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Review

Overview of bacterial cellulose composites: A multipurpose advanced material

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ABSTRACT

Bacterial cellulose (BC) has received substantial interest owing to its unique structural features and impressive physico-mechanical properties. BC has a variety of applications in biomedical fields, including use as biomaterial for artificial skin, artificial blood vessels, vascular grafts, scaffolds for tissue engineering, and wound dressing. However, pristine BC lacks certain properties, which limits its applications in various fields; therefore, synthesis of BC composites has been conducted to address these limitations. A variety of BC composite synthetic strategies have been developed based on the nature and relevant applications of the combined materials. BC composites are primarily synthesized through in situ addition of reinforcement materials to BC synthetic media or the ex situ penetration of such materials into BC microfibrils. Polymer blending and solution mixing are less frequently used synthetic approaches. BC composites have been synthesized using numerous materials ranging from organic polymers to inorganic nanoparticles. In medical fields, these composites are used for tissue regeneration, healing of deep wounds, enzyme immobilization, and synthesis of medical devices that could replace cardiovascular and other connective tissues. Various electrical products, including biosensors, biocatalysts, E-papers, display devices, electrical instruments, and optoelectronic devices, are prepared from BC composites with conductive materials. In this review, we compiled various synthetic approaches for BC composite synthesis, classes of BC composites, and applications of BC composites. This study will increase interest in BC composites and the development of new ideas in this field.

Contents

Abbreviations: AAc, acrylic acid; BC, bacterial cellulose; Ch, chitosan; CNTs, carbon nanotubes; COL, collagen; EFM, electric force microscopy; FeO, iron oxide; FE-SEM, field emission scanning electron microscopy; FRP, fiber reinforced polymer; GO, graphene oxide; HA, hydroxyapatite; HRP, horseradish peroxidase; MIP, molecularly imprinted polymers; MMT, montmorrilonite; NMMO, N-methy morpholine N-oxide; NPs, nanoparticles; OLED, organic light emitting diode; PAni, polyanaline; PBH, poly-3-hydroxybutyrate; Pd, palladium; Pt, platinum; PVA, polyvinyl alcohol; TEM, transmission electron microscopy; TiO_2, titanium dioxide; XRD, X-ray diffraction; ZnO, zinc oxide.

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1. Introduction

Since its discovery, bacterial cellulose (BC) has shown tremendous potential as an effective biopolymer in various fields. The structural features of BC are far superior to those of plant cellulose, which impart it with better properties (Ul-Islam, Khan, & Park, 2012a; Ul-Islam, Khan, & Park, 2012b). Fibril networks consisting of well arranged three dimensional nanofibers enable production of BC sheets with high surface area and porosity (Ul-Islam et al., 2012a). Moreover, the crystallinity and mechanical strength of BC are higher than those of plant cellulose, which has increased its utilization in biomedical and other related fields. Specifically, BC has been applied for wound dressings, burn treatments, tissue regeneration, skin substitutes, catalyst sensing materials, and electronic devices (Ciechanska, 2004; Czaja, Krystynowicz, Bielecki, & Brown, 2006; Czaja, Young, Kawechi, & Brown, 2007). However, lack of antibacterial, antioxidant, conducting and magnetic properties has diminished its capabilities in biomedical and electronic fields (Kim et al., 2011; Maria, Santos, Oliveira, & Valle, 2010).

Polymer composites have enhanced the material and biological properties of pure polymers (Bloor, Donnelly, Hands, Laughlin, & Lussey, 2005; Islam et al., 2011; Maneerung, Tokura, & Rujiravanit, 2007; Qian, 2004). As a result, composites of BC have been synthesized to overcome its limitations and increase its applications. The biomedical efficacy of BC has been increased by synthesizing its composites using bioactive polymers, nanomaterials, and solid particles. BC composites confer antibacterial, antiviral, antifungal, biocompatible, wound healing, conducting, magnetic and optical properties to BC (Evans, O'Neill, Malyvanh, Lee, & Woodward, 2003; Kim et al., 2011; Maneerung et al., 2007; Nakayama et al., 2004; Saibuatong & Philsalaphong, 2010; Shi et al., 2012; Ul-Islam et al., 2012a). Composite synthesis technology has also advanced with the development of new techniques. Currently, BC composites are synthesized through numerous routes based on the nature and size of the reinforcement material. Among the polymers used for composite synthesis, chitosan (Ch), gelatin, and collagen (COL) have been successfully combined with BC to improve its biological properties, while graphene oxide (GO) and polyanaline (PAni) increased the conducting properties of BC (Feng, Zhang, Shen, Yoshino, & Feng, 2012; Shi et al., 2012). Numerous nanoparticles (NPs) including silver (Ag), gold (Au), palladium (Pd), iron oxide (FeO), platinum (Pt) and titanium oxide (TiO_2) have been utilized in BC composites for various applications (Evans et al., 2003; Maneerung et al., 2007; Serafica, Mormino, & Bungay, 2002; Zhang et al., 2010). For example, BC-Ag has been synthesized through numerous synthetic strategies to impart antibacterial activities to BC, which consequently improves its performance when applied for wound healing. BC-Pd, BC-Pt, BC-Au, and BC-FeO have introduced conducting properties to BC sheets that are utilized in biosensors, display devices, electronic papers, security paper, and catalysis (Evans et al., 2003; Maneerung et al., 2007; Zhang et al., 2010).

Since BC has multiple applications, the compilation of fundamental synthetic approaches for enhanced production, environmental issues and industrial needs is of the utmost importance. Moreover, the approaches employed to date to overcome the limitations associated with pristine BC must be known before proceeding toward its practical applications. Accordingly, knowledge pertaining to possible routes of synthesis of BC composites and their consequent pros and cons can lead to the advancement of these materials for specific applications. Critical literature studies conducted to date have revealed that BC composites developmental strategies are governed by the nature of reinforcement materials and the desired application.

