



Advanced Biomedical Applications of Multifunctional Natural and Synthetic Biomaterials

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Abstract: Biomaterials are mostly any natural and synthetic materials which are compatible from a biological point of view with the human body. Biomaterials are widely used to sustain, increase, reestablish or substitute the biological function of any injured tissue and organ from the human body. Additionally, biomaterials are uninterruptedly in contact with the human body, i.e., tissue, blood and biological fluids. For this reason, an essential feature of biomaterials is their biocompatibility. Consequently, this review summarizes the classification of different types of biomaterials based on their origin, as natural and synthetic ones. Moreover, the advanced applications in pharmaceutical and medical domains are highlighted based on the specific mechanical and physical properties of biomaterials, concerning their use. The high-priority challenges in the field of biomaterials are also discussed, especially those regarding the transfer and implementation of valuable scientific results in medical practice.

Keywords: biomaterials; biocompatibility; biomedical; biodegradability; tissue engineering; biopolymers; bioceramics

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1. Introduction

Maintaining the quality of life through good health is required, and it is absolutely necessary worldwide. Currently, the permanent and growing interest in the study and advanced development of biomaterials is focused on medical or healthcare applications due to the requirements in the domain of drug administration, tissue engineering, medical device industry, permanent or temporary implants. As the need for high-accuracy and individualized treatments continues to grow, research pursues both the improvement of biomaterials in classic applications and the obtaining of new effective bioactive materials for multiple advanced applications, regenerative medicine or other health dysfunctions, including theranostic biomaterials (diagnosis, monitoring capabilities and combined therapy) [1–9].

In current medicine, biomaterials have an important role, both in stimulating healing and for regeneration of the initial biological or functional activity. Biomaterials can exist in natural or synthetic form. They are exploited in multiple applications in the medical field due to their biocompatibility. In this regard, biocompatibility can be defined as "the study and the knowledge of interactions between living and non-living materials" [10] and biomaterials as "any materials projected to interface with biological systems from the human body to evaluate, support, treat, enhance, augment, restore or replace any damaged tissue, organ, or function of the body" (definition proposed by the European Society for Biomaterials Consensus Conference II) [11]. Because of the fact that biomaterials are in direct contact with the human body fluids and tissues, they must accomplish several features such as stability, biocompatibility and safety.

Historical sources indicate the first use of biomaterials in antiquity [12]. In ancient Egypt, it was practiced since 3000 BC by using animal tendons as sutures, coconut shells to

repair injured skulls, wood and ivory as false teeth [13]. Likewise, in Greece and India since the 1st century BC, doctors performed plastic surgeries on wounded soldiers practicing the use of biomaterials [14]. The first more elaborate applications of natural biomaterials are dated in the modern period, with the first surgical operation used to replace the hip with ivory performed in Germany, in 1891 [15].

Starting with the 19th century, bone plates began to be integrated into successful operations, with the role to stabilize bone fractures and to speed up their healing. In 1951, the first allograft taken from a deceased person was successfully implanted, and in 1954, a synthetic arterial substitute was used for the first time to treat 10 patients. In 1958, the Dacron vascular prosthesis was obtained as a viable alternative to harvested human grafts, which presented certain complications after implantation [16,17].

The first artificial heart valves start from the early 1950s, when a methacrylate ball was implanted in the descending aorta by Charles Hufnagel. In 1960, the mitral valves were replaced using a flexible polyurethane mitral prosthesis with a Teflon chorda tendineae attachment, by Nina Braunwald. The last step in valves' evolution is represented by the progress in advanced valve alternatives called regenerative or tissue-engineered valves [18].

The history of biomaterial applications is presented in Figure 1 (adapted from reference [19]).

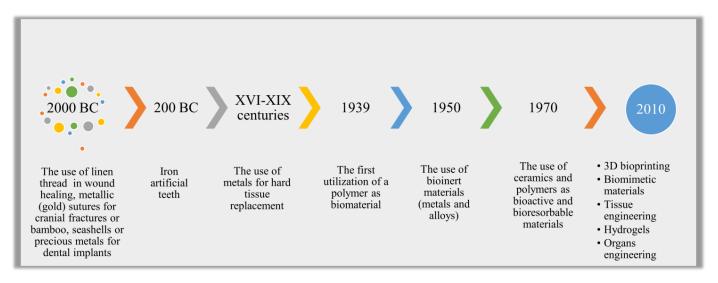


Figure 1. The evolution of materials as biomaterials from prehistory to recent time (adapted from reference [19]).

To exemplify the continuous growth in the publications on and the increasing research attention to this topic, a graphical representation showing the annual number of publications on biomaterials used for biomedical applications from 2003 to 2023 (Figure 2a) is included, which was obtained using the Scopus database [20]. The diagram highlights a significant growth in the publication numbers in the analysed 20 years. The search was performed using "biomaterials for biomedical" in all fields, "biomedical" in keywords and limited only to articles and reviews. In addition, a representation regarding the publications evaluated by subject area is shown in Figure 2b.

In this context, the present review aims to scrutinize the most recent technologies related to advanced biomedical applications of multifunctional natural and synthetic biomaterials and their classification based on their source. The purpose and the objective of the present work is to emphasize the design and functions of a new generation of biomaterials inspired by natural ones and capable of being critical mechanisms in various sides of modern civilization.

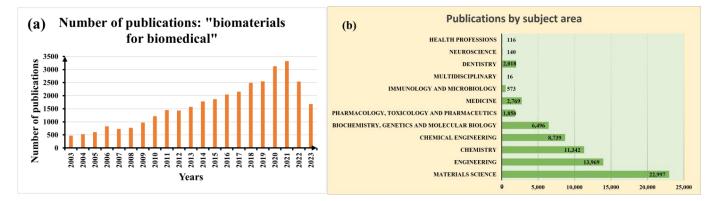


Figure 2. (a) Evolution of the number of scientific papers related to the search of "biomaterials for biomedical applications" phrase published between 2003 and 2023. (b) Evolution of publications by subject area. Source: Scopus (accessed on 12 August 2023) [20].

2. Biomaterials from Natural Sources

Presently, an increased interest has been focused on the development of biomaterials obtained from natural sources due to their several advantages such as biocompatibility, ease of production, renewability, low cost, availability, nonimmunogenicity [21]. Biomaterials derived from natural sources can be grouped in four main categories: protein-based materials, polysaccharide-based materials, glycosaminoglycan-based materials and extracellular matrix materials (Figure 3).

