Aerogel-Based Materials for Biomedical Applications

Subjects: Others

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Aerogel is one of the most interesting materials globally. The network of aerogel consists of pores with nanometer widths, which leads to a variety of functional properties and broad applications. Aerogel is categorized as inorganic, organic, carbon, and biopolymers, and can be modified by the addition of advanced materials and nanofillers.

aerogel silica biopolymer biomedical application wound healing drug delivery

1. Introduction

Aerogel is a nanostructured material that is gaining popularity as a structural alternative for insulation in a variety of uses, ranging from residences and commercial structures to offshore platforms and spacecraft. Aerogel insulator is thought to provide 40 times the shielding effect of fiber glass, allowing it to be used in space-constrained applications. It is a low-density, high dielectric strength, high specific surface areas, low thermal conductivities, and extremely porous foam with interconnected nanostructures [1][2]. Aerogel is composed of approximately 99.8 percent space, giving it a spectral look, and garnering the name of ‘solid smoke’ [3]. It is typically composed of silica and may take numerous shapes. However, organic polymers, inorganic, carbon allotropes, polysaccharides, transition metals, and nanostructures of semiconductors may also synthesize aerogels [4]. Aerogel is created by drying gels at extremely elevated heat.

In the early 1930s, Kistler and Learned invented the first aerogel by supercritical drying a wet gel and extracting the liquid [5]. It was employed as a tobacco filler and thickener, whereas silica aerogel was used as a thermal insulating blanket. Despite the numerous benefits that silica as well as other inorganic compounds can bring in the production of aerogel, conventional aerogel raw resources are still derived from petrochemical sources. On the other hand, the difficult multistage preparation method stymied the development of aerogel. Nonetheless, native aerogel with a single element is typically afflicted by serious issues such as weak mechanical properties, and a lack of functionalities. The name “aerogel” resurfaced in the 1970s, with the rising use of sol–gel synthesis processes and the usage of aerogel to store rocket fuels [6]. Following that, important efforts were made to simplify the synthesis methods, particularly drying to achieve a low-cost and simple synthesis of aerogel. This paved the way for a wide range of aerogel to be used in various fields of application due to their open structure and lightweight [5][7][8]. To improve aerogel performance, significant growth in the emergence of future aerogel with varied physicochemical features and functional abilities is required [9][10]. For example, aerogel-based biomaterials are now made from a variety of sources or components that imitate the structure of a biological extracellular matrix. The tissues that surround this structure serve as support cells and are affected biochemically by it. Even though an aerogel network has also hybridized with a wide variety of nanostructures and improved functional properties such as antifungal or antimicrobial performance.

2. Type of Aerogels and Properties

Different varieties of aerogels were produced during the last few decades as the methods for the synthesis and drying of aerogels improved. They can be classified as inorganic aerogels (silica, alumina, and titania), polymer-based, carbon allotropes (nanotubes and graphene), and natural macromolecule-based aerogels (alginate, starch, gelatin, protein, nanocellulose and chitosan) [11][12]. Typically, silica-based aerogels are the most potential candidate materials owing to their distinctive characteristics, such as low thermal conductivity (15–20 W/mK), low density (0.003–0.5 g/cm^3), and large surface area [13]. They are generally fragile, have poor mechanical properties, and require a lengthy processing technique, hence limiting their application range [10]. Many attempts to increase the quality of silica-based aerogels have already been made, including using (i) adaptable silica catalysts in the strand, (ii) enhanced polymer cross-linking, (iii) accelerated ageing processes in different solutions, (iv) adding nanofillers, and (v) polymerizing the precursor in advance of gelation. For example, it has been shown that the combination of silica with methacrylate polymer to improve the polymerization resulted in enhanced mechanical performance and other parameters, including densities, areas, pore diameters, and void content [14]. Silica aerogels through polymer modification are illustrated in **Figure 1**. They are classified as silica aerogels reinforced polymer, fabricated via cross-linked via water-oil aqueous solution in high-internal stage emulsion substance. This novel material shows a superior performance property over pure silica aerogels [15]. In addition, Posada et al. produced ceria-containing silica aerogel via a three-way catalyst approach in incorporation with a new rapid supercritical separation method. They employed a polyether to strengthen the aging process and accelerate the gelation time [16]. This innovative technique can reduce the time taken to prepare wet gels, including gelation, ageing, and solvent exchange from days to seconds [17].

