

Magnetic Bacterial Cellulose Biopolymers

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Bacterial cellulose (BC) is a biopolymer that has been widely investigated due to its useful characteristics, such as nanometric structure, simple production and biocompatibility, enabling the creation of novel materials made from additive BC in situ and/or ex situ.

electronics

devices

cellulose

1. Introduction

Magnetic materials are found in various types of devices with applications in everyday life such as sensors, transformers, magnets, stereos, electronic circuits, data storage systems, etc. [1]. Depending on the desired application, these materials must also have different properties such as biocompatibility for use in medical and pharmaceutical applications, and flexibility, as in the case of malleable sensors [2][3][4]. Several researchers have produced biotechnological magnetic materials, also classified as smart materials [5].

2. Magnetic Particles Added to Bacterial Cellulose

There are several options for dopants with magnetic properties. Iron oxides and derivatives such as magnetite, maghemite, hematite and ferrites can be found in nature as ores and can also be synthesized in the laboratory [6][7]. **Table 1** presents some studies that produced magnetic cellulose biomembranes, listing the respective magnetic dopants and applications.

Table 1. Magnetic materials added to bacterial cellulose and respective applications.

Magnetic Dopants	Forms of BC Magnetic Doping	Saturation Magnetization of Magnetic BC (emu/g) *	Coercive Field of Magnetic BC (Oe) *	Applications	References
Magnetite	In situ co-precipitation	60.0	15	Application in nonlinear optics, clinical applications such as contrast, agents for magnetic resonance,	[8]

Magnetic Dopants	Forms of BC Magnetic Doping	Saturation Magnetization of Magnetic BC (emu/g) *	Coercive Field of Magnetic BC (Oe) *	Applications	References
				hyperthermia and cell separation and sensors.	
	Incorporation of previously made particles	41.0	27	Absorption of heavy metals	[9]
	Incorporation of previously made particles	53.6	**	Actuators, sensors, flexible data storage	[10]
	In situ electrolysis	4.2–21.2	**	Electronic and magnetic devices, enzymatic assays, drug delivery systems	[11]
	In situ co-precipitation	***	***	Tissue reconstruction	[12]
	In situ co-precipitation	23.63	0.042	Drug delivery	[13]
	In situ co-precipitation	5.14–11.56	**	Electronic devices	[14]
	In situ co-precipitation	40.57	**	Electronic devices	[15]
	Thermal decomposition	***	***	Magnetic resonance device	[16]
	In situ co-precipitation	34.07	**	Device for enzyme immobilization	[17]
	Incorporation of previously made particles	***	***	Drug delivery	[18]
	Incorporation of previously made particles	0.14	**	Optical materials	[19]
Cobalt ferrite	In situ co-precipitation	3.769–5.026	5000	Electric actuators	[20]
				Sensors	[21]
Maghemite	Thermal decomposition	60	**	Sound amplifier devices	[22]

Magnetic Dopants	Forms of BC Magnetic Doping	Saturation Magnetization of Magnetic BC (emu/g) *	Coercive Field of Magnetic BC (Oe) *	Applications	References
Barium ferrite	Incorporation of previously made particles	24.1–49.5 [9][10][13][14][15]	5.31	Data storage devices, electromagnetic adsorbers	[23]
Magnetite and maghemite	In situ co-precipitation	55.0–61.0	**	Sensors, actuators, and metal adsorbents	[24][27]
Manganese ferrite	In situ co-precipitation	***	***	Sensors [25][28]	[25]
Nickel nanoparticles	In situ co-precipitation	2.8–3	28	Magnetic ink and magnetic scaffolds for tissue engineering	[26]

saturation magnetization of these minerals are generally high but vary depending on the size of the crystals. One of the challenges in the synthesis of these materials is the ease of oxidation, as the particles can be oxidized with atmospheric air. For instance, one of these oxidative processes is the formation of maghemite during the synthesis of magnetite due to the oxidation of the latter during the process. It is therefore common to obtain both types of particles in certain experiments [7][29].

* Measurements at room temperature. ** Values near to zero. *** The authors did not measure the values in such referenced articles.

Another interesting feature is that when these particles are in specific nanometric sizes, forming a magnetic monodomain, they change their magnetic property to superparamagnetic, which is a characteristic that makes the material magnetize only in the presence of an external magnetic field [30].

Magnetic ferrites are derived from iron oxides associated with ions of other metals. The production of this type of material takes place at high temperatures. The most common metals are barium, cobalt, manganese, nickel, and strontium [6][31]. Ferrites can be classified according to their magnetic coercivity, i.e., ease of magnetization and demagnetization, as soft (those with greater ease of magnetization due to an external magnetic field and demagnetization in its absence) or hard (those that do not lose their magnetization and are permanent magnets) [32]. Examples of soft ferrites are manganese and nickel, whereas strontium, cobalt and barium are hard ferrites [32][33][34].