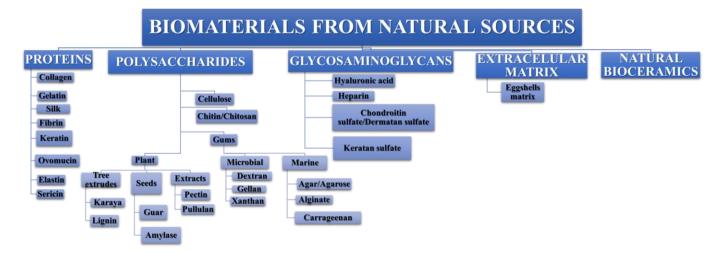


Figure 3. Classification of biomaterials obtained from natural sources.

Generally, biomaterials derived from natural sources are used (i) to restore or to replace a damaged tissue or organ, (ii) to promote the tissue regeneration, (iii) as a drug delivery system, (iv) to develop bone scaffolds.

2.1. Protein-Based Materials

Recently, a growing interest has been observed in the development of protein-based biomaterials due to the limited fossil source and also to their versatile characteristics such as high mechanical performance, biocompatibility and biodegradability [22–24].

2.1.1. Collagen

Collagen is the main protein from the whole animal body protein content (25–35%), with a significant role in providing support for tissues and cells and in sustaining structural and biological integrity of the extracellular matrix. Collagen is found in the skin, tendons, ligaments, cartilage, bones, corneas, blood vessels, intervertebral discs, gut and teeth [25].

Well-suited to build structures for tissue engineering, collagen is a biomaterial often used in biomedical applications based on its biocompatible, biodegradable and noncytotoxic properties [26,27]. Collagen can be processed and used in a variety of forms: (i) collagenbased films or membranes (2D) which are used especially for healing skin wounds and tissue regeneration (cornea, bones) due to good biocompatibility and biodegradability [28]; (ii) in different forms of scaffolds (3D), as pure collagen structures [29,30] or with synthetic polymers [31]. A schematic representation of collagen at different length scales and of different structures is shown in Figure 4 [32].

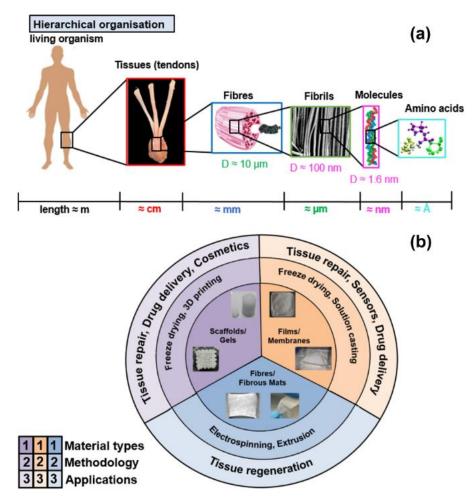


Figure 4. (a) Hierarchical assembly of collagen at different length scales; (b) various collagen structures with the corresponding processing techniques and related biomedical applications. (reprinted from reference [32]).

Collagen-based sponges are used in intraoperative surgery (in neurology) [33] as scaffolds in bone tissue engineering [34], as haemostatic dressings [35] or as an alternative biomaterial for the healing of cutaneous wounds with antibacterial properties [36]. Collagen blended with other biomaterials is used in reconstructive surgery [37] and as a haemostatic dressing for fast blood absorption [38].

2.1.2. Gelatine

Gelatine is a natural polymer derived from the hydrolysis of collagen, being a biocompatible, biodegradable, elastic and nontoxic material. In the field of tissue engineering, gelatine is often used in several systems, such as drug delivery systems, injectable hydrogels, scaffolds or wound dressing films. A mixture of demineralized bone matrix and gelatine together with absorbable gelatine sponges has been tested in bone tissue engineering to maintain haemostasis in multiple surgeries [39]. Gelatine has a disadvantage in terms of thermostability. Depending on the temperature, it can pass relatively easily from the solid state to the gel [40]. At the same time, one of its advantages is the ability to absorb water, useful in tissue regeneration, as in the case of formulations such as nanocomposite hydrogels for treating wounds [41] or injectable hydrogels to favour bone regeneration [42].

2.1.3. Silk

Silk, a fibrous biomaterial obtained from protein fibroin from silkworm cocoons or spider silk blends, is a biocompatible and biodegradable natural polymer frequently used for applications in regenerative medicine and tissue engineering [43]. The mixture with different natural or synthetic polymers or fibrous materials is used to improve its biodegradability, biocompatibility and to adjust the mechanical properties, by obtaining functional biomaterials in the form of hydrogels [44], thin films or coatings [45], nanoparticles [46], fibres, 3D printed structures [47] or in the form of composite scaffolds [48] for the medical field.

2.1.4. Fibrin

Recently, biomaterials or their composites were often used for tissue engineering, due to their bioavailability and low price. Fibrin is one of the most promising natural biomaterials for articular cartilage repair [49]. Fibrin polymers and composites are often used as transport vehicles for bioactive molecules to induce regeneration and promote wound healing or as delivery carriers for multiple cell lines (Figure 5) [50].

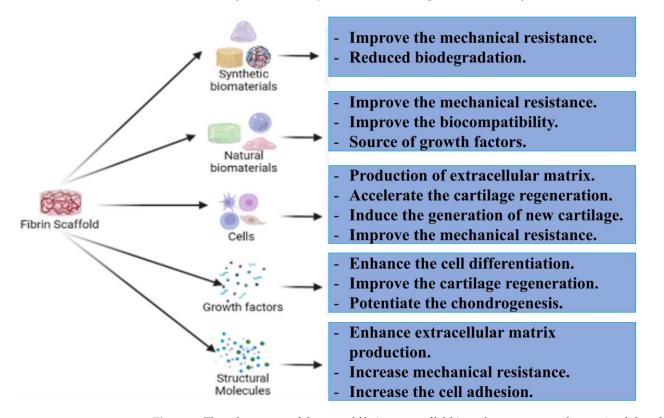


Figure 5. The advantages of the use of fibrin as a scaffold in order to promote the repair of chondral tissue (adapted from reference [50]).

