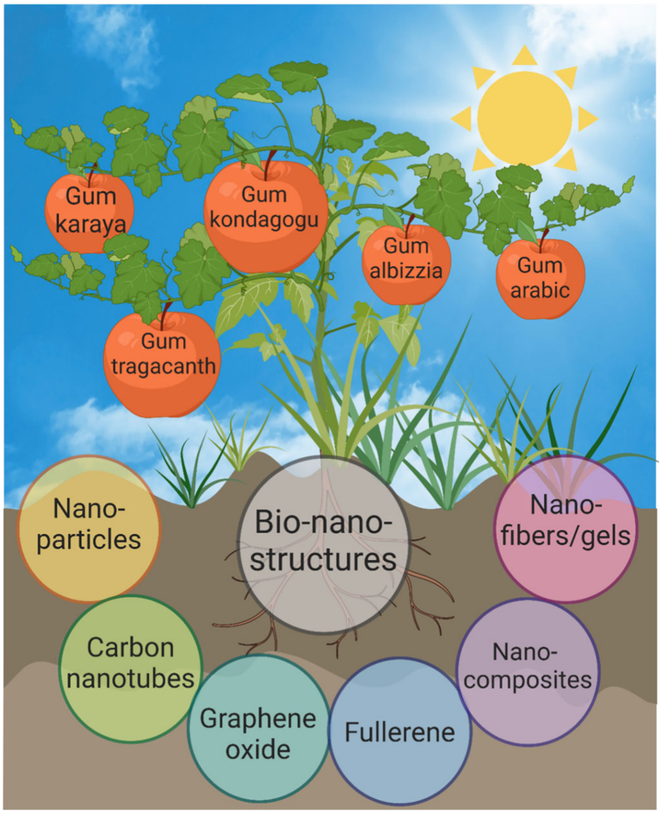

Figure 4. (A) comparative illustration of healthy cells environment vs cancer microenvironment. (B) Anticancer potentialities of plant-derived gum polysaccharides based engineered cues to destroy cancer cells and prevent metastasis by acting on tumor cells microenvironment via different mechanisms such as cell death via apoptosis.

Gums-based metallic nanoparticles

In recent years, much research attention has been diverted to the development and analysis of nanoparticles due to their wide-spectrum biomedical potentialities. Metal nanoparticles are commonly known as nanosized metals with a diameter range of 1–100 nm. These nanostructured particles exhibit improved biological, physical, and chemical properties that make them useful for several applications like thermal, optical, medicinal, and chemical sensing. A wide range of metallic nanoparticles has been explored and engineered to use against different microbial infections because of their great therapeutic and diagnostic potentialities. For instance, gold nanoparticles (AuNPs) have been utilized to quantify and identify bacterial strains in the human joint fluid within a short period of 1 h (Chang et al. 2015). Fabrication of silver nanoparticles (AgNPs) has shown potent and broad-spectrum biological activity against wounds causing microbes (Wu et al. 2014). Metallic nanoparticles offer numerous advantages due to their larger ratio of surface area to volume. Magnetic nanoparticles (MNPs) possess the potential of single-domain magnetism and can be used in Ferrofluid, magnetic tapes, and refrigerants due to their ultra-fine size dimension with magnetic domain size. It is demonstrated that living creatures also possess MNPs in their bodies. For instance, fish and migratory birds exhibit MNPs in the sensing system that direct their migration between north and south each year. Similarly, human beings also contain approximately 100 million MNPs per gram of tissue in their brains (Gieré 2016).

Different methods, such as electrochemical, chemical, radiolytic, and photochemical techniques, can be used to synthesize metallic nanoparticles. Various studies have suggested that the synthesis of MNPs is due to the reduction of metallic precursors like Cu, Au, Ag, and Zn. The stability of as-prepared nanoparticles can be increased by using other chemicals. Different MNPs depend on their chemical properties like metals, magnetic alloys, earth metals, and the oxidation of metallic nanoparticles (Dahman 2017). Notably, a variety of nanoparticles-based therapeutic products has been clinically approved. Among these, liposomal (AmBisome, Abelcet, Myocet, DaunoXome, and Caelyx) and polymeric-based products (Adagen, Neulasta, Macugen, Ranagel, and Copaxone) are most commonly used (Zhang et al. 2008). Recently, synthesis methods based on green chemistry principles have become important and popular to circumvent the use of environmentally-unsafe products. For this purpose, different reducing substances from green sources are being used, such as citrus fruits, coffee seeds, lactic acid, etc. (Bogireddy et al. 2018). More complex and elegant methods for the biosynthesis of these nanostructures involve microwave-assisted, sonochemical, and electrochemical methods (Roldan, Pellegri, and de Sanctis 2013; Sahoo et al. 2015). Metal nanoparticles such as copper, zinc, gold, and silver have been broadly investigated for various biomedical applications.

Gums-based silver nanoparticles

AgNPs possess different novel properties depending on shape, morphology, and size that enable their applicability to deal with animals, microbes, and plants. AgNPs have shown outstanding bactericidal properties against numerous microorganisms (Siddiqi and Husen 2016). Silver nanostructures have gained a significant position due to the essential characteristics, eco-friendliness, and broad-spectrum applications in catalysis, electronics, drugs, antimicrobial agent, and the development of biological systems (Lokina et al. 2014). It is predicted that around 320 tons of AgNPs are synthesized every year and are being used in food products, biosensing, nanomedical imaging, and theranostic applications. Moreover, they are also being used as topical creams to prevent wound infections, anticancer agents, and antimicrobial products in wound dressing (Jones et al. 2004).