Previous efforts identifying the shortcomings of pure BC, stepwise development of BC composites to overcome these limitations and approaches toward the practical applications of BC composites provide a platform for understanding the overall progress in this field to date. In the present review, we have summarized various efforts made to enable inexpensive and enhanced production of BC, various routes developed for synthesizing BC composites, pros and cons of available composite synthesis strategies, classes of BC composites and applications of these composites. To the best of our knowledge, no such comprehensive investigation has previously been conducted for BC composites. Accordingly, this review will provide ideas for the development of novel strategies for preparation of BC composites with specified applications.

2. Composite materials

Composites consist of two types of individual materials, the matrix and the reinforcement material. The matrix acts as a scaffold and supports the reinforcement material, while reinforcements impart physico-chemical and biological properties to the matrix. A synergistic reaction generates material properties that are not present in the individual constituent materials. The broad range of matrices and reinforcing materials allows synthesis of various composite materials with optimized properties. A number of natural and composite materials have been used throughout history; however, the first composite material produced on an industrial scale was fiberglass, which was introduced by Owens Corning in 1935. The combination of fiberglass with plastic polymer produced a much stronger structure that laid the foundation for the fiber reinforced polymer (FRP) industry.

Polymer composites have gained a great deal of attention due to their enhanced physico-mechanical properties and multiple applications relative to the virgin polymeric materials of which they are composed. Reinforcing materials of a different nature such as solid clay particles, metallic oxides, carbonates and fibrous materials have not only strengthened polymers, but also introduced various biological features to polymeric composites (Bloor et al., 2005; Islam et al., 2011; Maneerung et al., 2007; Qian, 2004; Ul-Islam, Khan, Khattak, & Park, 2013). Cellulose has been utilized as

both a reinforcing material and matrix (Ifuku et al., 2007). The well arranged fibrous network structure of cellulose can cage-in NPs, thus acting as a matrix (Ul-Islam et al., 2012a, 2012b). Similarly, cellulose nanofibers can reinforce other polymeric and non polymeric materials, cells and tissues (Entchev et al., 2004; Svensson et al., 2005). BC has similar chemistry and superior physical properties to plant cellulose and has therefore been used in the preparation of a number of composite materials for various applications.

3. Toward synthesis of BC composites

3.1. Bacterial cellulose

BC is produced from a variety of synthetic and non synthetic media by a class of acetic acid producing bacteria that includes *Acetobacter xylinus* and *Acetobacter hansenii* (Ha et al., 2008; Hestrin & Schramm, 1954; Shah, Ha, & Park, 2010; Shezad, Khan, Khan, & Park, 2009; Ul-Islam et al., 2012a). During the synthetic process, the glucose chains produced inside the bacterial body extrude out through tiny pores present on their cell envelope. The glucose chains then combine, forming microfibrils that further aggregate to form cellulose ribbons (Ross, Mayer, & Benziman, 1991). These ribbons (nanofibers) subsequently generate a web shaped network structure with plenty of empty spaces between the fibers. The well separated nano-fibrils of BC create an expanded surface area and highly porous matrix (Dahman, 2009; Ifuku et al., 2007; Maria et al., 2010; Meftahi et al., 2010). The well arranged nanofibers, pure nature, extended surface area and variable pore geometry have imparted BC with remarkable properties including high mechanical strength, improved crystallinity, high water holding capacity, slow water evaporation capability, broad chemical modifying ability, biodegradability, biocompatibility and the ability to be molded into three dimensional structures during synthesis (Jeon, Oh, Kee, & Kim, 2010; Ul-Islam et al., 2012a)

3.2. Synthetic approaches and trends in BC production

BC has been synthesized through a number of routes, which are broadly classified into static and agitated cultures. In static cultures, BC is produced at the air–water interface as an assembly of reticulated crystalline ribbons that are formed into a gel (pellicle) that increases in thickness with increasing cultivation time (Mormino & Bungay, 2003; Ul-Islam et al., 2012a). The pellicle then grows downward until all cells entrapped in the pellicle become inactive or die due to oxygen deficit (Borzani & Desouza, 1995). Medical and several other applications utilize BC sheets produced through static culture. Unfortunately, the low yield of the static production process has restricted the commercial efficacy of BC. Therefore, a major goal of BC research has been improvement of the product yield. Indeed, various efforts have been made to enhance the microbial conversion of glucose into cellulose. Some of these approaches include isolation of high rate production strains (Kojima, Seto, Tonouchi, Tsuchida, & Yoshinaga, 1997), in situ pH control (Vandamme, De Baets, Vanbaelen, Joris, & De Wulf 1998), controlled side product formation (Ha et al., 2011), supplementation with additional substrates and variation of carbon sources (Hornung, Ludwig, Gerrard, & Schmauder, 2006). Hornung et al. developed a comprehensive model by evaluating several factors including bioreaction (substrate consumption and product formation), transport of oxygen and carbon source, and removal of cellulose fibers involved in the BC production process (Hornung et al., 2006). In addition, several modified reactors, including a rotating disk bioreactor (Serafica et al., 2002), cylindrical silicone membrane vessel (Yoshino Asakura, & Toda, 1996) and a direct oxygen and glucose feeding reactor (Hornung et al., 2006), have been developed to improve BC production.

An alternative approach to BC production is submerged fermentation through aerated or agitated cultivation. Presently, most BC used for commercial purposes is produced through agitated fermentation, which generates small pellets or granules instead of pellicles (Mormino & Bungay, 2003; Shah et al., 2010). These BC pellets exhibit a lower degree of polymerization, mechanical strength, and crystallinity than those produced under static cultivation. However, the production process is much faster during agitated culture than static culture. The high cell density and better contact with the provided oxygen has resulted in very high volumetric productivity in agitated cultures. Several factors including production rate, desired applications, capacitance and economic feasibility are involved in the acquired synthetic approach.