2.1.5. Keratin

Due to their intrinsic biological properties and excellent biocompatibility, keratin-based biomaterials have shown increased interest recently and are widely used for biomedical applications such as tissue engineering, drug delivery, wound healing [51]. The main sources of keratin extractives are wool, horns, hair, nails and feathers. Keratin is a structured protein with a high cysteine content (7–15%). The main methods used to extract and

solubilize keratin are reduction, oxidation, alkaline extraction, microwave irradiation, steam explosion and by means of ionic liquids [52]. Keratin-based formulations are used as (i) hydrogels for diabetic wound dressing material, regeneration of pupal tissue, in substrates for cellular attachment and proliferation; as (ii) films for reconstruction of ocular surface, drug delivery and as (iii) fibres for tissue engineering [51].

2.1.6. Ovomucin

Ovomucin is a glycoprotein carbohydrate (representing between 2 and 4% of total egg albumin) with high molecular weight, composed of a carbohydrate-rich β -subunit (50–57%, a molecular weight of about 400 kDa) and a carbohydrate-poor α -subunit with approximately 11–15% of carbohydrates [53]. Ovomucin possesses antibacterial, antitumor, antiviral, antioxidant activities and immune properties with multiple applications in intestinal injury treatment of gastrointestinal tract after dietary intake or as biocompatible porous hydrogel for additional development as an implant material for bone or tissue engineering [54,55].

2.1.7. Elastin

Obtained by extraction from different biological sources, elastin, a hydrophobic amino acid with a high degree of intermolecular cross-linking, is used as a dressing to speed up the healing of chronic wounds or for large surfaces [56]. Elastin, an extracellular matrix protein, is recognized for providing elasticity to organs and/or tissues. Consequently, elastin is present in human organs with elastic properties such as blood vessels, elastic ligaments, in lung and in skin. Elastin-based biomaterials are used in a wide range of applications such as tissue engineering, skin substitutes, vascular grafts, heart valves and elastic cartilage [57].

2.1.8. Lactoferrin

Lactoferrin is a mammalian iron-binding glycoprotein which belongs to the transferrin class. Recently, lactoferrin has gained potential as a therapeutic agent and as a pharmacological compound. Lactoferrin possesses immunomodulatory, anti-inflammatory, antioxidant, antiviral, antimicrobial and anticancerogenic activities. Lately, various technologies have been developed to advance the lactoferrin's role in the ocular drug bioavailability (Figure 6) [58].

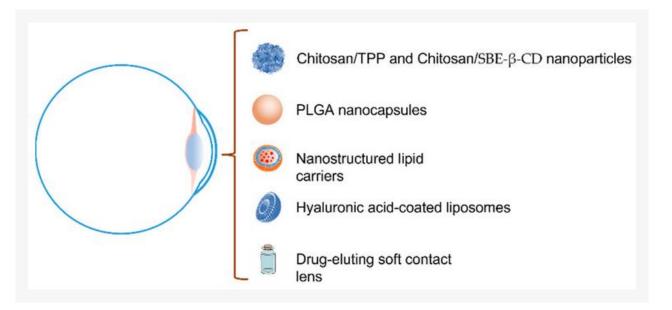


Figure 6. The improvement methodologies of lactoferrin ocular drug bioavailability. (reprinted from reference [58]).

2.1.9. Sericin

Sericin, a component of silk known as silk sericin, has grown consideration in recent years in sericin-based biomaterials. Due to its role in the sustenance of cell proliferation and attachment and stimulation of cell differentiation, sericin has been used in tissue engineering and cell culture [59–71]. A summary of sericin-based biomaterials for biomedical applications is presented in Figure 7.

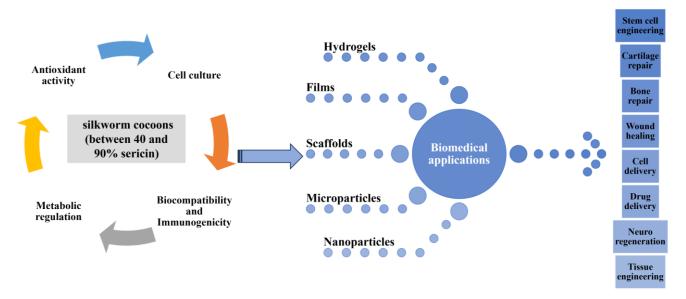


Figure 7. Examples of sericin-based biomaterials, their properties and biomedical applications.

2.2. Polysaccharides

2.2.1. Cellulose

The main sources of cellulose in nature are plants (wood and secondary products from agriculture), algae, bacteria and marine animals, being practically an abundantly available biomaterial [72]. Cellulose can be found in three forms, namely cellulose nanocrystals, cellulose nanofibers and bacterial or microbial nanocellulose. Cellulose nanocrystals demonstrate unique characteristics, such as large surface area, good crystallinity, as well as excellent mechanical strength. [73]. Cellulose-based materials can be used as antibacterial agents, for tissue engineering, for wound dressing, for artificial blood vessels and for drug delivery in the form of aerogels, hydrogels, three dimensional scaffolds and membranes [74].

2.2.2. Chitin/Chitosan

Chitosan and its precursor chitin are bioavailable polymers that come from the shells or skeletons of marine and nonmarine organisms [75]. They have been studied and used in a very large range of biomedical and nanobiotechnology applications because they have advantageous properties, such as nontoxicity, biocompatibility, biodegradability. Because chitosan is water-soluble and easily forms ionic and hydrogen bonds with drug structures, it can be widely used in drug delivery systems, cancer therapy, wound healing and tissue engineering [76]. To maximize the therapeutic potential and bioavailability of drugs in a controlled and continuous manner [77], various micro- and nanoparticles have been developed as their active carriers [78,79], hydrogels [80–83] scaffolds [84–87] and organic or inorganic matrices based on chitosan or chitin [88].

2.2.3. Gums

Natural gum-based polysaccharides and their derivatives are biodegradable polymeric materials which present advantages over synthetic polymers, mainly since they are nontoxic, bioavailable and inexpensive [89]. Natural gums can be divided based on their sources and chemical structures as follows: (i) of plant origin (tree exudates, seed gums and extracts), (ii) of microbial origin and of (iii) marine origin. Gums are principally synthesized from the following three categories: (a) tree or shrub exudates, (b) plant exudate gums and (c) the endosperm of some seeds [90]. A graphic illustration of the natural gum sources, gum-based materials and their possible biomedical applications is shown in Figure 8.