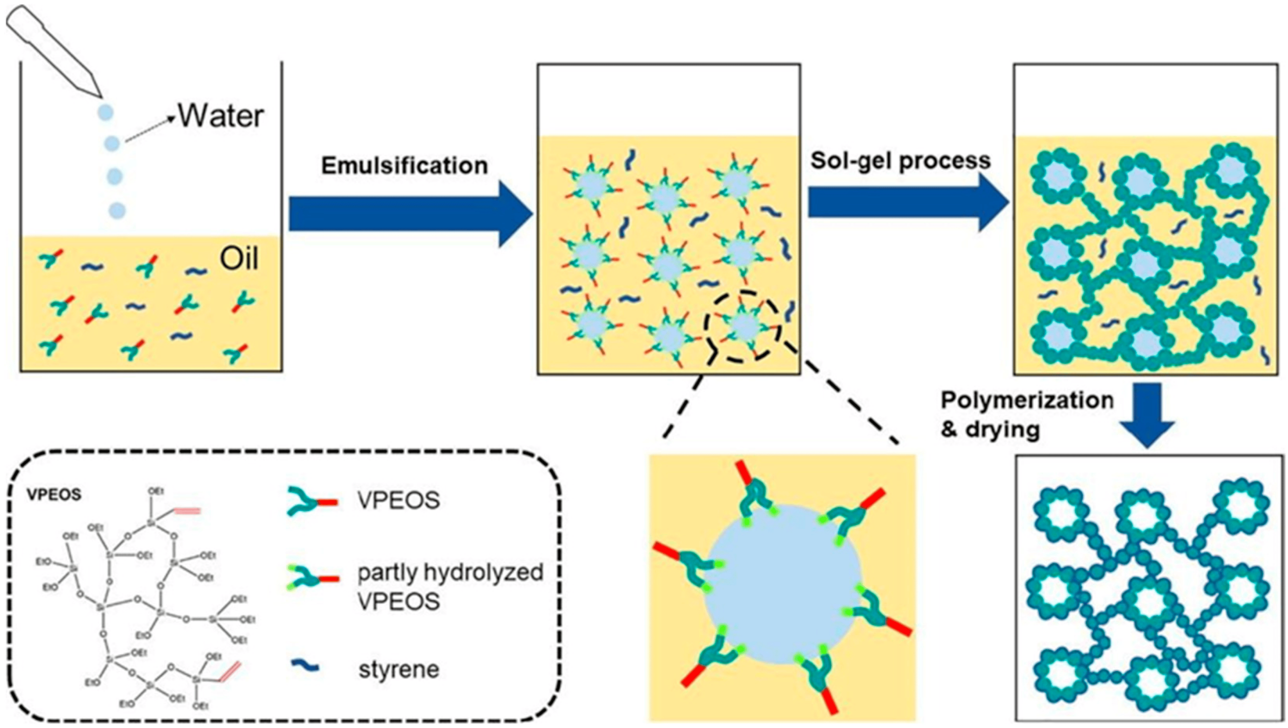

Figure 1. Polymer modification via polymerization of water-in-oil HIPE templates [15].

In addition, a nanofiller such as graphene nanoplatelets (GnPs) can also be employed to enhance the mechanical behavior of aerogel. This GnPs can speed up the gelatinization of nanostructures and reduce nanopore shrinkage throughout the hydrothermal process [18]. In addition, many studies are concentrating on improving the performance of silica aerogel by utilizing various approaches in native silica aerogel. A trifunction alorganoalkoxysilane, such as methyltrimethoxysilane was also used to provide agility to silica aerogel. However, the high costs of these precursors make them unsuitable for long-term use. As a result, many researchers adopted the organic-inorganic hybridization method, which entails cross-linking the silica aerogel with organic molecules [19]. This distinctive aerogel has a high degree of hydrophobicity and thermal insulation, giving it appealing properties such as self-cleaning, infrared stealth, and heat insulation compared with rival commercial items. The cellular structure shown the construction of multidimensional nanomaterials with synergistic action of organic–inorganic components contributed to the excellent multifunction of aerogel [20][21] and a strong interfacial effect is formed between the two components [22]. In general, other inorganic aerogels, such as alumina and titania, have garnered huge attention due to their unique microstructures. However, the extreme brittleness and manufacturing expense of these aerogels severely limit their industrial advantages. These aerogels may be modified with other materials, such as organic and polymer substances to provide numerous meshwork formation, high porosity, lightweight structure, moduli of elasticity, and low thermal conductivity [23][24][25]. Multifunctional inorganic aerogel with high open porosity and enormous surface area is a promising material that might be extended for extensive applications [26][27]. Additionally, the agglomeration of inorganic nanoparticles and nanofibers are recognized as a very viable approach for creating extremely flexible, readily accessible, and versatile composite aerogels [22].