To date, most efforts have been devoted to identification of inexpensive raw sources for economically feasible production of BC. Some success has already been achieved with the significant production of BC from waste of beer fermentation broth (Ha et al., 2008), agricultural wastes (Kongruang, 2008), molasses (Bae & Shoda, 2004), fruit juices (Kurosumi, Sasaki, Yamashita, & Nakamura, 2009), and cotton based textile wastes (Hong et al., 2012). The combined effects of several carbon sources and addition of supplementary sources have led to BC production. Some previous efforts intended to enhance BC production with static and agitation cultivation cultures using a variety of BC producing strains, carbon sources, alternate inexpensive sources, and supplementary materials are summarized in Table 1.

3.3. Applications and limitations of BC

The unique properties of BC have inspired attempts to use it in a number of commercial products. Commercial items utilizing BC include tires, headphone membranes, high performance speaker diaphragms, high-grade paper, make-up pads, diet-food and textiles (Cannon & Anderson, 1991; Evans et al., 2003). The most exciting applications of BC are in the biomedical field, where they are used as wound dressing materials (Ciechanska, 2004; Czaja et al., 2006), artificial skin, vascular grafts, scaffolds for tissue engineering, artificial blood vessels, medical pads and dental implants (Czaja et al., 2007; Klemm, Schumann, Udhardt, & Marsch, 2001; Shezad et al., 2009; Wan, Hutter, Millon, & Guhados, 2006). Biofill, a membrane prepared from BC, is used as a temporary skin substitute for patients with burns and ulcers (Evans et al., 2003; Fontana et al., 1990). BC has also been proposed for use as a material source of environmentally compatible ion-exchange membranes for fuel cells (Choi et al., 2004; Finkenstadt, 2005). Furthermore, BC has been utilized as biodegradable and biocompatible sensors and actuators (Jeon et al., 2010; Kim & Seo, 2002).

Improvements and modifications are required to enhance the capabilities of all materials prior to their use for different applications. Similar to other polymers, BC has limitations that restrict its application to a certain extent. For example, BC is an excellent wound dressing material since it provides a moist environment that enables swift healing (Shezad et al., 2010). However, it has no antimicrobial properties to prevent infections (Maria et al., 2010). Moreover, if a wound healing material is antioxidant in nature, it can provide protection against reactive oxygen species; however, pure BC does not possess any such activity. Pristine BC also lacks optical transparency, electrical conductivity, magnetism, and hydrophobicity; therefore, it cannot be utilized directly in electrical devices, batteries, sensors, electromagnetic shielding, or electrochromic devices (Evans et al., 2003; Shi et al., 2012). BC has

Table 1
Production of BC in static and agitation cultures with variety of BC producing strains, carbon sources, and supplementary materials.

Microorganism	Carbon source	Supplementary materials	Culture time (days)	Yield (g/l)	Cultivation mode	References
Gluconacetobacter xylinus, Trichoderma reesei	Glucose	Fiber sludge	14	6.23	Static	Cavka et al. (2013)
Gluconacetobacter xylinus	Glucose	Cellulosic fabrics	14	10.80	–	Feng et al. (2012)
Gluconacetobacter medellensis	Glucose	None	14	4.50	–	Castro et al. (2012)
Glucon acetobacter hansenii PJK (KCTC 10505 BP)	Glucose	Glucuronic acid oligomers	10	7.4	–	Ha et al. (2011)
G. xylinus (PTCC, 1734)	Glucose	Date syrup	14	40.35	–	Moosavi-Nasab and Yousefi (2011)
Gluconacetobacter Persimmonis (GH-2)	Glucose	Fructose, beef extract	14	5.14	–	Hungund and Gupta (2010)
Gluconacetobacter xylinus strain (ATCC 53524)	Sucrose	None	4	3.83	–	Mikkelsen et al. (2009)
Glucon acetobacter hansenii PJK (KCTC 10505 BP)	Waste from beer culture	None	14	8.6	–	Ha et al. (2008)
Glucon acetobacter xylinus strain (K3)	Mannitol	Green tea	7	3.34	–	Nguyen et al. (2008)
Gluconacetobacter xylinus (IFO 13773)	Sugar cane molasses	None	7	5.76	–	Keshk and Sameshima (2006)
Acetobacter xylinum (ATCC 700178)	CSL-Fru	Carboxymethylcellulose	5	13.00	Agitated	Cheng et al. (2011)
Acetobacter xylinum	CSL-Fru	Sodium alginate, agar, carboxymethylcellulose.	5	7.05	–	Cheng et al. (2009)
Gluconacetobacter sp. (RKY5)	Glycerol	None	6	5.63	–	Kim et al. (2006)
A. xylinum (BPR2001)	Molasses	None	3	7.80	–	Bae and Shoda (2004).
A. xylinum (BPR2001)	Fructose	Agar/Oxygen	3	14.10	–	Bae et al. (2004).
Glucon acetobacter hansenii PJK (KCTC 10505 BP)	Glucose	Ethanol	3	2.50	–	Park et al. (2003)

high mechanical properties, but the presence of numerous pores reduces its stress bearing capabilities.

3.4. Need for BC composites

The deficiencies mentioned above limit the application of BC in different fields; thus, there is a need for synthesis of its composites. Owing to its structural features, BC has shown tremendous potential as both a matrix and a reinforcement partner in the synthesis of various composite materials (Kim et al., 2011; Ul-Islam et al., 2012a, 2012b). Different composites of BC have already been synthesized to increase its mechanical properties, biological activities, conductivity, magnetic properties and biomedical applications (Evans et al., 2003; Kim et al., 2011; Maneerung et al., 2007; Nakayama et al., 2004; Saibuatong & Philsalaphong, 2010; Shi et al., 2012; Ul-Islam et al., 2012a, 2012b). Table 2 represents composites of BC with different materials and illustrates their potential applications. As shown in Table 2, various composites of BC are synthesized through different synthetic strategies, and the formed composites have additional properties relative to unamended BC. Here, we summarize a portion of the literature related to BC composites. BC composites have been formed using a broad range of materials ranging from NPs to polymers, many of which have been synthesized to enhance medicinal applications of BC. In addition, BC composites with conducting materials have been applied in electric and magnetic devices.