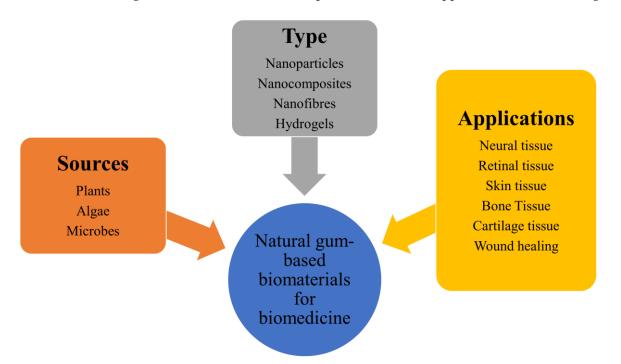


Figure 8. Diagram of sources of natural gums and their advanced biomedical applications (adapted from reference [90]).

The significant characteristics of natural gums are (i) the ability to form viscous solutions by attracting and binding water molecules, (ii) the ability to act as fibres and (iii) the ability to form films. Besides their origin, natural gums are also grouped and classified into the following categories:

- (a) Anionic, cationic and nonionic, based on the surface charges;
- (b) Linear and branched chains, based on their structure [91].

Plant Gum-Based Biomaterials

(i) Tree extrudes

Gum arabic, a natural polysaccharide obtained from *Acacia nilotica*, is part of the Leguminosae family. It is composed of different compounds: galactose (39–42%), arabinose (24–27%), glucuronic acid (15–16%), rhamnose (12–16%), protein (1.5–2.6%), nitrogen (0.22–0.39%) and moisture (12.5–16%). Its main application is in the food industry, pharmaceuticals and cosmetics. Recently, gum arabic has been extensively used for drug delivery and in a biomedical domain, as nanoscaffolds [92–95].

Gum karaya is produced from Sterculia species trees. Gum karaya is an exceptional natural vegetable bioproduct, with good biocompatible, biodegradable, renewable properties, it is easy to handle and store, has a low cost and is frequently used in food, pharmaceutical, denture-fixation, wound-dressing, nanoencapsulation and other industrial applications (cosmetics, textile and paper industry) [96–99].

Lignin is the main component of plant cell walls, which protects the plant from different stress factors and provides mechanical support. The chemical structure of lignin is complex, being composed of methoxy, phenolic and aliphatic hydroxyl groups. Lignin has satisfactory biocompatibility. Its cytotoxicity depends on the process of chemical extraction, as well as antioxidant and antimicrobial properties. The application of lignin in the biomedical domain can be summarized as follows: for wound healing as hydrogelbased materials, in tissue engineering as hydrogels and nanofibers, in drug delivery due to its ability to encapsulate hydrophobic/lipophilic drugs for oncological therapy [100–105].

Gum tragacanth, an acidic and anionic polysaccharide having a high molecular weight and durability, is produced from various species of Astragalus. This gum possesses good stability and biodegradability, is available in nature and is nontoxic [106]. Gum tragacanth is used in many biomedical applications for wound healing, prevention of growing of cancer cells, tissue engineering and as a gelling agent in pharmacy [107–109].

(ii) Seed gums

Guar gum, a natural hydrophilic polysaccharide derived from the endosperm of *Cyamopsis tetragonolobus* or *Cyamopsis psoraloides* seed, is formed by linear 1,4 linkages of β -D-mannopyranose, and the branch contains 1,6 linkages of α -D-galactopyranose. Guar gum is highly soluble in water, is nontoxic over a wide range of pH; its hydrogels are biodegradable in nature, have higher flexibility and are biocompatible. Based on these properties, guar gum has been widely applied in the biomedical domain for targeted drug delivery and in tablets [110–113].

(iii) Extracts

Pectin extracted from biomass, using different treatment conditions, can be used in biomedical applications. Several applications of pectin can be mentioned in 3D printable inks as scaffolds with chitosan [114], in drug delivery applications [115], as a hydrogel for cancer-targeted drug delivery [116], in tissue engineering [117] and for wound healing [118].

Pullulan, a fungal exopolysaccharide which is produced by *Aureobasidium pullulans* and *Aureobasidium melanogenum* under aerobic conditions, has several properties that make it highly soluble in water, tasteless, edible, odourless, nontoxic and biodegradable [119]. Among biomedical applications of pullulan that can be mentioned are for bone formation and/or repair [120], wound healing [121], tissue regeneration [122] and drug delivery [123,124].

Microbial Gum-Based Biomaterials

Dextran is a nontoxic, hydrophilic, biodegradable and biocompatible homopolysaccharide derived from bacterial attack of sucrose with dextransucrase or maltodextrins with dextrinase [125]. This biocompatible homopolysaccharide has been broadly applied in pharmaceutical and biomedical applications to stimulate wound healing and rehabilitation of skin and to decrease the inflammatory response [126–129].

Over the past few years, gellan gum, a high-molecular-weight linear anionic exopolysaccharide, formed by microbial fermentation of *Sphingomonas paucimobilis*, has been paid attention and considerably used in some areas such as clinical and biomedical applications [130]. Due to its adaptable characteristics such as tuneable mechanical properties, biocompatibility, biodegradability, easy functionalization and fabrication, gellan gum has attracted the researchers as a promising biomaterial in regenerative medicine (cartilage and intervertebral disc repair), drug delivery and tissue engineering [131–133]. Generally, gellan gum is used in composite biomaterials by blending it with additional biopolymers or nanomaterials in order to improve the poor mechanical strength and lower stability [134–136].

Xanthan gum, an anionic polysaccharide is produced by a Gram-negative bacteria named *Xanthomonas* bacteria. Xanthan gum due to its biodegradability and biocompatibility has been used in numerous biomedical applications alone or in combination with other natural and/or synthetic polymers in tissue regeneration [137,138], wound healing [139–141] and the controlled release of drugs [142].

Marine Gum-Based Biomaterials

Agar, a hydrophilic polysaccharide, is extracted from red seaweed with a composition containing 30% agaropectin and 70% agarose (the main component responsible for the

gelation process). Its usage as hydrogels or air-dried films was reported in wound dressing and wound healing [143,144].

Alginate is an anionic biopolysaccharide formed by units of mannuronic acid and glucuronic acid, randomly arranged [145]. Alginate has been extensively used in tissue engineering, cancer therapy, wound care, controlled drug delivery and nanomedicines due to its biocompatibility [146–152].