Silver is a white, soft, and shiny transition metal with high electrical and thermal conductivity. It is used in many different forms like vessels, coins, sutures, foils, solutions, and colloids as ointments or lotions. Silver is the prime therapeutic agent used in medicines to treat surgical infections and infectious diseases (Alexander 2009). Ag salts are being utilized for the inhibition of different bacterial growth in the human system. Silver is also used in cuts, catheters, wounds, and burns to prevent infections. In a study, Das, Gang, and Nath (2011) demonstrated that AgNPs are significant bacterial growth inhibitors. Biogenic synthesis of AgNPs involves fungi, bacteria, actinomycetes, plant extracts, and yeast. The morphology, stability, and size of AgNPs are based on the synthesis, concentration, temperature, nature of the solvent, and strength of reducing agents used (Siddiqi and Husen 2016). Eghbalifam et al. (2020) fabricated gum arabic-based antimicrobial nanofibers mat with appropriate porous structure, mechanical strength, water vapor permeability, and high-water absorption capacity, loaded with AgNPs and applied against *E. coli*, *S. aureus*, *C. albicans*, *and P. aeruginosa*. A green and simple approach was used employed for the biosynthesis of gum acacia-assisted AgNPs as highly selective sensing for Hg(II) and a triarylmethane dye malachite green. The redox reaction between Hg(II) and Ag(0) results in the formation of Hg(0) and Ag(I) and transference of electron from gum acacia to Ag(I), leading to form an Ag@Hg nanoalloy. The as-developed new nanosensor showed the effective sensing and detection ability to malachite green via an inner filter effect (Abbasi, Hanif, and Shakir 2020). Sharma et al. (2019) fabricated gum Salai guggal-mediated silver nanocomposites, which presented potential antipathogenic bioactivity against four different bacterial isolates, including *E. coli*, *B. cereus*, *P. aeruginosa*, and *S. aureus* bacterial isolates representing their biomedical potentiality to prevent bacterial infections.

Gums-based copper nanoparticles

Copper nanoparticles (CuNPs) have garnered significant attention because of their physicochemical properties like magnetism, small dimensions, and sensors (Huang et al. 2009), catalyst (Kantam et al. 2007), high melting point, antimicrobial, thermal and electrical conductivity, heat transfer and light absorption. CuNPs are being employed as antimicrobial agents as they easily interact with other compounds by enhancing their antimicrobial potential because of their high surface to volume ratio. Besides, CuNPs have been applied as a heterogeneous catalyst for various environmental processes like decomposition of NO_2, oxidation of CO, and nitric oxide reduction. Copper oxide is formed from CuNPs when exposed to air. To avoid this oxidation, the nanoparticles are covered with inorganic and organic coatings like carbon and silica. The resistance of functionalized CuNPs against oxidation was higher as compared to bare copper. Studies related to the use of CuNPs have recently expanded into many fields, such as in membranes for water purification (Ursino et al. 2018), electronics (Roy, Foong, and Cullinan 2018), agriculture (Hernández et al., 2018; López-Vargas et al. 2018), and catalysis.

Numerous synthetic methods have been designed for the synthesis of CuNPs, e.g., sonochemical and thermal reduction, electron beam irradiation (Zhou et al. 2008), metal vapor synthesis (Ponce and Klabunde 2005), reverse micelles, polyol process (Park et al. 2007), microwave irradiation (Zhu, Zhang, and Yin 2004), chemical reduction and pulsed laser ablation (Tilaki, Iraji Zad, and Mahdavi 2007). Some of these methods have few factors that hinder their use in the fabrication of CuNPs. Among the above-stated techniques, chemical reduction is the most common and appropriate method. It possesses several advantages over the remaining methods such as versatility, inexpensive, the ability of solubilization, and the obtained nanoparticles containing well-defined sizes and shapes. Due to the chances of oxidation of Cu with air, the synthesis of CuNPs is more laborious and tiring than other metals. High surface to volume ratio, small size, and improvement in shape and size are factors that CuNPs have gained more attention than many other metals like Au, Pt, and Ag. In recent times, natural gum polymers are increasingly used to reduce and stabilize agents to synthesize CuNPs, which prevent the aggregation of the nanoparticles. Padil and Černík (2013) designed and developed CuO nanoparticles from a naturally occurring and plant-derived nontoxic gum karaya that served as a templating and a reducing agent. The synthesized nanoconstructs were 4.8 nm in diameter, incredibly stable, and exhibited promising antibacterial activities against *Staphylococcus aureus* and *Escherichia coli* strains. Recently, CuNPs were fabricated by ascorbic acid and sodium borohydride-driven reduction reaction in the presence of a natural biopolymer stabilizer cashew gum (CG). The CG-CuNPs were very tiny spherical particles with an average size of 10 nm and showed high bioactivity against Staphylococcus aureus without any toxic effects on BALB/c murine macrophages (Amorim et al. 2019). Dong et al. (2014) put forward a facile approach to fabricate colloidal CuNPs in aqueous media using gum acacia, copper sulfate, and hydrazine as capping, copper precursor, and reducing agents respectively. The prepared particles were well-dispersed, nanosized, and spherical with a size of 3 to 9 nm that depends on the amount of gum acacia, reaction temperature, and duration.

Gums-based gold nanoparticles

AuNPs are frequently used nanomaterials due to their electric, optical, photonic, textile, and antimicrobial properties. These nanostructures have fascinated a considerable deal of interest in biotechnology because of their spectacular properties like multiple surface functionalities, catalytic potential, surface chemical activity, excellent chemical stability, and environmentally-friendly nature (Choudhary et al. 2017; Ran et al. 2018). Bioconjugates of AuNPs have become a promising agent in designing novel biomaterials for the exploration of biological systems. There are few chemical and physical methods for synthesizing AuNPs, e.g., chemical reduction, ion sputtering, sol-gel, and hydrothermal (Yazid et al. 2010; Vieaud et al. 2018). The versatile nature of AuNPs has offered many materials for a wide range of applications in the biomedical field. In diagnostics, the binding between AuNPs and analyte leads to the detection of signals (Uehara 2010).