Furthermore, polymer aerogels have a variety of forms, including polyamide (PI), polyvinylpolydimethylsiloxane (PDMS), and phenolic-based aerogels. All polymer aerogels have closely similar structures and properties [28][29][30]. In contrast to silica aerogel which are fragile and hygroscopic, aerogels derived from polymers have a broad variety of uses owing to their excellent mechanical attributes, such as high strength and fatigue resistance. These organic aerogels have thermal conductivity close to silica aerogel, comparable density and can be produced with very little shrinkage during the manufacturing process. Depending on the polymer type and fabrication circumstances, it may range between sheet-like skeletons and colloidal nanoparticles to nano/micro-fibrillar networks. The structural properties of aerogel materials, such as shape, size, and even pore ordering, have a substantial impact on their ultimate mechanical performance [31]. For example, a PI reinforced graphene oxide/cobalt (PI/rGO/Co) polymer produced by a unique cross-linking process demonstrated great heat stability and low thermal conductivity [32]. Additionally, multifunctional polyvinylpolydimethylsiloxane (PDMS)-based aerogels were reported to have high hydrophobicity and super-flexibility, thermal superinsulation, effective water, and oil separation, integrate selective absorption, and strain sensing [33]. In contrast, cellulose-based aerogel offer high porosity, higher surface area, and lightweight [34]. Aerogels containing organic precursors such as resorcinol formaldehyde, phenol formaldehyde, or melamine formaldehyde, on the other hand, have extremely poor electrical conductivities and dramatically lowered heat transmission throughout the aerogel’s backbone phase. Compared with cellulose-based aerogels, they may also be mechanically more flexible and confined to surface areas of less than 1000 m^2/g [35].

Meanwhile, carbon allotrope aerogel is generally porous materials made up of small interstitial pores (less than 50 nm) and interconnected with homogeneous carbonaceous particles (3 nm–30 nm) [36]. This aerogel has strong thermal and electrical conductivity. It provide a more brittle structure with higher backbone porosity due to micropore structures at specific areas of approximately 2000 m^2/g for certain meso- and macrostructures [37]. The typical synthesis process of polymer or carbon aerogel is illustrated in **Figure 2**.