Carrageenan is a naturally occurring, linear sulphated polysaccharide extracted from red seaweeds. It is composed of the disaccharide units alternating 4-linked α -D-galactopyranose (the B unit) and 3-linked β -D-galactopyranose (the A unit) residues. Carrageenans hold several advantages such as abundant availability, biocompatibility and biodegradability, antiviral activity, antimicrobial properties, sustainability permitting their usage as remarkable functional biomaterials for advanced biomedical applications [153–159].

2.3. Glycosaminoglycans

Both fibrous proteins and distinct polysaccharides named glycosaminoglycans are present in the composition of the extracellular matrix. Glycosaminoglycans are formed by a linear chain represented by repeated disaccharide units [160,161].

2.3.1. Hyaluronic Acid

Hyaluronic acid, a linear anionic polysaccharide, is a major macromolecular component of the extracellular matrix in the most connective tissues. Due to its biocompatible, biodegradable and bioresorbable nature, limited immunogenicity, recognized by cell surface receptors, its flexible and unique viscoelasticity properties, hyaluronic acid has been extensively studied in recent years for biomedical applications [162–164]. This biopolymer is useful in medicine for the treatment of diverse pathological situations such as arthritis but is also used in drug delivery and tissue engineering [165,166].

2.3.2. Heparin

Heparin is an acidic sulphated polysaccharide belonging to the glycosaminoglycan family which is isolated by extraction from animal tissues such as porcine intestine. Heparin is formed by repeating disaccharide units of $1 \rightarrow 4$ -linked hexuronic acid and glucosamine saccharide residues. Heparin is used in the manufacture of membranes [167] or in the novel polyelectrolyte-scavenging method for biomedical applications [168]. This glycosaminoglycan has been proposed for the cystic fibrosis treatment, for neurodegenerative diseases, as an antimicrobial agent, and in the pancreatitis treatment [169].

2.3.3. Chondroitin Sulphate/Dermatan Sulphate

Studies initiated in the 1950s on bovine material and developed later, led to a better understanding of the biosynthesis and homeostatic roles of chondroitin sulphate/dermatan sulfate, especially in the control of bone development and in the regulation of the assembly of collagen fibrils as well as cytokines. These findings were followed by highly significant correlations between CS/DS synthesis and certain genetic abnormalities in humans [170].

2.3.4. Keratan Sulphate

Keratan sulphate (KS) is the newest glycosaminoglycan, an elaborated molecule with a distinctive structure, whose function is still to be revealed; cornea, cartilage and brain being the main tissue sources of KS in the human body. It has an important role in the regulation of cells in epithelial and mesenchymal tissues, as well as in bones [171].

2.4. Extracellular Matrix

The role of the extracellular matrix and vascular basement membranes and their influence in the regulation of angiogenesis and the progression of some tumours has been studied and highlighted [172]. Various biomaterials of synthetic and natural origin

have been investigated as new scaffolds for heart valve tissue engineering, especially for paediatric patients. Among them, glycosaminoglycans such as chondroitin sulfate and hyaluronic acid are the main components of cardiac cushions, as important parts that regulate the functionality of the complex system of heart valves [173].

Eggshells Matrix

By recycling biological waste such as eggshells and using different natural extracts or other compounds, materials similar to natural bone minerals have been obtained for orthopaedic and dental applications [174], such as hydrogels with super absorbent properties for drug delivery systems [175] or composite materials in electrochemical sensors for the determination of ascorbic acid, dopamine and uric acid in human urine and blood serum [176].

2.5. Bioceramics

Based on their origin, bioceramics can be classified as follows:

- (i) Natural. Natural bioceramics occur naturally in natural corals, algae, natural pearls, shells, eggshell matrix, teeth and bones.
- (ii) Synthetic. Synthetic bioceramics are produced artificially and include hydroxyapatite, calcium phosphate-based materials, bioglass, alumina, zirconia, silicon nitride.

In Figure 9, a schematic representation of the main natural and synthetic bioceramic materials is shown.

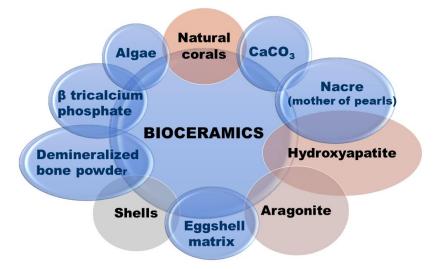


Figure 9. Classification of main bioceramics-based biomaterials from natural and synthetic sources.

Bioceramics are classified based on tissues response into the main three classes as follows:

- Bioactive ceramics represented by hydroxyapatites, fluorapatite-based composites, bioactive glasses and bioactive glass ceramics;
- Biodegradable/bioresorbable ceramics with the presence of calcium phosphate, tricalcium phosphate, aluminium-calcium phosphate, zinc-calcium phosphorous oxides, zinc-sulphate-calcium phosphates, ferric-calcium phosphorous oxides, coralline, calcium aluminates;
- (iii) Bioinert/nonresorbable ceramics which includes alumina, zirconia, carbons and silicon nitride.

Table 1 summarizes the most recent applications of natural and synthetic bioceramics in the biomedical area.

Bioceramics	Examples	Applications	References
Bioactive ceramics	hydroxyapatites	coatings on metallic implants	[177–179]
	fluorapatite-based composites	bone applications	[180]
bloactive cerainics	bioactive glasses	bone substitute and drug carrier	[181]
	bioactive glass ceramics	chemo hyperthermia	[182]
	aluminium-calcium phosphate	biomedical application	[183]
	zinc-calcium phosphorous oxides	postoperative tumour treatment	[184]
Biodegradable (bioresorbable)ceramics	zinc-sulfate-calcium phosphates	tissue engineering	[185]
(bioresorbable)ceramics	ferric-calcium phosphorous oxides	scaffolding for cell and drug delivery	[186]
	coralline	drugs	[187]
	calcium aluminates	bioactive dental materials	[188]
Bioinert (nonresorbable) ceramics	alumina	drug delivery	[189]
	zirconia	biomedical applications	[190]
	carbons	biomedical applications for tissue engineering	[191]
	silicon nitride	biomedical applications in medical implants	[192]

Table 1. Bioceramics used in biomedical applications.

3. Biomaterials from Synthetic Sources

The classification of biomaterials obtained from synthetic sources is presented in Figure 10.