In general, particles of maghemite, magnetite and ferrites used as magnetic dopants for BC have a size on the nanometric order to ensure a better interaction with the nanometric fibers of the biopolymer. Thus, a biotechnological and magnetic BC material is produced. There are also several ways of inserting these dopants into BC fibers, such as methods that carry out the synthesis of dopants in situ and ex situ, which will be discussed in the next section.

2. Forms of Bacterial Cellulose Magnetic Doping

Table 1 demonstrates the recurrence of methods used among the cited works. The method most used is in situ co-precipitation, followed by the incorporation of previously made particles, thermal decomposition and in situ electrolysis.

With the in situ co-precipitation process, precursor compounds are adsorbed by the BC and other agents are then added, thus transforming it into a small bioreactor: a medium where the synthesis of the dopant takes place. At the end of the synthesis, the magnetic particles precipitate inside the BC nanofibers [35].

Chanthiwong et al. [24] used co-precipitation to produce magnetic nanoparticles within BC fibres. The authors left the membrane immersed in an aqueous solution containing Fe^{2+} and Fe^{3+} ions. The membrane absorbed the ions and was then immersed in an oxidizing solution, converting the metallic ions into Fe_3O_4 and Fe_2O_3 (magnetite and maghemite, respectively). Another interesting point was presented in the work by Vitta et al. [26], who used BC in its coconut gel cube form, which is a sweet commonly consumed in Asia made from the fermentation of biocellulose with coconut water, demonstrating a little more of the versatility of this material. Other works describe addition via in situ co-precipitation [8][13][14][15][17][20][21][25].

Zeng et al. [22] and Mira-Cuenca et al. [16] performed the synthesis of magnetic particles in situ using a microwave device to perform a synthesis of thermal decomposition within the membranes. Zhou et al. [11] synthesized magnetite via electrolysis within BC membranes.

Thus, several methods are available for incorporating magnetic particles into a BC matrix, which can guide other researchers who intend to obtain this type of material with different techniques and compounds capable of adapting to different contexts and goals.

Another common form of incorporation is the addition of ready-made magnetic compounds synthesized ex situ, which is described in several works [9][10][18][19][23]. For instance, Sriplai et al. [10] incorporated magnetite in BC in a very simple way. The authors immersed BC films in aqueous solutions with different proportions of a commercial ferrofluid and maintained the BC submerged in the solutions for 1 h under agitation at 80 °C to facilitate the dispersion of the magnetic particles over the membrane. At the end of the process, the authors obtained uniform magnetic films. While co-precipitation methods depend not only on the absorption of particles and reagents in the BC, but also on the synthesis of the magnetic dopant, this type of method generally depends on the absorption capacity, since the particles have previously been made.

As shown in **Table 1**, most researchers opted for the synthesis of co-precipitation of magnetic particles within the structure of the biomembrane, making it a bioreactor. According to Chanthiwong et al. [24], this method became popular due to its simplicity, the good distribution of particles in the BC and the possibility of adjusting the reagents. However, not all studies use this method. Salidkul et al. [23], for example, performed ex situ doping with barium ferrite. According to the authors, there is no way to carry out the safe in situ precipitation in BC, as the synthesis of these nanoparticles occurs at very high temperatures (>800 °C), which would degrade the BC membranes. There is also a discussion of the control of the amount, distribution, size, and shape of the dopant particles added to the

BC in in situ and ex situ precipitation methods. Some authors claim that classic in situ co-precipitation does not enable good control over these variables, whereas others suggest adaptations and adjustments of the reaction conditions to obtain greater control over the particles [13][15][26]. Therefore, the choice of doping method depends on factors such as the nature of the dopant, degree of complexity, availability of materials and the desired application of the biomaterial.

3. Applications

One of the main advantages of magnetic cellulose (BC and VC) production is the diversity of cellulosic fibres and dopant particles that can be added [36][37]. This wide variety of options enables the production of different biotechnological materials, which can assume different shapes and applications. There are several suggestions and applications for magnetic materials derived from BC biofilms, as presented in **Table 1**. Most applications are in the field of electronics and materials used in medical devices, due both to the characteristics of the magnetic material incorporated and the intrinsic properties of BC [38].

There are reports of applications of magnetic BC in several fields, such as in the manufacture of enzyme and protein immobilization systems, application in the food industry, as adsorbent material and for the separation of heavy metals [9][17][23][24]. The literature indicates that, in addition to a wide variety of synthesis methods, there is also a diversity of potential applications for magnetic BC biomembranes, making this a promising material for further research and investigation [39][40][41].