4. Approaches for synthesis of BC composites

Polymer composites are synthesized through numerous methods depending on the nature of the polymer and the combining partner. The synthesis strategy also varies with the required applications. In general, there are two basic composite synthesis approaches, in situ and ex situ (Fig. 1(a and b)). The in situ method utilizes the addition of reinforcement material to the polymer during its synthesis, which then becomes part of the polymer structure (Saibuatong & Philsalaphong, 2010; Serafica et al., 2002; Ul-Islam et al., 2012a). In ex situ synthesis, the polymer matrix is impregnated with reinforcement materials to produce composites. These techniques can be further specified based on the nature of the combining agents and the developmental approach. BC is a biopolymer that can be subjected to all of the aforementioned techniques for polymer synthesis. Here, we attempt to recapitulate a few common approaches utilized for BC composites synthesis for various applications. A comparison of all three approaches based on their advantages and disadvantages is provided in Table 3.

4.1. In situ BC composites synthesis

This method utilizes a synthetic approach in which reinforcement materials are added to BC culture media at the beginning of the BC synthetic process. Microfibrils of BC become denser with time and produce a web shaped structure (Horii, Yamamoto, & Hirai, 1997; Tang, Jia, Jia, & Yang, 2010) that can trap various materials added to the BC synthetic media (Ul-Islam et al., 2012a). The encaged materials become part of the BC fibril network, resulting in BC composites. A schematic diagram showing the synthesis of BC composites through this in situ strategy is provided in Fig. 1(a). Several studies have provided examples of the in situ formation of BC composites. For example, Saibuatong and Phisalaphong synthesized BC-Aloe vera composite films by adding various quantities of aloe vera to BC synthetic media cultured under static conditions. The aloe vera components were entrapped in the BC fibrils and the resulting composite had superior physico-mechanical properties (Saibuatong & Philsalaphong, 2010). Similarly, multi walled carbon nanotubes (CNTs) were added to synthetic media and incubated for two weeks. Structural analysis revealed that the CNTs were trapped between the BC fibrils (Yan, Chen, Wang, Wang, & Jiang, 2008). Ruka et al. recently reported BC-composite with a water insoluble poly-3-hydroxybutyrate (PBH) prepared using an in situ composite synthesis strategy (Ruka, Simon & Dean, 2013). Initial addition of various concentrations of PBH not only affected BC production, but also changed the morphology and crytallinity of BC (Ruka et al., 2013). Several other examples of BC composite materials synthesized through the in situ composite synthesis strategy are also available (Chen, Chen, Huang, & Lin, 2011; Yano, Maeda, Nakajima, Hagiwara, & Sawaguchi, 2008).

Composite synthesis via static BC culture is rather difficult because the particles only remain suspended in the BC synthetic media for a short time. In static culture, a BC sheet is formed on the

Table 2
Various BC composite materials showing the synthetic strategy, composites type, improved properties, and concerned applications.

Reinforcement material	Synthetic approach	Composite type	Improved properties	Applications	Reference
Chitosan	Ex situ Solution penetration	Polymer–polymer	Physical, mechanical antifungal	Biomedical and industrial	Kim et al. (2011)
Gelatin			Physical, mechanical	Biomedical	Nakayama et al. (2004)
Poly aniline			Conducting, medicinal	Electronic devices and biomedical	Shi et al. (2012)
Polyethylene glycol			Thermal stability, biocompatibility	Biomedical	Cai and Kim (2010)
Graphine oxide (GO)			Thermal, mechanical, conducting properties	Electronic devices and industrial	Feng et al. (2012)
Collagen	In situ addition		Tissue engineering scaffold	Biomedical	Luo et al. (2008)
Aloe vera gel			Mechanical, physical	Biomedical and Industrial	Saibuatong and Philsalaphong (2010)
Montmorrilonite	Ex situ Particle penetration	Polymer–particle	Physical, mechanical, antibacterial, thermal	Biomedical, Industrial	Ul-Islam et al. (2012a, 2012b)
Silver nanoparticle			Antibacterial activities	Biomedical	Maneerung et al. (2007)
Palladium			Conducting	Sensors and electronic	Evans et al. (2003)
Gold nanoparticle			Optical, photocatalysis, biosensing	Biosensors and optical devices	Zhang et al. (2010) and Pinto et al. (2007)
Hydroxy apatite			Biocompatible	Biomedical	Hong et al. (2006)
Carbon nanotubes	Ex situ Particle penetration/In situ addition		Mechanical, conducting	Electronic devices	Yoon et al. (2006) and Yan et al. (2008)
Silica			Mechanical properties	Industrial	Yano et al. (2008)
Inorganic particles	In situ addition		Mechanical properties	Industrial	Serafica et al. (2002)