Plants-derived natural gums are readily available, economical, and renewable materials and have found wide applications in the bio-fabrication of AuNPs. Gangapuram et al. (2017) prepared AuNPs-mediated colorimetric modalities for highly selective and sensitive sensing copper (II) ions using plant-originated nontoxic polysaccharide carboxymethyl gum karaya as a capping, reducing, and modifying agent. The gums-stabilized AuNPs presented a discriminating response to copper ions and showed an excellent linear correlation and minimum limit of detection in real

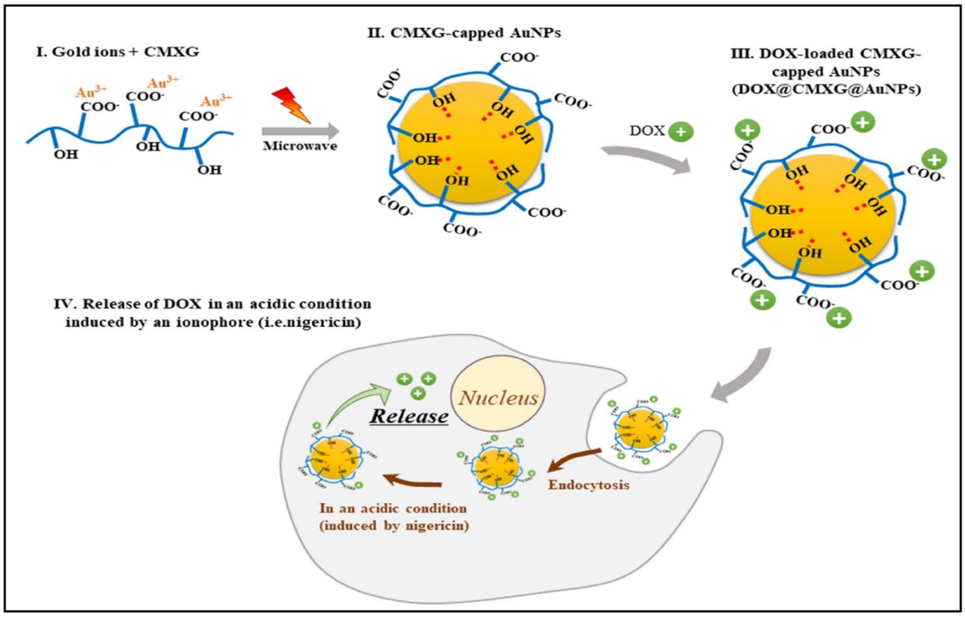

Figure 5. Schematic illustration showing the preparation of CMXG-capped AuNPs, subsequent loading of DOX onto CMXG@AuNPs, and the in vitro release of DOX from DOX@ CMXG@AuNPs under acidic conditions (induced by ionophore, i.e., Nigericin). Reprinted from Alle et al. (2020) with permission from Elsevier. © 2019 Elsevier Ltd. License Number: 4977821285443.

biological samples. Alle et al. (2020) developed carboxymethyl xanthan gum-imprinted AuNPs adopting microwave irradiation strategy (Figure 5). The newly-synthesized nanoparticles possessed high crystallinity and spherical shape with a size range of 8 to10 nm. Results revealed an increased release of doxorubicin encapsulated on CMXG@AuNPs in acidic conditions but insignificant at physiological pH. In another study, AuNPs were prepared using carboxymethylated gum kondagogu as a reducing and stabilizing agent. The reducing capability of as-obtained AuNPs (11 nm) was evaluated to reduce 4-nitrophenol pollutants in the presence of $NaBH_4$. The gum-based AuNPs presented marked antibacterial activities against *Bacillus subtilis* and *E. coli* (Seku et al. 2019).

Gums-based zinc-oxide nanoparticles

Zinc oxide is usually considered a safe compound that exhibits photo-oxidizing and photocatalysis effects on biological and chemical species (Espitia et al. 2012). Zinc oxide nanoparticles (ZnONPs) have received consideration due to their wide varieties of antimicrobial, luminescence, anti-corrosive, photocatalytic, thermal, UV filtering, chemical stability, chemical and physical properties (Hatamie et al. 2015; Sharmila et al. 2018). These nanoparticles inhibit prokaryotic growth by cytotoxicity and induce cells' death through oxidative bursting and photoactivation in eukaryotes. Moreover, they can be used in biomedical engineering for implant coatings, tissue regeneration, wound healing, bio-imaging, and the development of cancer therapies (Iqbal et al. 2019; Mirzaei and Darroudi 2017; Ullah, Zainol, and Idrus 2017).

Synthesis of ZnONPs by chemical methods such as laser ablation, ultrasonic fields, and lithography requires high temperature and pressure and generates toxic products. Thus, the synthesis from green sources like plant extracts is acquiring more attention and being widely used nowadays. It provides simple and cost-effective methods for the synthesis of metallic nanoparticles. Different methods for the synthesis from bacteria (Yusof, Mohamad, and Zaidan 2019), and fungi (Sanghi, Verma, and Puri 2011) are being used in many types of research. Plant extracts for the synthesis of ZnONPs can be more beneficial than chemical and microbial methods as there is no chance of hazardous chemicals and bacterial contamination (Rai and Ingle 2012).

Notably, plant gums play a pivotal role in the fabrication of various metallic nanoparticles. Suganya and Vivekanandhan (2019) reported the biosynthesis of ZnONPs following an Azadirachta indica (neem) gum-mediated sol-gel fabrication process. Neem gum acts as a promising chelating candidate for Zn^{2+} ions, leading to the identical dissemination of metal ions in the gum network. The prepared ZnONPs revealed exceptional photocatalytic activity for the degradation of organic dye trypan blue under UV exposure. Ullah, Munir, and Shahid (2019) used crude and carboxymethylated *Bombax malabaricum* gum for the eco-friendly biosynthesis of zinc and AgNPs to explore their potential as a bio-binder for the preparation of drug tablets. The drug tablets formulated from crude and purified gums exhibited slow dissolution and release of drugs compared to the tablets formed using functionalized gum-based nanoparticles.

Gums-supported carbon nanomaterials

In technology science, carbon-based nanostructures are becoming important candidates due to their inimitable chemical and physical features (Tiwari et al. 2016; Zhang, Zheng, et al. 2017; Mukhopadhyay et al. 2016). Due to the presence of various allotropes of carbon such as diamonds, graphite, CNTs, fullerene, graphene, carbon nanostructures have recently gained increasing importance (Mostofizadeh et al. 2011). Carbon is one of the most versatile and abundant elements in the universe (Siqueira and Oliveira 2017).