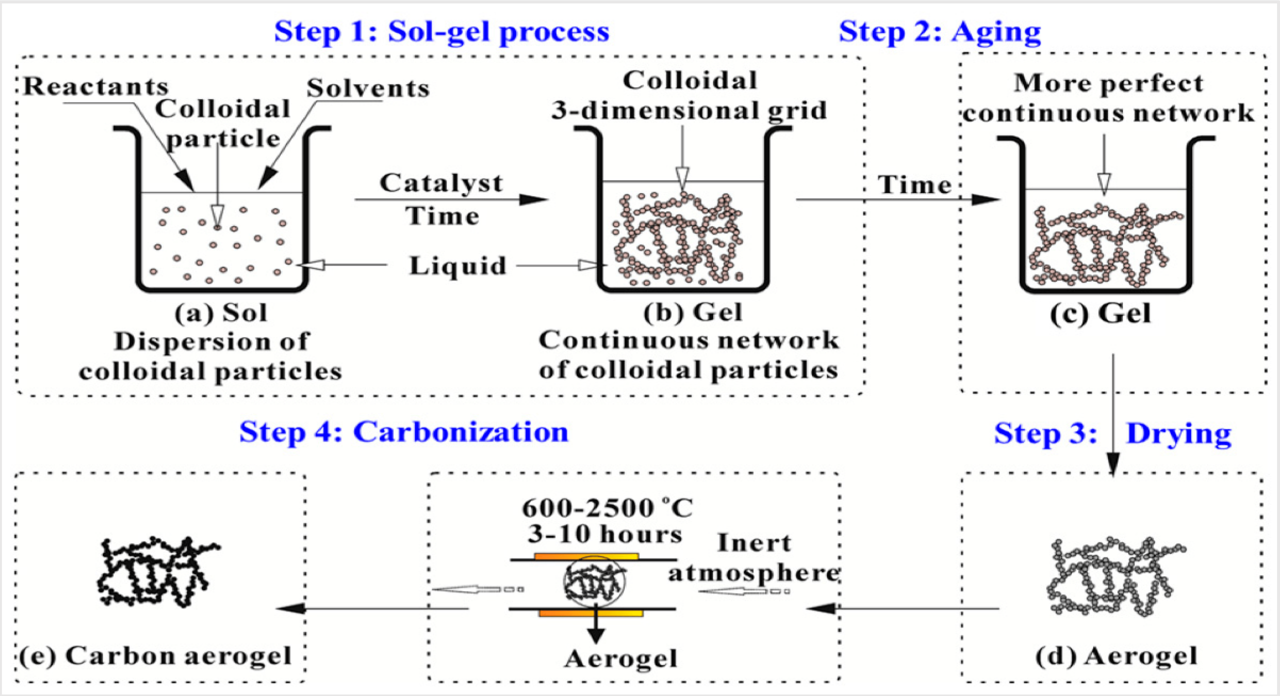

Figure 2. Basic method of producing carbon or polymer aerogels [31].

Macromolecules or polysaccharides-based aerogels are made from biopolymers derived from renewable raw materials such as cellulose, chitosan, alginate, chitin, and protein. For example, cellulose aerogel is identical to ordinary silica and polymeric aerogel in terms of compressive stress (5.2 kPa–16.67 MPa) and better recyclability [7]. As stated by Gong et al. the spongy morphology of this aerogel was steadily enhanced with the raising of the carboxyl proportion of nanofibrils in the structure. Carboxymethyl element could also effectively increase the total area of aerogel, due to the elimination of horrification [38]. Moreover, chitosan-based aerogel has much better physicochemical properties of the functional groups than cellulose-based aerogel and can be used in biomedical applications. When it was incorporated with graphene oxide, the adsorption capacity of this material improved [39]. In contrast, alginate-based aerogel is highly promising for low-flammability performance; however, it exhibits poor mechanical properties [40]. Interestingly, with the addition of graphene oxide, the catalytic property of this biomass aerogel can be increased by 30 times, resulting in an improvement in its mechanical property. The properties of different types of aerogels are shown in **Table 1**.

Table 1. Different types of aerogels with their respective properties.

Types	Main Component	Properties	Weakness	Methods for Improvement	Applications	References
Silica	Tetraethylorthosilicate (TEOS) and methyltrimethoxysilane (MTMS)	Low heat conductivity, large built-up area, low density	Fragile, have poor mechanical properties and require a lengthy processing technique	Use precursors in the backbone, surface-crosslinking with a polymer, prolonged aging incorporating, polymerizing	Photocatalysts, Thermal insulation, absorbent pollutants	[13][41]
Polymer	Cellulose/ conducting polymer	High moduli and fatigue resistance	Monolithic, prone to defects, length processing and costly	Usage of synthetic polymer	Additives (foods, cosmetics) construction, materials, drug delivery carrier	[28][42]
Carbon	Carbon/CNT/ graphene	High specific surface area and porosity, low density, good electrical conductor, good chemical	Low electrical conductivities and reduced heat transmission via the	Focused on carbon aerogel-based biomass	Electrodes, in supercapacitors, adsorbents for phenol	[37][43]

Types	Main Component	Properties	Weakness	Methods for Improvement	Applications	References
		stability, and hydrophobicity,	aerogel backbone phase with related organic precursor			
Inorganic	Oxide/ metallic/ chalcogenide	Ultra-high surface area and high open porosity	High production cost	Hybrid aerogel formation	Energy conversion, storage application	[44][45]
Organic	Biopolymer	High compressive strength, high surface area	Poor mechanical properties	Incorporated with inorganic fillers	Biosensor, Medical implantable device.	[46]

3. Biomedical Applications of Aerogel

Aerogel is an appealing substance for the biomedical field due to their distinctive properties, which include low density, porous structure, extensive surface area, and high strength. It is also versatile in terms of sol–gel biochemistry. Aerogel typically used in biomedicine to encapsulate bioactive compounds with low solubility or stability as well as to create artificial scaffolds for tissue engineering and materials for chronic wound dressings. Other than that, many studies also discussed the applications of aerogel, such as for drug delivery carriers, anti-toxicity, and antioxidants. Although the technology and composition of aerogels are varied, the aerogels applied in the biological system must be made of biocompatible, and preferably biodegradable material.