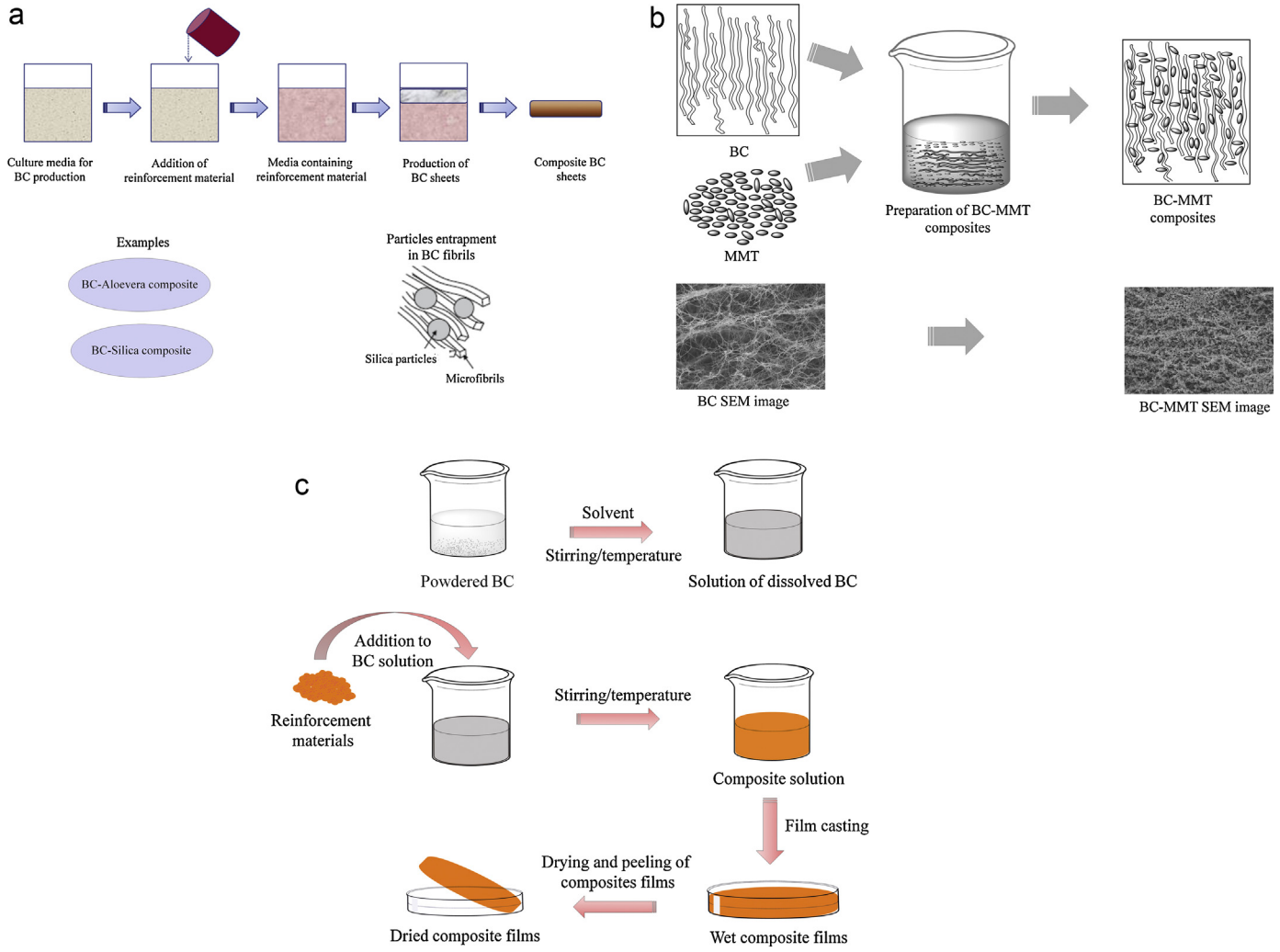

Fig. 1. (a) Schematic representation of BC composites synthesized through an in situ synthetic strategy. Particles entrapped between growing BC fibrils are visible in the figure. (b) Schematic representation of BC composites synthesized through an ex situ synthetic strategy. The example illustrates the penetration of particles in the BC matrix. (c) Schematic representation of BC composites synthesized from dissolved BC solutions. The composite solution was casted to prepare BC films.

Table 3
Comparative study of various BC composites synthesis routes, their advantages and limitations.

Synthetic approaches	Advantages	Drawbacks	References
In situ synthesis of BC composites	Easy processing	Difficulties in composites synthesis due to quick penetration of suspended particles	Saibuatong and Phisalaphong (2008)
	Variety of solid and liquid reinforcement materials	Limitation of BC composites synthesis with antibacterial agents	Serafica et al. (2002)
	Varity of BC synthesis methods (Static, Shaking and Agitated)	Limited applications of BC composites synthesized through agitation method	Cheng et al. (2009)
		Disturbance in basic structural features of BC	Ul-Islam et al. (2012a)
Ex situ synthesis of BC composites	Composite synthesis using BC films	Only submicron to nano sized particles can penetrate in BC	Shi et al. (2012)
	No problems with antimicrobial materials	Difficulties in homogenous distribution of particles in BC	Yano et al. (2008)
	Retention of basic structural features of BC	Hydrophobic materials can't be used for BC composites	Ul-Islam et al. (2012b)
	Variety of liquefied and suspendible materials		Katepetch and Rujiravanit (2011)
Synthesis of BC composites from BC solution	Controllable composite composition	Limited solubility of BC	Lindman, Karlström, and Stigsson (2010)
	Wide range of reinforcement materials	Expensive approach	
	No restriction with bactericidal, magnetic, conducting materials	Destruction of BC primary structure.	

surface of the media at the media air interface (Cheng, Catchmark, & Demirci, 2009). After being precipitated or moving lower in the media, the particles cannot be entrapped in the surface BC fibers. Several strategies have been applied to overcome this problem, including composite synthesis during agitated culture (Cheng et al., 2009; Yan et al., 2008) and designing vessels equipped with spinning discs (Serafica et al., 2002). Agitation culture provides a much better environment for the added materials to become entrapped in the BC fibrils. This is because the constant movement in the media prevents the particles from settling. The homogeneous BC formation and distribution keeps the particle entrapment constant. Serafica et al. reported the synthesis of BC composites with a variety of materials entrapped inside BC produced through agitation culture using a specially designed rotating disk. The density of penetrating particles was dependent on the particle size, rotation speed and suspension concentration (Serafica et al., 2002). The in situ BC composite synthesis technique is the most widely used approach that employs a wide range of modifications in agitation equipment and operating methods. However, this technique has certain limitations that prevent the synthesis of many BC composites. For example, several important bioactive (antibacterial) agents (Ag, ZnO, TiO_2 etc.) cannot be added directly to the media because of their toxic effects on microorganisms. Moreover, BC composite synthesized through agitation culture cannot be applied as a gel or sheet in biomedical applications, while gels/sheets formed in static culture cannot entrap reinforcement materials for composite synthesis.