Carbon-based materials have shown great versatilities because they can chemically combine/react with other carbon-based materials and different elements by forming strong covalent bonds. As a result, these materials showed excellent characteristics like high density, high hardness, and high strength. Depending on its allotropic factors, carbon can form different compounds that exhibit various features depending on the arrangement of carbon atoms adjacent to each other. Graphite and diamond are two common examples. Graphite is brittle and fragile, whereas diamond possesses the highest hardness (Koziol, Boskovic, and Yahya 2010).

The development of multiwall CNTs (MWCNTs) from carbon arc discharge was first realized by Sumio Iijima in 1991. After a few years, Doanld Bethune and Prof. Sumio Iijima independently observed single-wall CNTs (SWCNTs) (Monthioux and Kuznetsov 2006). Later on, research about CNTs expanded quickly. These nanostructures were explained as hollow cylinders that consisted of graphene sheets and were further divided into SWCNTs and MWCNTs. SWCNTs are cylindrical nanostructures prepared by rolling up a single sheet of graphene with a high aspect ratio. While MWCNTs consist of few graphene sheets in the rolling pattern with 3.4A° of interlayer spacing (Eatemadi et al. 2014). Due to their unique electrical, structural and mechanical diversity, CNTs offer high strength, electrical conductivity, and flexibility toward different biological entities that are potentially useful for medical diagnosis, sensing, and the treatment of various infectious diseases (Roldo and Fatouros 2013). Among the different allotropes of carbon, graphene is known as the most attractive allotrope due to its exclusive intrinsic properties. Around 70 years ago, Wallace in 1947 determined the electronic structure of graphene, and in 1956, McClure explored the corresponding wave equation (Sun et al. 2013). Herein, we highlight and discuss graphene, CNTs, and fullerenes among the many carbon structures.

Gums–graphene oxide framed nanostructures

Graphene nanomaterial is arranged in a two-dimensional layer of carbon atoms, connected in the hexagonal lattice structure through sp^2 hybridization. In 2001, Konstantin Novoselov and Andre Geim from the University of Manchester discovered graphene's structure (Novoselov et al. 2004). Graphene can be used in batteries, sensors, supercapacitors, and fuel and solar cells. Due to the enormous significance in different fields, graphene's discoverers were awarded the Nobel Prize in 2010. Because of the crystalline structure, graphene has a significant band structure. Graphene forms a two-dimensional structure resulting in a hexagonal lattice in which the carbon atoms are 1.42 A° form their three neighboring atoms through a σ bond with each other (Geim 2009; Novoselov et al. 2012).

Currently, graphene oxide (GO) is regarded as an ideal material in the field of carbon-based nanostructures. Though GO is analogous to graphene, the plane of graphene hexagons is garnished by oxygen that render GO solvable in an aqueous solution. Many other polar solvents new functionalities to this material. These exceptional physical and chemical features furnish GO, a highly appealing candidate in drug delivery, energy, catalysis, etc. (Adeel et al. 2018; Sehar et al. 2020). The utilization of natural plants derived gum polymers has achieved popularity in synthesizing GO-based novel nanostructured for various technological applications. For example, Oliveira et al. (2020) integrated GO and gum Arabic to design an electrode that was used as a potentiometric pH sensor. Xanthan-incorporated titanium dioxide functionalized GO was prepared by an eco-friendly, facile, and cost-efficient ice-templating method. The as-prepared 3D-graphene nanocomposite showed porous sponge-like architecture, chemical functionalities, and high thermal steadiness. It also demonstrated potential adsorption capabilities in a highly recyclable way under milder conditions (Lai et al. 2020). A novel environmentally-friendly method was adopted to synthesize nanofibers based on an amalgamation of polyvinyl alcohol and gum arabic exudate. In the presence of GO, the fabricated nanofibrous membrane showed 50% higher adsorption capacity for methylene blue, which offers the opportunity for the effective utilization of GO/GA/PVA nanofibers for environmental remediation (Silvestri et al. 2019). Venkateshaiah et al. (2019) generated reduced GO-supported platinum nanoparticles using gum waste of a nontoxic gum kondagogu using an eco-benign synthesis approach. The as-synthesized RGO-Pt nanostructures exhibited superior catalytic performance for the hydrogenation of 4-nitrophenol pollutants.

Gums–CNT composites

In the past few years, CNTs are the most studied nanostructures (Li, Wu, and Chopra 2015). They form a one-dimensional structure and have a diameter of few nanometers. CNTs possess unique electrical and mechanical characteristics that make them different from other allotropes of carbon. Their discovery has not been fully explained, and CNTs are known since 1991 in the scientific community after the publication of Sumio Iijima that described a method for forming concentric and cylindrical carbon structures, which were later known as MWNTS (Iijima 1991). Donald Bethune and Iijima, in 1993, published a new study that covers the methods for the formation of SWNTS. After this discovery, nanotubes became the main object for research in different fields of science (Iijima and Ichihashi 1993). In the aeronautics industry, CNTs are being used to produce new composites (Gohardani, Elola, and Elizetxea 2014). They also find utilization in electronic nanocircuits and nanodevices to manufacture new chips for computers (Maiti et al. 2014). Ferreira et al. (2015) carried out the functionalization of boron nitride nanotubes (BNNTs) with gum arabic, which is a naturally occurring polysaccharide with exceptional biocompatibility and good water solubility. The gum-modified BNNT were found to be attractive candidates for the specific delivery of drugs, proteins, and nucleic acids. Makhado et al. (2018) fabricated, characterized, and surface-modified xanthan gum-cl-poly (acrylic acid)/o-MWCNTs hydrogel nanoscale composites by microwave-assisted co-polymerization strategy using N, N′-methylenebisacrylamide as a cross-linker. The newly synthesized XG-cl-

PAA/o-MWCNTs nanocomposites demonstrated an efficient and reusable adsorptive capacity for methylene blue dye.