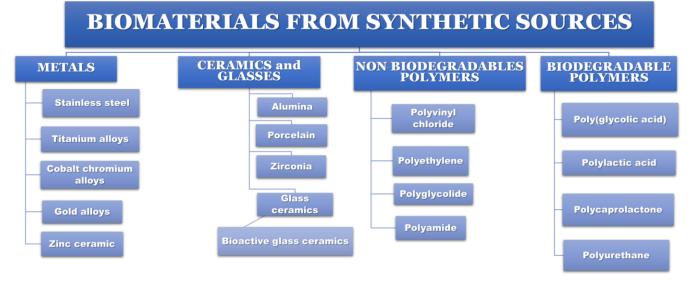


Figure 10. Classification of biomaterials obtained from synthetic sources.

Table 2 lists the main advantages and disadvantages of biomaterials obtained from synthetic sources and their most relevant biomedical applications (adapted from reference [19]).

Category	Advantages	Disadvantages	Applications	References
Metals	high mechanical properties; low friction; high fatigue resistance; ductility; low cost	poor biocompatibility; stiffness; high specific weight; low resistance to corrosion	orthopaedic, orthodontic, cardiovascular, joint prostheses	[193–196]
Ceramics	good biocompatibility; chemical inertness; high compressive strength; low thermal and electrical conductivity; corrosion resistance	low impulsive tensile strength; high specific weight; brittleness; reproducibility; not easy to process	orthopaedic, orthodontic, cardiovascular; coatings; bone tissue regeneration; surgical implants	[197–199]
Nonbiodegradable and biodegradable polymers	toughness; low specific weight; low frictional properties; good processability	low mechanical strength; degradability over time; deformability over time	orthopaedic, orthodontic, cardiovascular, breast implants, scaffolding for soft tissues; eye lenses, artificial tendons	[200–204]
Biologically-derived materials (porcine/bovine pericardium)	superior biocompatibility	poor reliability; difficult handling and storage	bioprosthetic heart valves, total artificial heart	[205,206]

Table 2. Advantages and disadvantages of biomaterials obtained from synthetic sources and their most relevant biomedical applications.

3.1. Metals

Metallic biomaterials are predominantly used to replace damaged hard tissues. Various types of high-strength alloys containing nontoxic elements are used for various biomedical applications. Functional devices based on NiTi (Nitinol) alloys have been developed due to their exceptional shape memory and super elastic properties. The alloy is used for the manufacture of self-expandable stents for cardiovascular surgery as well as super elastic bone staples for orthopaedics or super elastic orthodontic wires for dentistry [207]. Patient-specific medical implants such as bone plates, screws, cranial or dental devices are manufactured with the necessary biocompatibility, bioactivity, surface integrity and wear resistance [208].

Table 3 summarizes recently developed metal-based biomaterials for the use in the biomedical domain.

Table 3. Metal biomaterials used in biomedical applications.
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Metals	Characteristics	Applications	References
Titanium alloys	biocompatibility, corrosion resistance, high mechanical properties, high fatigue resistance, ductility	orthopaedic implant materials, bone plates, dental implants, spinal internal fixation devices, orthodontic wires, intramedullary nails	[209]
Mg alloys	nontoxicity, biocompatibility	bioresorbable orthopaedic materials	[210,211]
Conventional stainless steel	corrosion resistance	metal implants	[212,213]
Platinum	systemic treatment	chemotherapy	[214]
Nickel	therapeutic activity	antibacterial agent	[215]
CoCr alloys	mechanical properties	dental prosthesis; dental restorations	[216]
Nitinol	super-elasticity, shape memory	endovascular stents	[217]

3.2. Ceramics and Glasses

Among the materials commonly used for clinical applications, the biocompatible characteristic of ceramics makes it suitable for implants, tissue regeneration and engineering applications. The demand for high-quality devices is determined by global health care problems of the increasingly numerous populations, such as defects or dysfunctions of the bone skeleton, osteoarthritis or those in the field of dentistry. Ceramics find numerous applications in biomedicine due to their excellent biocompatibility in the dental field, from dental restoration to implants, for joints or bone substitutes, bone fillings and scaffolds for tissue engineering. Some ceramic implants can be used as porous substrates to help the ingrowth of new bone tissue and can be resorbed after tissue regeneration.

A porous nanocomposite material in the form of hematite nanoparticles covered by amorphous alumina was produced by sol-gel combustion synthesis. Porous nanocomposite particles have proven capable of inducing special magnetic properties, making them suitable as contrast agents for imaging applications, namely in ultra-high field MRI [218]. Alumina and zirconia have been used as ceramic fillers for many types of polymer-ceramic composites developed as biomaterials in dental prosthetics [219]. Biofunctional materials, such as silicate glass ceramics, are studied and applied for the regeneration and healing of bone tissue [220], in cancer treatment by hyperthermia [221], in implants [222] or for total replacement in articulating surfaces to promote osseointegration [223]. Larry Hench is one of the pioneers of studies regarding the influence of the chemical structure of bioactive glasses and glass ceramics in general, on the interaction with the physiological system, as well as the connection between the glass surface and the living tissue [224]. Beginning in 1969, Hench developed for the first time a bioactive and biocompatible melt-derived glass material called bioglass, with the potential to form bonds with mineralized bone tissue even in the body's physiological environment [225]. The main purpose of his study was to replace inert implants made of other materials (for example plastic or metal) that were not well supported by the human body. Most bioglasses contain Na₂O, CaO, P_2O_5 and SiO₂ in their formula. The latter was added in a weight percentage <55%, because a larger amount of SiO₂ could lead to the loss of bioactivities. In 1969, studies were successfully carried out on rats, on the adhesion of a bioglass to bone and muscle 6 weeks after implantation. This material had a composition of 45SiO₂-24.5Na₂O-24.5CaO-6P₂O₅ by weight (%) and was named 4555 Bioglass[®] [226]. Later, in 1991, Hench proposed the development of bioactive gel-derived glasses [227]. These studies marked a shift in interest from bioinert materials to bioactive materials. Bioactive glass has multiple uses: (i) in solid form, as different prostheses, in dentistry, bone regeneration medicine or in tissue engineering; (ii) in the form of powders for covering biomedical devices; (iii) in the form of composites, as different filling materials and (iv) as a drug delivery system [228–235].