Interest in the medical and pharmaceutical applications of BC is generally due to its biocompatibility [42]. By conferring magnetic characteristics to the biopolymer, a biocompatible, magnetic material is produced, which can then be used in the manufacture of various systems and devices such as systems for contrast in magnetic resonance imaging exams, tissue engineering and drug delivery [43][44]. Another important point of doping with magnetic nanoparticles, especially magnetite, is the antimicrobial effect on different viruses, bacteria, and fungi [13].

Mira-Cuenca et al. [16] developed an ink with crushed BC fibers and iron oxide nanoparticles (magnetite). With this ink, the authors demonstrated that it was possible to produce different types of drawings in the form of a film. In in vivo tests involving a muscle implant, the ink was easily identified through magnetic resonance imaging. Thus, the material performed very well as a transverse relaxation contrast agent, enabling the monitoring of surgical implants containing this dye in a less invasive way.

Another example of a medical application is found in the work by Chaabane et al. [13]. The authors modified the structure of BC and carried out magnetite precipitation in situ, forming a magnetic BC composite. The antimicrobial properties were evaluated, and the material was also used as a chemotherapeutic agent in mice with tumours. The authors observed the effectiveness of the magnetic BC composite as an antifungal and antibacterial agent and found that the composite performed well in chemotherapy treatment in mice, preventing tumour growth.

Sensors for monitoring health conditions have been also studied. In this type of application, there is interdisciplinarity with the field of electronics, which is constantly growing and directly and indirectly impacts many other fields.

4. Electro-Electronic Applications of Magnetic Bacterial Cellulose

Electro-electronics is a constantly growing field, as other scientific fields depend on electronic devices. Such devices are also found in homes and industries, making everyone's work and life easier. Magnetic BC also has applications in this field, since many devices have parts made of magnetic materials and there is a continuous search for their optimization. Therefore, magnetic BC has been attracting the attention of researchers ^[45]. Indeed, the literature reports the association between magnetic BC biofilm and electronic devices such as actuators, sensors, memory storage devices, sound amplifiers and displays ^{[19][22][38][40]}.

The study by Chen et al. ^[21] is a good example of a practical application of magnetic BC with the development of an electromagnetic motion sensor. The sensor was composed of a flexible magnetic BC tape connected to a copper coil. The ribbon was sewn onto the sleeve of a jacket, while the bobbin was sewn to the waistband. The sensor was able to monitor the shape of the movement by detecting the oscillation frequency and speed of the arm, which was in direct contact. The volunteers in this experiment ran at different speeds. The periodicity of the arm movement generated displacement of the magnetic BC tape and an oscillation in its magnetic field, thus inducing a voltage difference between the tape and copper coil. The signals were captured in waveforms by a monitor connected to the copper coil and had different amplitudes and frequencies, corresponding to each volunteer's running speed, showing the performance of magnetic BC as a sensor. In addition to the performance shown in the prototype, the authors also listed the flexibility and biocompatibility of the magnetic BC as advantages of the material in the application of sensors of this type ^[46].

Sriplai et al. ^[25] and Marins et al. ^[8] also proposed the application of magnetic BC in the production of sensors. Nakajima et al. ^[47] stated that magnetic characteristics and flexibility are important for the applicability of modern sensors, since flexibility gives devices freer, more adaptable forms. Sriplai et al. ^[35] pointed out the piezoelectric properties (ability to generate electrical voltage in response to a mechanical stimulus) in magnetic BC films, which were conferred by manganese ferrites for the development of a greater variety of sensors.

Salidkul et al. ^[23] recommended the application of a magnetic BC produced in their study in data storage devices due to its high magnetic coercivity. Zeng et al. ^[22] highlighted the use of the magnetic membrane in electrical circuits and in the assembly of sound amplifiers, since magnetic BC showed good flexibility and magnetization in this research. The authors also relate this last possibility of application to a work carried out by Galland et al. ^[48], who produced a prototype with a magnetic VC membrane, which dispensed with the presence of a magnet inside. Another work in which the authors built an amplifier with a material similar to magnetic BC, supporting the application proposed by Wu et al. ^[49], was that of Tarrés et al. ^[50], who performed treatment on a VC extract, making its structure nanometric, which was then magnetized with magnetite.

With the great diversity of possible direct applications of magnetic BC according to its characteristics, performance in prototypes and associated with prototypes made with similar materials, magnetic BC has considerable biotechnological potential for the construction of materials and electronic devices, and should receive more attention from researchers regarding practical applications and advances in making more prototypes.

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