Fullerene

Fullerene is known as the third allotropic type of carbon, after diamond and graphite (Rocha and Filho 1996; Yadav and Kumar 2008). In 1985, Kroto et al. discovered that fullerene possesses an icosahedral and closed cage structure that is composed of 12 pentagons and 20 hexagons in which every carbon atom bonds to the 3 other carbon through sp^2 hybridization. Due to similarity with a building designed by an American architect Richard Buckminster Fuller, fullerene is also known as *buckminsterfullerene* (Kroto et al. 1985). Fullerene contains 60 carbon atoms arranged in the shape of a soccer ball having a 7 A˚ diameter. Because of its shape, it is also called Buckyball. Fullerene is a zero-dimensional carbon structure, as it forms molecular crystals. The discovery of fullerene has revolutionized research for new allotropes of carbon and different nanostructured materials. In 1996, its discoverers were awarded the Nobel Prize in the field of Chemistry (Kroto et al. 1985; Siqueira and Oliveira 2017).

Applications of natural gums

Carbohydrate polymers are preferred to synthetic materials for their low cost, nontoxicity, and relative abundant availability. Gums exhibit abandoned rates of hydration and thickening decreases in microbial contamination, and viscosity on storage, which can be addressed by manipulating the physical/chemical attributes via the modification process. The following are the applications of natural gums in different fields.

Natural gums: confectionery and salad dressings

Natural gums have good flavor-release properties and a creamy mouthfeel. These gums as thickeners and stabilizers are used in the salads formulation to preserve its stability and eliminate phase separation. Gum Arabic as emulsifiers and gum xanthan as a stabilizer can be applied for the emulsification and stabilization of the olive oil lemon juice in a salad recipe. Salads containing Arabic gum and xanthan were less stable and more stable in terms of creaming and oil droplet coalescence, respectively (Paraskevopoulou, Boskou, and Kiosseoglou 2005). Lipid oxidation is the major reason for spoiling quality in salad dressings. Gum polysaccharides used in salad dressings also affect lipid oxidation. Emulsions of natural gums were stored for 6 to 8 months at room temperature. The natural gums have shown a potential ability to reduce the lipid oxidation rate, presumably because of their ability to increase viscosity and amphiphilic features (Paraskevopoulou, Boskou, and Paraskevopoulou 2007).

Natural gums: frozen products

In baked products, flour is an essential component. The shelf life of different bakery and baked goods can be increased by freezing the dough. Using gum arabic and locust bean gum, the bakery products show the improved character of the gluten network when evaluated after different storage periods. Therefore, frozen dough's shelf life can be improved during the extended periods of frozen storage (Sharadanant and Khan 2006). Natural gums are added to give a smooth texture and protection during the storage of frozen desserts hydrocolloids. High-pressure freezing techniques are also used to enhance the quality of the product (Fernández et al. 2007).

Natural gums: emulsifying and suspending agents

Gums also act as suspending and emulsifying agents and stabilize water-oil emulsions by creating a robust multimolecular film surrounding each oil globule. Pharmaceutical suspensions are solid dispersions of sparingly soluble or insoluble drugs in an aqueous/oily vehicle. They are used for topical application, oral and parental administration of drugs. Since the infants and elders face difficulty swallowing tablets or capsules, the formation of drugs as a suspension for oral administration is a convenient way to give sparingly soluble or insoluble drugs. Drugs as suspensions for oral administration are also useful to control the absorption rate and to mask the taste of drugs. Film coating of pharmaceutical dosage forms has evolved from its basic origin to become a sophisticated science that has contributed immensely to drug delivery advancement (Miller et al. 2008). In tablet formulations, gum albizia and karaya have been successfully used as binders in tablets' formation (Odeku et al. 2005). They serve as a drug delivery carrier to the colon (Ofori-Kwakye, Kwapong, and Adu 2010). Gum Grewia's role as a film coating agent in praziquantel tablets has been investigated (Sharma and Patni 2012). Gums and mucilages of different plants have been utilized as gelling agents due to low-cost, nontoxicity, abundant availability, nonirritating, and emollient nature (Prajapati et al. 2013).

Some naturally occurring polymers such as dextran (Ravi et al. 2009) and okra mucilage have been used in medicine as plasma expanders. A highly pure gum from *Abelmoschus esculentus* has been evaluated as a plasma expander, intravenous circulating agent, and emulsifying agent (Ravi et al. 2009). Gum Arabic is usually added to wine as a natural stabilizer and used to stabilize color (Nussonovitch 2010). Some gums (apple pectin and gum arabic) and blends of some gums with sugar solutions are applied as edible glues and adhesives in the food industries. They can also be utilized to enhance thickening and viscosity. In meat products, natural gums have several uses, such as the supplementation of gum karaya (∼0.25%) provides sufficient water-holding capability and gives products with a much smoother appearance (Nussonovitch 2010). Gums serve as a structural material to the plants, as adhesives, energy reserves, information-transfer agents, and defensive materials (Palaniraj and Jayaraman 2011). Gums are an essential constituent in liquefied foods for controlling viscosity and mouthfeel. For several uses, such as coating or binding agents for water-based paints on paper, gums have been used since ancient times, such as in textiles, cosmetics, paints, and paper manufacturing industries.

Medical applications of gums-based bio-nanostructures

Therapeutic applications

Gum-based nanomaterials offer various applications, from sensing environmental pollutants to removing harmful microbes and chemicals present in the environment and water (Padil et al. 2018). Polymers of plant origins have been intensely studied and used for their medical preparations, such as tablets and ophthalmic solution formulations, suspensions, implants, surgical lubricants, medium for bacterial growth, and laxatives nanomedicine, and several other applications. Due to their natural existence, biosafety, biocompatibility, and biodegradability, gum polysaccharides are applied in different medical fields specialties such as diagnostics, surgery, therapeutics, etc. Nanoparticles are prepared from several types of materials and mostly are polysaccharides such as gums and phospholipids. Such nanoparticles have a broad range of applications in therapeutics of a variety of diseases. As examples, here we discuss a few of those applications in which gum-based nanoparticles were applied in the therapy of different diseases.