4.2. Ex situ BC composites synthesis

The problems associated with in situ BC composite synthesis can be resolved by incorporating liquid and NPs into the structural matrix of the prepared BC (Fig. 1(b)). The interaction might be physical or occur through definite hydrogen bonding between the BC and reinforcement materials. Liquid substances and tiny solid particles can easily penetrate and become engrossed inside the porous BC matrix. In addition to this physical absorbance, the presence of OH moieties in cellulose chains results in hydrogen bonding with the invading reinforcement materials. This strategy is much simpler and more valuable than the in situ composite synthesis strategy. The most important factor in utilizing this method for composite synthesis is that the original structure of BC remains almost completely unchanged. BC sheets obtained through static cultivation are usually utilized for composite synthesis, which can be effectively employed in biomedical and other industrial applications (Maneerung et al., 2007). Accordingly, the strategy has been applied to a number of BC composites, including those formed from polymers, inorganic materials, metals and metallic oxides (Evans et al., 2003; Maneerung et al., 2007; Nakayama et al., 2004; Shi et al., 2012; Ul-Islam et al., 2012a, 2012b, 2013; Yano et al., 2008; Yoon, Jin, Kook, & Pyun, 2006).

The properties of BC are differently affected by the reinforcement material. Our studies of BC-Ch composites illustrated that Ch molecules penetrated the BC matrix and combined through a strong hydrogen bonding interaction between the O—H and N—H moieties. The composite strengthened the BC fibrils by enhancing their mechanical properties (Ul-Islam et al., 2012b). BC composites formed with gelatin, hydroxyapatite (HA), etc., were synthesized through the same strategy to enhance their biomedical applications (Hong et al., 2006; Nakayama et al., 2004). To accomplish this, wet BC sheets were kept in polymeric solution for different lengths of time to ensure sufficient penetration and attachments of the polymer to BC. A number of BC composites have been synthesized with inorganic materials using the same synthetic strategy for different applications. The antibacterial activities of BC were enhanced through incorporation of Ag NPs into the BC sheets. The composites were prepared through a green route without using any chemical reagent, utilizing BC as both a reducing and stabilizing agent. Pieces of BC were rinsed with $AgNO_3$ solution and then heated gently to enable hydrothermal production of Ag NPs. The particles deposited in the BC matrix produced high antibacterial activities (Yang, Xie, Deng, Bian & Hong, 2012). BC-Pd, BC-CNT and BC-MMT composites were synthesized through the same ex situ composite production strategy (Evans et al., 2003; Ul-Islam et al., 2012b; Yoon et al., 2006).

The major hurdle associated with the ex situ composites synthesis strategy is the size and nature of the reinforcement material. Specifically, only submicron to nanosized materials can be impregnated into BC matrix. This is because larger particles cannot enter the BC pores, and hydrophobic materials are not able to combine with BC. Moreover, the structural arrangement of the BC fibril is not always uniform; therefore, penetrating materials might not be homogenously distributed inside the BC matrix. Accordingly, there is a need to identify new BC composite synthesis routes to resolve this problem.

4.3. Synthesis from solution of dissolved BC

The formation of BC composites from solutions of dissolved BC is a better approach that can be utilized to produce a broad range of BC composites with a variety of materials. Moreover, the composition of matrix and reinforcement materials can be easily controlled. The homogenous distribution and mixing of either partner is also possible. A generalized scheme for BC composite development through this approach is shown in Fig. 1(c). The major problem associated with this method is the limited solubility of BC. Indeed, BC is very difficult to dissolve because it is insoluble in water and most widely used organic solvents (Lindman, Karlström, & Stigsson, 2010). It is also insoluble in non polar solvents owing to its polar nature, but its aqueous insolubility is more difficult to understand. The strong inter and intra molecular hydrogen bonding and the high crystallinity of BC have been suggested as possible reasons for its low solubility (Lindman et al., 2010). This unusual behavior has created great interest in studying its solubility and further modification. However, few classes of compounds can completely dissolve BC (Lindman et al., 2010). Some of these solvents include N-methy morpholine N-oxide (NMMO) (Gao, Shen, & Lu, 2011), ionic liquids (Chen et al., 2011a), $ZnCl_2$ ($3H_2O$) (Lu & Shen, 2011), NaOH (Łaszkiewicz, 1998), and LiOH/urea/thiourea (Zhang & Luo, 2011). Dissolved BC can easily be utilized to synthesize BC films, microfibers, nanofibers, nanocrystals and composite materials. Gao et al. (2011) prepared regenerated BC films from BC dissolved in NMMO and found them to possess uniform fibril arrangement with superior mechanical and thermal properties. The successful dissolution of BC and synthesis of regenerated films have generated tremendous interest in utilization of this method for composites preparation. Different materials added to dissolve BC can be mixed well in BC solution and processed with ease to generate composite films or fibers. This approach has not been intensely applied for BC composites, but has been comprehensively employed for the synthesis of various polymeric composites with inorganic materials including clay, metals and NPs (Gao, 2004). Additionally, we recently synthesized RBC-ZnO composite using the same technique, and the RBC-ZnO nanocomposites were found to possess enhanced thermal, mechanical, and antibacterial properties [unpublished data]. Composite produced through this method has stronger interactions between BC and the combining material; accordingly, this approach will be very useful in future strategies of BC composite development.

5. Classes of BC composites

BC composite was prepared using materials ranging from bulk polymers to NPs. Based on the nature of the reinforcement material, BC composite can be classified into organic materials and inorganic materials. These two main classes can be further divided into subclassifications of BC composites with polymers, NPs, metals, metal oxides, clays and macro-sized solid particles. A detailed sketch of the composites classification with specified examples is given in Fig. 2(a and b).