Biocompatibility is the ability of materials to be functional in a biological system without causing harm. Biocompatibility must be determined before any substance can be used in biological applications. **Table 2** summarises biocompatibility testing of various materials using in vitro and in vivo methods. Bajpai et al. studied the biocompatibility of 3D-structure graphene aerogel (GA). This 3D, ultra-lightweight and hydrophobic GA was produced by the one-step pyrolysis of sugar and ammonium chloride. GA showed excellent adsorption capacity for various biogenic amines, bacteria contaminants such as Staphylococcus aureus, and other toxins, especially in food safety applications. The biocompatibility of the synthesised GA is also determined by cell proliferation efficiency and wound healing ability [47]. In another work, Liu et al. created an extremely porous aerogel consisting of graphene oxide (GO) and Type I collagen (COL) using the sol–gel approach. This study demonstrated that 0.1% GO-COL aerogel had good biocompatibility in vivo, making it a potential scaffold to support bone regeneration and tissue engineering [48]. Zhou et al. found that the combination of hydroxyapatite and graphene to produce a new aerogel improved microbial electrocatalysis due to its higher interfacial and biocompatibility for bacterial growth [49].

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Other biomaterials, such as bacterial cellulose (BC) also had excellent biocompatibility and had a non-immunogenic potential [50]. BC aerogel is known for being fragile, very light, open-pored and transversally isotropic materials for various biomedical applications. Salehi et al. put clay nanoplatelets over the BC membrane to form a nano-fibrillated template for aniline in situ polymerization, creating a double linked network of electrically conductive pathways in the aerogel. Clay and polyaniline had a synergistic effect on biocompatibility and cell adhesion, with no mutagenic or carcinogenic effects [51]. In another study, a novel biocompatible BC aerogel modified with poly (glycidyl methacrylate) (PGMA) was fabricated. The incorporation of PGMA and BC aerogel improved its biocompatibility following the immobilisation of catalase [52]. BC aerogel had the highest modulus, porosity and specific surface area among cellulose aerogels. Even so, the production of BC was hindered by a lengthy production period, a low yield, and a high price, which diminished interest in its further clinical applications.

Another biomaterial with exceptional properties and being more biocompatible within cells is silica aerogel. Their main limitations however, are fragility and high hygroscopicity [53]. In a study by Lazar et al., silica aerogel was hybridised with industrial manufactured bovine casein, using tetramethyl orthosilicate (TMOS) and co-gelation. The CHO-K1 Chinese hamster ovary cell line was used to test the in vitro biocompatibility of hybrid aerogel. It has been demonstrated that silica-casein aerogel are highly biocompatible and, to all intents and purposes, non-toxic to CHO-K1 cells [54]. According to the findings by Sani et al., the hydroxyapatite (HA)-mixed with silica aerogel with a weight ratio of 0.5 had the highest bioactivity and biocompatibility [55]. Resveratrol has been thought to help with or even cure osteoarthritis. Qin et al. synthesized a resveratrol-loaded silica aerogel (RSA) using the sol–gel method and exploited it as a drug delivery vehicle. According to the results of the study, RSA is inexpensive, biocompatible, and has relatively high loading rate of 19%. Initial in vitro toxicity testing revealed that RSA is biocompatible stable, and may be used to treat osteoarthritis due to its anti-inflammatory effects [56]. In another report, nanofibrous silica hybrid aerogel was biocompatible to healthy cells but their antitumour activity significantly increased when loaded with camptothecin (CPT) [57]. Kiraly et al. in their study injected a fluorescein-labeled silica-gelatin aerogel microparticles (FSGM) into the peritoneum of mice to assess acute toxicity. They reported no physiological abnormalities or disorder were discovered after a three-week-long experiment [58]. A nano-porous silica aerogel was developed for drug delivery for oral administration of paclitaxel (PTX), an anticancer drug. The excellent biocompatibility of this aerogel was proven by the reduced side effects of drug and inhibited tumour growth [59]. Furthermore, silica–gelatin hybrid aerogel has a potential for local and non-invasive drug delivery because they are biodegradable and biocompatible within tissue cells [60]. In addition, aerogels produce from marine polymer such as chitosan exhibit a potential prospect in wound healing due to their antimicrobial activity. Piatkowski et al. developed a new chitosan-based aerogel with enhanced properties to improve the healing of burn wounds. The studies demonstrated that the proposed chitosan aerogel containing Au nanoparticles were biocompatible and promoted fibroblast proliferation [61]. Batista et al. developed a hybrid alginate-chitosan aerogel fibre and assess their effect in wound healing application using the emulsion gelation method. In vitro model assessment revealed that they were non-cytotoxic and promoted wound healing [62]. In another study, the hybrid chitosan-alginate aerogel microparticles were also prepared using the emulsion gelation technique. The toxicity test showed that the alginate-chitosan carrier induced moderate lung inflammation along with some damage to kidneys and liver [63].