Cancers are very prevalent and one of the most common causes of death worldwide. The word cancer itself is recognized as terror in humanity's history. Intertwined and complex carcinogenesis mechanisms, tumor progression, and tumor metastasis with vague understanding have become challenging to treat cancers. Several modalities and approaches are being used to overcome this challenge, and nanomedicine is one of them. NPs interact at the subcellular level and can be used in intracellular drug delivery. Nanocomposites of pectin-guar gum-zinc oxide (PEC-GG-ZnO) prepared with the size of 50–70 nm were used for the modulation of immune systems to provoke lymphocytes to improve their cytotoxicity against cancer cells (Figure 6) (Hira et al. 2018). Peripheral blood lymphocytes were used to evaluate the immunostimulatory effect and these stimulated lymphocytes were co-cultured with breast cancer cell lines (MCF-7). Cytotoxicity of lymphocytes in peripheral was significantly increased after exposure with PEC-GG-ZnO nanocomposites. This study suggests that gums can play an important role in the immunomodulation of cancer therapy.

An emerging field of nanotechnology has opened new horizons for nanoparticles' applications, like in medicine, increase the therapeutic efficacy of drugs, and in several other disciplines of life. Antibacterial application in various gums-mediated NPs has been reported to produce a synergistic effect on different antibiotics' bactericidal activity (ampicillin, erythromycin, chloramphenicol, and kanamycin) against Gram-positive and gram-negative bacteria. Gum-based nanoparticles showed good stability and high antibacterial activity when AgNPs were at 5 μg concentrations. Bactericidal effect of gum Arabic (GA)-based zinc oxide NPs against *Bacillus cereus* and *Pseudomonas aeruginosa* has been observed. Subsequently, they witnessed low toxicity in cells that resulted in faster wound healing in infected wounds injuries as compared with the ZnONPs. Similarly, Kandagogu gum-based AgNPs used in combination with different antibiotics like streptomycin, ciprofloxacin, and gentamicin against *staphylococcus*, *Pseudomonas aeruginous* and *Escherichia coli*. These two studies by Rastogi et al. justified the feasibility of more rational use of antibiotics by introducing such a combination in therapy invitro. Quelemes et al. evaluated the antimicrobial activity of chemically modified cashew gum through quaternization with ammonium reagent against different strains of *Staphylococcus* including methicillin resistant *Staphylococcus aureus* (MRSA). They found the significant antimicrobial activity of modified cashew gum that was variable in different strains of *Staphylococci*. It was further confirmed that purified and natural cashew gum did not show activity against various strains of *Staphylococci* at the tested concentrations. Minimum inhibitor and bactericidal concentrations against MRSA were 62.5 $\mu g/mL$. It was found to be biocompatible as well. This study may provide a novel antimicrobial agent against MRSA. Combining all of these studies, this combination may be safe for clinical application in bacterial infection due to gums' biocompatible nature. However, clinical translation of in-vitro-experiment is yet to be evaluated for a secure application. Drug resistance is also a significant public health problem being reported in a variety of diseases, especially in cancers and infectious diseases. Plant gum-based nanoparticles may also play an essential role in overcoming the challenge that needs to evaluate.

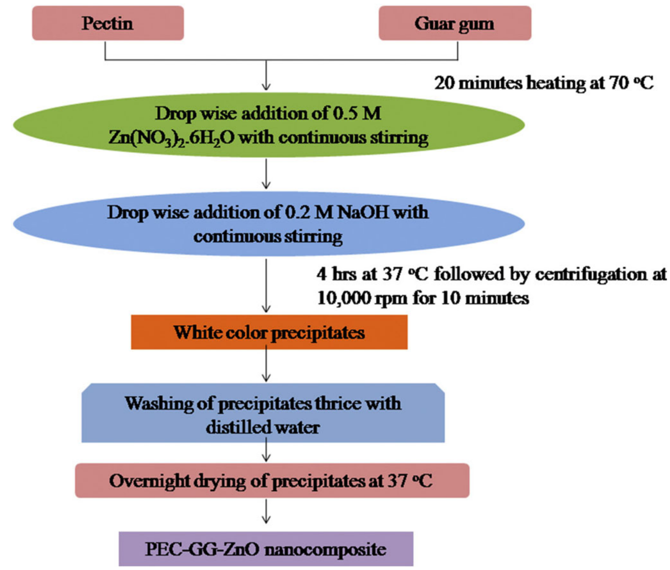

Figure 6. Schematic illustration of the synthesis of PEC-GG-ZnO nanocomposite using precipitation method for enhancing cancer cells killing capabilities of human lymphocytes. Reprinted from Hira et al. (2018) with permission from Elsevier. © 2018 Elsevier B.V. License Number: 4977830237077.

Hyperphosphatemia is defined as increased phosphate levels in the blood and is a commonly observed complication in renal failure patients who undergo dialysis at regular intervals. This hyperphosphatemia rives the different metabolic reversible and irreversible changes in various organs of the body. Hyperphosphatemia in renal failure patients contributes to the rapid renal damage via a vicious cycle, i.e.,