In their work, M Montazerian and ED Zanotto presented an interesting review of the history of bioactive glass ceramics, highlighting their biochemical and mechanical properties, as well as trends in the manufacture of commercial bioglass products for various biomedical applications [236].

Glass-ceramic materials can be made from glasses through a suitable heat treatment, resulting in nucleation phases and the growth of specific crystalline phases inside the remanent vitreous matrix. It has a very fine microstructure with a small number of or no residual pores. These characteristics improve the mechanical qualities of the final product. A favourable microstructure can be achieved both through appropriate heat treatments and through the use of additives that can act as nucleating agents [237]. The main methods of obtaining bioactive glasses and glass ceramics are the traditional melting techniques [238], the sol–gel method [239–241], the synthesis assisted by ultrasound [242], microwaves [243] or the hydrothermal synthesis [244].

3.3. Nonbiodegradable Polymers

Nondegradable polymers are used in biomedicine, being resistant substrates over time and proving a good performance throughout the life of the patients. They are easily processed in different shapes and can be bio-integrated together with other materials into larger or smaller surfaces, in various thicknesses. They are robust and present excellent mechanical properties [245]. In the medical field, nondegradable polymers are used for the manufacture of tissue engineering scaffolds, orthopaedic implants, heart valves, bone and cartilage substitutes, vascular grafts, artificial hips, eye lenses, fillings and other components in the dental field, bone cements. Many nondegradable polymers such as poly(ethylene), poly(propylene), poly(tetrafluoroethylene), poly(methyl methacrylate), polyurethanes, poly(dimethylsiloxane), poly(ethylene terphthlate), poly(sulfone), poly(ethyleneoxide) are processed under trade names by different companies as parts or medical devices for diverse medical dysfunctions. The most important parameters of long-term implantable nondegradable materials are their biocompatibility, biostability and mechanical resistance

3.4. Biodegradable Polymers

to wear [246].

Synthetic biodegradable polymers can be tailored to have an improved and stable range of mechanical and chemical properties compared to natural polymers. They do not present the risk of causing an immune reaction or transmitting microbes or viruses, but the issue of biocompatibility and possible long-term effects such as the appearance of scars or inflammations arises. Among the most commonly employed biodegradable synthetic polymers for biomedical applications are saturated aliphatic polyesters, polyanhydrides and polyurethanes.

Poly(glycolic acid) (PGA), polylactic acid (PLA) and the copolymer poly(lactic-coglycolic acid) (PLGA) are saturated aliphatic polyesters frequently used for the manufacture of 3D scaffolds applied in tissue engineering, due to their biocompatibility and biodegradability [247]. PLA is a material used extensively in biomedical applications and very suitable especially for cardiac, dental or orthopaedic fixation devices, because it is biocompatible. However, it is biodegradable over a long period of time and also can be used for other implantable devices, especially in paediatric patients [248]. It is used in regenerative medicine due to its ability to stimulate hard tissue regeneration in bone grafting procedures, having an extraordinary capacity for bioresorption. Many studies have been reported on PLA-HA [249] or PLA-polycaprolactone-hydroxyapatite composite materials that have highlighted their promising preliminary results for biomedical applications [250]. As a hydrophilic and highly crystalline polymer, with a relatively fast degradation rate in aqueous solutions or in vivo, PGA can be used in drug delivery [251], biological adhesives/glues [252], oral surgical treatments and injectable microspheres [253].

Poly(ε -caprolactone) (PCL) is a useful biodegradable biomaterial that can slowly degrade (in few years) under physiological conditions and is therefore used for long-term implants (in the treatment of bone defects), drug delivery systems [254] and as delivery platforms for various extracellular matrix proteins [255] or 3D scaffolds [256]. Polyanhydrides are hydrophobic polymers whose degradation occurs more through surface erosion than volume. This feature is very practical in the case of the release of certain drugs and especially important in the case of extremely strong drugs. Because water does not penetrate before the polymer is eroded, the implanted drugs are kept safe [257]. The PCL is conventionally used as a local (targeted) delivery vehicle especially for the controlled release of chemotherapeutic agents [258].

Polyurethanes are biocompatible, biodegradable polymers with excellent flexibility, durability and resistance. Due to their biostability, they are favourable inert materials for development of medical devices [259], vascular grafts [260], prostheses [261], heart valves [262,263], catheters [264], drug delivery and as porous scaffolds for tissue regeneration [265].

Table 4 summarizes the recent developments of polymer-based biomaterials in the biomedical domain.

Polymers	Characteristics	Applications	References
Polyesters	degradability, bioresorbability (new polymer classes)	tissue engineering, nanoscaled drug delivery systems	[266]
Polyurethane	easily malleable and flexible, biocompatibility, prominent mechanical properties (high tensile strength, toughness and resistance to degradation)	biomedical devices (cardiovascular surgery, orthopaedic surgery and traumatology, reconstructive surgery, gynaecology and obstetrics, gene therapy, implantable vascular grafts)	[267,268]
Polyamides	biocompatibility, controlled porosity, high processability, excellent stress crack resistance, biocompatibility, good mechanical strength and good stability in human body fluid	antimicrobial wound dressings, wound healing applications	[269,270]
Polysiloxanes	extensible elastomer, low tensile strength and tensile modulus	implantable devices (vascular prostheses), magneto-responsive 4D-printed bioproducts	[271,272]
Acrylics	good mechanical strength, resistant to a wide range of chemicals, excellent UV-light transmittance, good scratch resistance, biocompatibility	odontological applications including artificial teeth, dentures and denture bases, obturators, provisional or permanent crown, biomedical applications	[273,274]
Polymeric composites	biocompatibility, high corrosion resistance, very high compressive yield strength, bioresorbable implant material, antimicrobial properties	orthopaedic applications, tissue engineering, flexible antibacterial surfaces, antiadhesive surface biomaterials, acrylic bone cements, dental adhesives, antiadhesive surfaces for biomedicine, medical sutures	[275–277]

Table 4. Polymeric materials used in biomedical applications.

4. Composite Biomaterials

Composites are materials based on two or more different compounds in order to improve the individual properties of materials such as surface characteristics, mechanical strength and biocompatibility in order to be easy for manufacturing. Polymer-based composites have shown an increased interest in biomedicine applications for drug release, tissue engineering, regenerative medicine, wound dressings, surgical operations, medical imaging in cancer detection and dental resin composites [278,279].

The recently developed biomaterials for their applications in the biomedical area are shown in Table 5.