5.1. BC-composites with polymers

Being a biopolymer, BC has been utilized in the synthesis of various composites with different polymeric materials. Polymers are organic in nature, and most have specific hydrogen bonding sites. The similar nature and presence of the binding sites facilitates the synthesis of BC composites with numerous polymers of varying nature. There are several examples of BC composites with biopolymers that have led to improvements in biomedical applications, physico-mechanical properties and conducting capabilities of BC. Progress in composite synthesis is driven by limitations in BC application in various fields. Limitations such as antimicrobial, antioxidant, wound healing, and conducting properties have been overcome to a certain extent through the use of composites.

The synthesis of BC-Ch composites is based on the active role of Ch in biomedical fields. Chitosan has been recognized for its broad spectrum of activities, which include absorption of wound exudates, wound-healing effects, tissue-engineering scaffolds, drug delivery, anti-microbial, anti-fungal and anti-viral properties (Ai, Wang, Xia, Chen, & Lei, 2012; Dutta, Dutta, & Tripathi, 2004). Composites of BC with Ch have been prepared through both in situ and ex situ penetration methods. The similar structural features and presence of O—H and N—H groups produce strong bonding between the BC and Ch molecules. BC-Ch composites further show pronounced improvements in physico-mechanical and biomedical properties (Ciechanska, 2004; Kim, Cai, & Chen, 2010; Ul-Islam, Shah, Ha, & Park, 2011). BC composites with gelatin are based on the same strategy as BC-Ch. The impressive biomedical properties of gelatin have led to investigations of its utilization in combination with scaffold materials such as BC. Composites of BC-Gelatin have been applied in medical fields owing to their enhanced cell adhesion and proliferation activities. Several other biopolymers have been combined with BC to increase its biomedical applications, including BC-COL (Luo et al., 2008), BC-Alginate (Zhang & Luo, 2011) and Novo aloe vera (Saibuatong & Philsalaphong, 2010).

BC composites with some conducting polymers have been produced to impart conducting properties to BC. Shi et al. (2012) reported a BC-PAni composite prepared by the in situ polymerization of PAni in solution containing BC pellicles in which the polymerizing PAni formed a layer on the BC surface. The composites showed high conductivity, which was further improved by controlled doping with acids (Shi et al., 2012). Similar results of enhanced conductivity in BC-PAni composites have been reported elsewhere (Hu, Chen, Yang, Liu, & Wang, 2011; Lee, Kim, & Yang, 2012).

In addition to these particular examples, various BC–Polymer composites have been synthesized to improve different applications of BC. BC-Polymeric composites have greatly improved the thermal, physical mechanical and biological properties of BC.

5.2. BC composites with metals and metal oxides

Metals and metal oxides are a separate class of materials that have received a great deal of attention owing to their exceptional properties. Their small size, charged nature, and individual behavior govern the specific characteristics needed by the polymeric materials. Accordingly, incorporation of metals and metal oxides into polymeric materials has been in practice for a long time. Prominent features of metallic particles include their conducting and magnetic capabilities, photo-oxidizing activities, and antimicrobial properties (Qian, 2004). Metals and metallic oxides have been extensively used in polymer composites to introduce electrical, optical, conducting and antimicrobial properties in polymers. They have shown enhancement in the physical and mechanical properties of the polymers (Bloor et al., 2005; Islam et al., 2011; Maneerung et al., 2007; Qian, 2004). Like other polymeric materials, BC can be used to make composites with metals and metallic oxides through a variety of synthetic approaches. Here, we describe a few of these composites and their synthetic approaches and applications.

BC-Ag nanocomposites are primarily synthesized to incorporate antibacterial properties into BC. Maneerung et al. (2007) prepared BC-Ag nanocomposite by impregnation of BC sheets in $AgNO_3$ solution that was reduced with $NaBH_4$ to produce metallic Ag. Nanoparticles adsorbed onto the BC surface were observed through UV absorption and XRD spectral peaks. BC-Ag