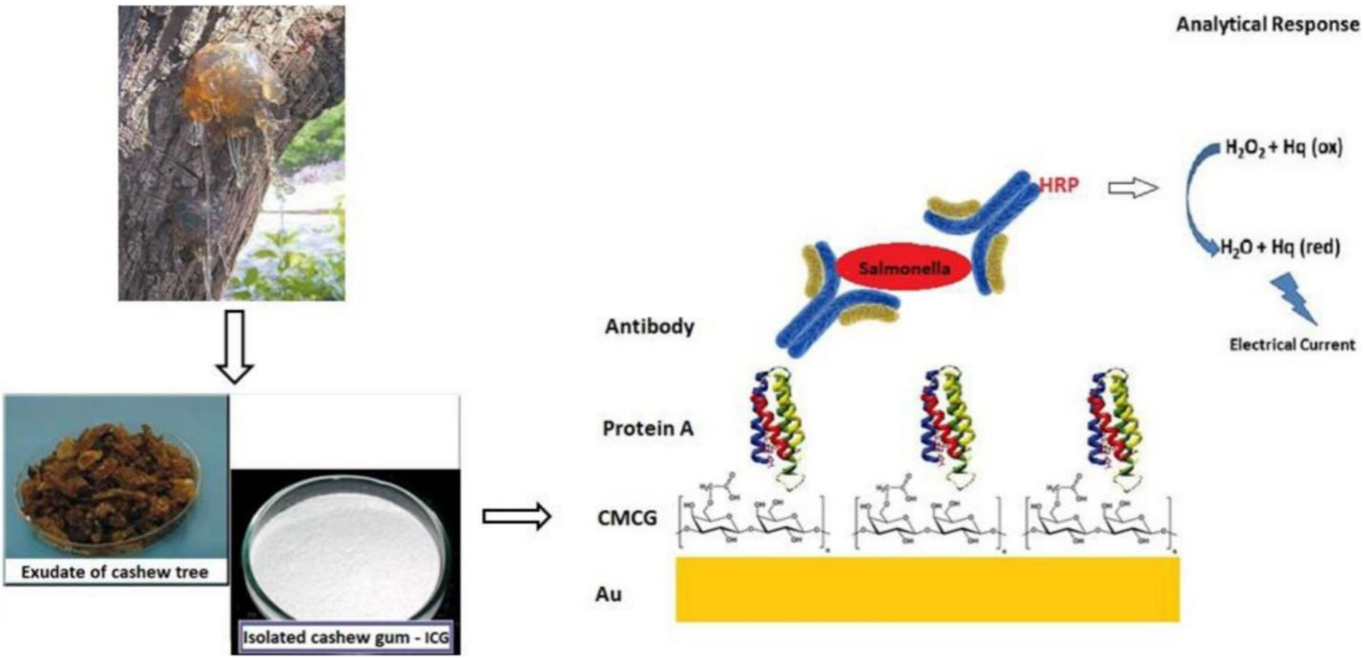

Figure 7. Preparation and characterization of carboxymethyl cashew gum grafted with the immobilized antibody for potential biosensor application. Reprinted from Melo et al. (2020) with permission from Elsevier. © 2019 Elsevier Ltd. License Number: 4977830606277.

phosphorous renal clearance, damage kidneys, and induces electrolyte imbalance and cardiovascular problems. It is believed that the concentrations of phosphate in the saliva of patients with hyperphosphatemia are high. Previously, it was controversial to use chewing gum to absorb the salivary phosphate during the salivation and digestion process, as mentioned by Oh and Urribarri in their report.

Application in biosensors

Gums are an important platform for the immobilization of antibodies, enzymes, and various other biomolecules. A cashew gum platform was designed for protein A-mediated fixation of antibody (Figure 7) (Melo et al. 2020), which was successfully used to detect *Salmonella typhimurium* bacteria with efficiency and rapidity. Nanoparticles of gum tragacanth were used to synthesize highly absorbent hydrogel for biosensing, and quantum dots and glucose oxidase (GO) were fixed on the hydrogel. When glucose comes to contact with GO enzyme, GO oxidizes glucose while generating hydrogen peroxide. The rate of and amount of hydrogen peroxide production depends on the concentration of glucose in the sample. The amount of hydrogen peroxide synthesized was estimated by the characteristic of fluorescence, which was proportional to the concentration of glucose in the sample.

To detect the inflammation of gum caused by implants reduces tissue damage. Ritzer et al. reported that in the peri-implant disease of the oral cavity, matrix metalloproteases are upregulated, and they used upregulated proteases in the oral cavity to improve the diagnosis of peri-implant disease. They reported proteases catabolize a chewing gum that consists of a peptide sensor bond. This catalysis results in the synthesis of a bitter substance and a microparticle. Quantification of bitterness or sourness was measured by potentiometry. Another notable application of plant gums in the biosensor is the estimation of glyphosphate by the principle of potentiometry. This biosensor used agarose-gaur gum (A-G) to entrap the urease bionanoconjugates with gold NPs and was the first device of its kind to detect glyphosphate. Glyphospate inhibits the activity of urease enzyme, and inhibition of enzyme activity was potentiometry ammonium ion-selective electrodes.

Application in tissue regeneration

Tissue regeneration and wound healing are a complex series of events occurring sequentially and are facilitated by inflammatory cells synthesizing growth factors, chemokines, and metabolites. Collagen formation is commonly observed. If regeneration delays and injury become chronic due to certain factors, it may result in fibrosis and tissue or organ death. Several bacteria are known to infect the wounds and delay the process of tissue regeneration and wound healing. The application of antibiotics and other drugs like corticosteroids is an essential factor responsible for reduced risk of wound infection and fasten tissue regeneration. The critical role of gum-based NPs has been reported to mediate wound healing and reduce scar formation and fibrosis risk in infected wounds. To minimize the effect of infection toxicity and improve wound healing and tissue regeneration, GA-mediated zinc oxide NPs were applied to *Bacillus cereus*, and *Pseudomonas aeruginosa* infected wounds. As a result, it reduced the cytotoxicity, and rapid wound healing was observed as compared with ZnONPs. A hydrogel synthesized by calcium ion mediated cross-linking of sodium alginate with GA preserved the control and sustained release of recombinant human mitsugumin protein in-vitro. In vivo experiments were carried out on mice, which reflected the efficient release of the protein at the injured site resulting in enhanced epithelial tissue regeneration and reduced scar formation.

Stimulation of static electromagnetic and magnetic field (MF) also plays an essential role in the resolution of bone injury In addition to this combined MF, rotating MF, and sinusoidal MF also have the potential to promote tissue regeneration and wound healing Either MNPs improve the activity of growth factors, stem cells and scaffolds, applied alone or in combination with a magnetic field that enhances bone regeneration. Growth factors, stem cells, and scaffolds are key modulators for bone regeneration. MNPs and magnetic fields influence cellular pathways and ion channels. Magnetic field and MNPs-based scaffolds can optimize cell recruitment through magnetic and mechanical stimulation, and magnetic cell labeling can drive the patterning, targeting, and gene modification. MNPs may serve as cargo for the delivery of drugs, certain growth factors, and transfection of genes. Magnetic field possibly modulates cell cycle, proliferation, differentiation, gene expression, and morphology. Differentiation of osteogenic cells like bone marrow stem cells (SCs), a cell line of human osteosarcoma, human adipose-derived mesenchymal SCs, dental pulp SCs, rat calvarias cells, and calvarial osteoblasts were enhanced by magnetic field application. Several types of nanoparticles are formulated with gums to improve their efficacy in drug delivery, in which plant gums are used as linking and coating agents to increase their retention in circulation and bioavailability. More importantly, different types of gums are used to link the targeting moieties in targeted drug delivery and certain other modalities of diagnostic imaging. MNPs are widely used in the delivery of a variety of modalities in diagnosis and therapies. Therefore, combining the delivery of different materials through MNPs and using different MF effects in tissue regeneration could play a synergistic role and improve the effects of therapy by modifying various factors.