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However, conventionally prepared chitosan aerogel exhibited a number of defects, including low porosity, an irregular structure, and an easiness to deform, which limited their biocompatibility [64].

Beside chitosan, alginate is another biomaterial that has been intensively researched in biomedical fields. For example, Franco et al. used mesoglycan (MSG) and impregnated calcium alginate aerogel (CAA) to treat a wound. Both human keratinocytes and fibroblasts were resistant to the cytotoxic effects of MSG in CAA, as shown by an in vitro experiment [65]. In other applications, alginate microspheres-based on alginate and hyaluronic acid demonstrate high porosity and good in vitro aerodynamic properties [66]. Carbon-based aerogels are unique since it consists of networks of 3D nanostructures with a high volume of air-filled nano-porous, high porosity, low density, and a large surface area [67]. These properties endow aerogels with a rapid response signal, high selectivity, and super sensitivity for sensing a variety of biomedical targets. The synthesis of carbon-aerogel scaffolds containing biocompatible ceramic nanoparticles of tricalcium phosphate has been disclosed by Tevlek et al. Due to their high gelatine content and highly porous structure, the materials exhibited good biocompatibility and supported cell growth for 14 days [68].

Table 2. Summary of studies on compatibility of aerogels for biomedical applications.

	Aerogels	Method	Remarks	References	Year
1	Cellulose	Freeze drying and polymerization	Higher biocompatible with catalase immobilization	[52]	2019
2	Silica	Freeze-drying	Biocompatible for drug carrier	[59]	2019
3	Graphene oxide-collagen	Sol–gel process	0.1% GO-COL aerogel presented reliable biocompatibility	[48]	2019
4	Graphene	Pyrolysis	Cell viability was observed even at high concentrations	[47]	2019
5	Chitin	Supercritical CO2 drying and freeze-drying	Good biocompatibility (cell viability >90%	[69]	2019
6	Alginate-chitosan	Supercritical drying of CO_2	Cell viability values >70 %	[62]	2020
7	Alginate-Chitosan	Emulsion gelation	Resulted in mild lung-congestion	[63]	2020
8	Silica	Aqueous sol–gel ambient pressure drying	Not toxic to normal human osteoblast cell line	[55]	2020
9	Silica	Co-gelation in the sol–gel, supercritical CO_2	Highly biocompatible and practically inert towards CHO-K1 cells	[54]	2020

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	Aerogels	Method	Remarks	References	Year
10	Silica	Sol–gel	Good biocompatibility	[56]	2020
11	Silica	Freeze-drying and cross-linking	Excellent biocompatibility to human cells	[57]	2020
12	Carbon	Freeze-drying	Cells able to adapt to microenvironment and able for growth	[68]	2020
13	Composite	Freeze-drying	Good biocompatibility of mouse lung fibroblasts (L929) cells on the membrane	[51]	2021
14	Silica	Sol–gel combined with co-gelation	All mice were healthy after being injected with aerogel	[58]	2021
15	Graphene	Hydrothermal thermal dialysis and freeze-drying	Excellent biocompatibility	[49]	2021
16	Chitin	Supercritical CO_2 drying	Lower haemolysis ratio (<1%)	[70]	2019
17	Alginate	Supercritical CO_2 drying	Not cytotoxic	[65]	2020
18	Cellulose	Supercritical CO_2 drying	Excellent conditions for cell viability and proliferation	[71]	2021
19	Magnetic	Sol–gel	Biocompatible	[72]	2022
20	Silica	Sol–gel, supercritical drying	Biocompatible for local and non-invasive drug delivery	[60]	2022

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