Material	Characteristics	Applications	References
Smart elastomer composites	rapid light-responsive self-healing ability and shape memory	surgical sutures to promote wound healing (as spiral-like stand)	[280]
HNT-PVA-ALG-Hap composite on alkali-treated Ti-6Al-4V alloy substrate	biocompatible with the human tissues	biomedical applications (bone regeneration)	[281]

Table 5. Composite biomaterials for biomedical applications.

Material	Characteristics	Applications	References
Polydopamine (PDA)-coated hydroxyapatite (HA)-reinforced polyvinyl alcohol (PVA) films	higher mechanical properties, homogeneous mineral distributions, high antibacterial capacities against Acinetobacter <i>Baumannii</i> (A. <i>Baumannii</i>), <i>Staphylococcus aureus</i> (S. <i>aureus</i>) and <i>Streptococcus mutans</i> (S. <i>mutans</i>), good biocompatibility with fibroblast (L929) cells and MCS cells	biomedical fields (tooth-bone treatments for coating, filling or occlusion purposes)	[282]
Electrospun graphene oxide/calcium hydroxyapatite/ polycaprolactone composites	biocompatibility, antimicrobial activity	excellent biological compatibility for prospective application in medicine and clinical dentistry biologically (compatible matrix for potential bone tissue regeneration with antimicrobial effect)	[283]
Ternary nanocomposites, including graphene oxide (GO), hydroxyapatite (HAP) and cadmium selenite (CdSe) encapsulated into nanofibrous scaffolds of polylactic acid	good wettability and mechanical properties, cell viability and cell growth in vitro	advanced bioactive material for effective and fast wound healing, for tissue engineering application	[284]
Dual mineral-substituted hydroxyapatite (DM-HAP) combined with biodegradable polymer alginate-chitosan (ALG-CS) and graphene oxide	improved antibacterial activity performance and bioactivity	tissue engineering applications (orthopaedic applications as soft bone tissue replacements)	[285]
NiFe ₂ O ₄ /NG/cellulose composites	antibacterial activity against Escherichia coli (Gram-negative bacteria) and Bacillus subtilis (Gram-positive bacteria)	biomedical applications (as antibacterial material)	[286]
Biocomposite based on lginate/gelatine crosslinked with genipin	thermal stabilization	bioengineering (support for β-galactosidase enzyme immobilization)	[287]
Chitosan/poly(vinyl alcohol)/graphene oxide (CS/PVA/GO) nanocomposites	biodegradable films with good mechanical, chemical and biological properties	tissue engineering and cell regeneration	[288]
CNF-NCG reinforced drug-loaded PVA/MC/PEG glutaraldehyde cross-linked novel electrospun nanofibrous bio-nanocomposites (BNCs)	biocompatibility, biodegradability	transdermal drug delivery systems	[289]
Biowaste-derived nanophase yttrium-substituted hydroxyapatite/citrate cellulose/opuntia mucilage (Y-nHAP/CC/OM) biocomposites	potential bacterial resistance against both Gram-positive and Gram-negative strains, in vitro cytocompatibility, considerable mechanical, antibacterial and biological properties, valorisation of biowaste material	biomedical applications	[290]

good in vitro bioactivity biocompatibility and antibacterial

properties,

ceramic composites could induce apatite formation in SBF biomaterial for clinical applications

such as orthopaedic and dentistry

[291]

Hydroxyapatite

(HA)/nanostructured monticellite

ceramic composites

17 of 31

Table 5. Cont.

Summarizing, the main properties of biomaterials required for their applications in the biomedical domain are presented in Figure 11, and the strategy for synthesis and processing technologies applied in order to develop biomaterials for the use in biomedical fields is presented in Figure 12.

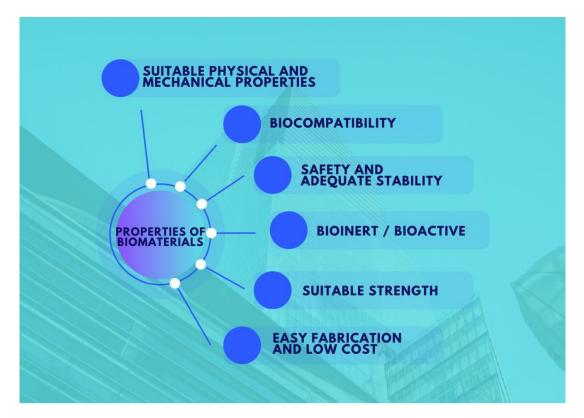


Figure 11. Required properties of biomaterials to be use in biomedical applications.

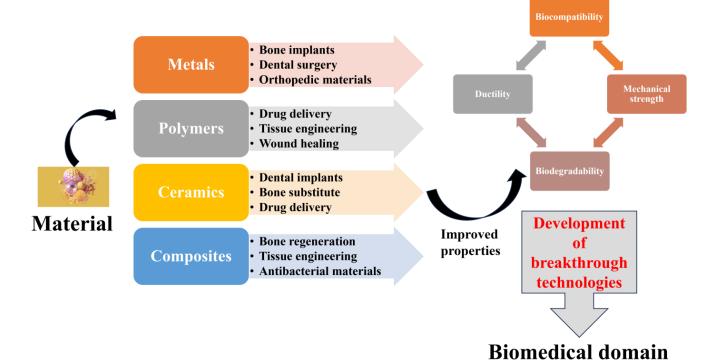


Figure 12. Biomedical applications of biomaterials.

5. Conclusions and Future Perspectives

Progress in biomaterials will comprise the advance of supplementary functional medical biomaterials and the extended usage of biomaterials in the novel areas of their application. Nevertheless, the future may also suggest a prospect for researchers in the field to reconsider essentially the method in which motivation is drawn from natural science. Consideration of the natural mechanisms of complex dynamic performances of materials in their medium may lead to the design/synthesis of innovative materials that replicates the nature by reproducing the functional comportment of these biomaterials in order to acquire new properties that are presently unreachable. The future research will be focused on a new generation of bioinspired "smart", multifunctional biomaterials for monitoring health and for preventing biological crises. A serious rational stage in biomaterial design and synthesis will be their recognition as models or templates of biopolymers and organisms for multifunctional, active devices. The economic and social impact will be diminished by reducing the costs related to the manufacturing process of existing chemicals and drugs. This will need the progress and application of new methods from different areas such as engineering, biology and the physical and chemical sciences.

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