Conductive adhesive properties and pharmaceutical applications

The biomedical significance of gum-imprinted renewable bio-nanostructures mainly metal/metal oxide NPs has been studied in numerous areas such as conductive adhesive properties and pharmaceutical applications. Gums convert powder mass into granules, and these granules are compressed into tablets. Due to their adhesive nature, in this way, they act as a binder. In addition to adhesion and cohesion, gums also exhibit dissolution property and a swelling ratio of five times their actual volume by absorbing water. Swelling results in disruption of the tablet into smaller fragments that further increase the dissolution speed. Albizia gum was assessed as a tablet binder along with its mechanical properties and disintegration and compared with gelatin. Ferris and In Het Panhuis (2009) proved the conduciveness of gellan gum, a bio-materials obtained from *Pseudomonas elodea*. In another study, Keller et al. (2017) conducting hydrogels for edible electrodes. Both the gum membranes and fibers display noteworthy antibacterial activities against Gram classes of bacteria. Their potential antibacterial efficacies and surface modification result in biocompatibility and cytotoxicity, used in various pharmaceutical and biomedical applications.

Application in imaging and cell studies

Another important application of Gum-based nanoparticles is their use in cell studies and imaging. It is commonly used to specific couple molecules and is widely used in targeted drug delivery to link the nanoparticles with a targeting moiety. Gold (Au) nanoparticles coupled with folic acid (FA) to deliver the nanoparticles at the target site in and fluorescein isothiocyanate (FITC) induce fluorescence for cell imaging. Coupling of AuNPs with FA and FITC achieved by using gum kondagogu (Kumar et al. 2018) Various NPs are applied to track the tumors or cells by Magnetic resonance imaging (MRI) that is a noninvasive imaging technique to study the NPs concentration in a specific organ and or their distribution inside the body. Patitsa et al. synthesized ferrous NPs and furnished them with a polyarabic acid. They witnessed that the polyarabic acid markedly enhanced the Doxorubicin binding reversibly with NPs. Then he studied the cellular uptake of NPs by breast cancer cells in-vitro. They concluded that polyarabic acid-coated nanocarriers provide an efficient platform for drug delivery (Patitsa et al. 2017). Such a Ferromagnetic NPs can be used in MRI for diagnosis, distribution of drug inside body and image directed therapy. GA is a natural glycoprotein and widely used in nanomedicine. It provides stability and reduces the clustering of nanoparticles inside the body. AuNPs synthesized and stabilized by GA as an X-ray contrast agent for tumor imaging in the animal model study.

Conclusions and futuristic outlook

In summary, engineering gums polysaccharides based renewable bio-nanostructures with unique structural, morphological, and functional attributes is a ground-breaking platform with an optimistic vision and applications. Aiming to strengthen the 21st century, bio- and non-bio sectors, the plant-based gums polysaccharides and their engineered bio-nanostructures signify/spotlight the progressive properties for several applications, as discussed above in the representative sections/subsections of this review. Briefly, the current work presented insight and applied perspectives of various gums, i.e., gum Arabic, gum albizzia, gum karaya, gum tragacanth, gum kondagogu, and others, as candidates to construct nanocubes with therapeutic and functional values to tackle the current challenges and limitations of in practice counterparts. The overall ineffectiveness or inefficiency of these in practice synthetic counterpart's necessities new robust and highly effective materials of natural origin for 21st-century medical and biotechnological settings. Though the current technological and nanotechnological advancement has made a significant improvement in the pharmaceutical sector of the modern world. However, several methodological measures and their resulted engineered constructs are still at their fantasy and facing challenges due to poor pharmacokinetics and inefficient or ineffective mode of action. The current limitations of synthetic counterparts can be tackled by deploying or imparting the inherited bio-active features and tunable functional attributes of gums polysaccharides-based bio-nanostructures. Plant-based gums polysaccharides offer numerous advantages, such as renewable, biocompatible, biodegradable, high cost-effective, structurally and functionally tunable, and nontoxic, all that urges researchers to use to design therapeutically effective and industrially efficient carriers. With reference to considerable applied aspects, gum-polysaccharides have appeared valuable materials with significant retention capabilities, tunable active units, swelling, and colloidal features for modulating drug delivery, tissue regeneration systems, etc., and biosensing cues without compromising their inherited traits.

As conferred above with suitable examples, the literature specifies the state-of-the-art advances to extract and stabilize the gums polysaccharides to fabricate renewable bio-nano-structures using different co-materials, such as metallic nanoparticles and others. In turn, the engineered nanocubes can effectively be used as new carriers to treat wounds of numerous categories and related skin disorders, controlled or sustained drug delivery, cancer therapy, diagnosis, sensing, and imaging platforms. Considering all these points, gums polysaccharides based on renewable bio-nanostructures remain a subject of intensive future research. Besides the above-mentioned advancement and technological inputs, further in-depth analysis is needed to understand the exact action mechanism responsible for the high efficacy of gums polysaccharides based renewable bio-nanostructures in the industrial and biomedical sector of the modern world. This review's compendium is that gums polysaccharides and their engineered bio-nanostructures are the emerging and most promising area in the industrial, biotechnological, biomedical, and healthcare applications.

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Conflict of interest

The authors declare that they have no conflict of interest.

ORCID

Muhammad Bilal http://orcid.org/0000-0001-5388-3183
Hira Munir http://orcid.org/0000-0002-9732-0419
Muhammad Imran Khan http://orcid.org/0000-0001-6898-4630
Mohsin Khurshid http://orcid.org/0000-0002-3196-2857
Tahir Rasheed http://orcid.org/0000-0001-9265-6303
Komal Rizwan http://orcid.org/0000-0002-2011-1600
Marcelo Franco http://orcid.org/0000-0002-7827-789X
Hafiz M. N. Iqbal http://orcid.org/0000-0003-4855-2720

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