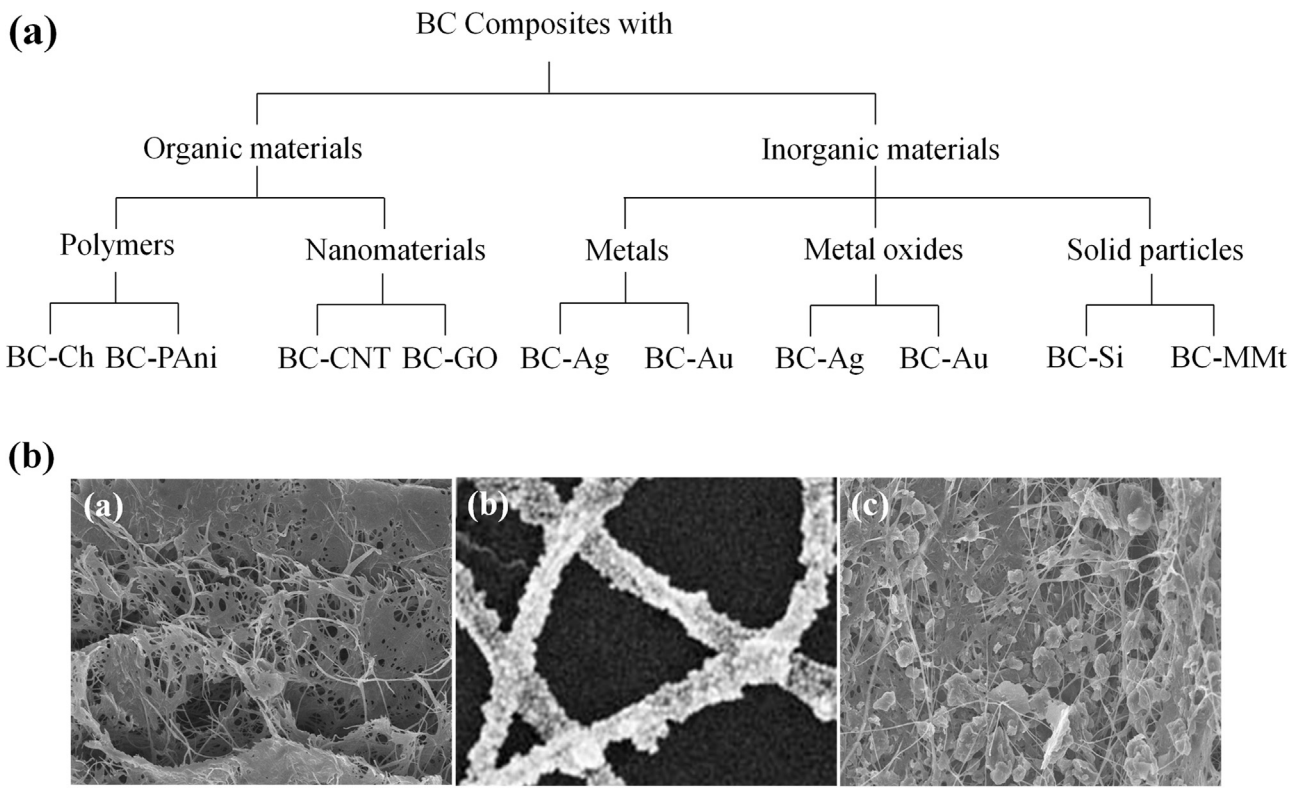

Fig. 2. (a) A generalized classification of BC composites prepared with various materials of organic and inorganic nature. (b) FE-SEM images of a few BC composites including BC-Ch (a), BC-Au (b) and BC-MMT (c).

nanocomposite produced very strong antibacterial effects against gram positive and gram negative bacterial species (Maneerung et al., 2007). A similar study that used a slightly modified synthetic approach was conducted by Maria et al. (2010). The obtained BC-Ag nanocomposites showed large bactericidal effects against *Escherichia coli*. BC-Ag nanocomposite membranes have the potential for application in wound dressings, where they can provide protection against invading microorganisms in addition to their primary role in wound healing. Sureshkumar et al. reported a facile method for preparing magnetic BC-Ag nanocomposite. They first homogenized the 3-D nanofibrous structure of BC with a mixture of ferric and ferrous. As the pH increased, magnetic NPs were precipitated and attached to the BC surface. A polydopamine layer was then coated onto the magnetic BC nanofibers, which reduced the Ag NPs from silver nitrate solution onto the magnetic BC surface. The magnetization of the as-prepared Ag nanocomposite was well maintained and the magnetic Ag nanocomposite presented impressive antimicrobial activity against the model microbial species (Sureshkumar, Siswanto, & Lee, 2010)

Gold NPs have received immense attention owing to their potential for use in sensors, catalysis, and several related fields (Berry, Gola, Kundu, Murphy, & Saraf, 2010). BC-Au composites have been prepared through specific techniques and utilized for bio-sensing and in bio-devices. Zhang et al. (2010) utilized BC nanofibers as robust biotemplates for BC-Au nanocomposite synthesis. In their study, the pellicles were disrupted to separate the BC nanofibers, after which Au NPs were deposited onto the fibers. The field emission scanning electron microscopy (FE-SEM) morphology of the BC-Au nanocomposites clearly demonstrated the attachments and depositions of Au NPs onto the surface of individual nanofibers. The composites had the potential for use in enzyme immobilization and in bioelectronics systems (Zhang et al., 2010).

Another striking capability of some metallic particles is their conductivity and magnetic properties. BC is deficient in these properties and therefore requires modification to be applicable in a number of fields. An effort to address this deficiency has been made through the synthesis of BC-Pd composites. Specifically, these composites are synthesized through deposition of metals onto the BC surface by soaking BC in ammonium hexachloropalladate solution. Metallic ions are then reduced by the cellulose and produce a coating on the BC surface. BC-Pd nanocomposites were found to be capable of catalyzing the generation of hydrogen when incubated with sodium dithionite. Moreover, BC-Pd generated an electrical current from hydrogen in membrane electrode assemblies (Evans et al., 2003). Another recent study of BC-Pt) nanocomposites showed that they have significant conducting efficiency and can be used as fuel cells and biosensors. BC-Pt nanocomposites were prepared through the liquid phase chemical deoxidization method, and the generated composites produced high electro catalytic activity. Moreover, the current density was dramatically increased by doping the membrane with protonic acid or inorganic acid (Yang et al., 2009).

In addition to metals, a few metallic oxides have been combined with BC to enhance its biological and conducting activities. Gutierrez et al. (2012) recently prepared BC-titanium oxide nanocomposites. To accomplish this, NPs of TiO_2 prepared by the sol gel method were deposited onto a hydrophilic BC membrane. The hydrogen bonding interaction strongly held the NPs between the BC fibrils. Comparative EFM (electric force microscopy) studies of pure amorphous TiO_2 NPs and TiO_2 attached to BC nanofibrils revealed a stunning scenario. Specifically, amorphous pure TiO_2 NPs were electrically neutral and therefore did not respond to the applied voltage. However, BC-TiO_2 nanofibers were able to respond to voltage applied to the EFM tip, regardless of the sign of the applied voltage. The electrostatic dipole–dipole interactions produced between BC and TiO_2 NPs influenced their conductive behavior, and the response to the applied voltage from the BC-TiO_2 surface increased with increasing voltage (Gutierrez,

Tercjak, Algar, Retegi, & Mondragon, 2012). Therefore, the BC-TiO_2 nanocomposite was shown to be an important conducting material and it has since been utilized as a controlled drug delivery material. Sun et al. reported another approach for synthesis of TiO_2 NPs in the presence of BC. In their method, BC not only acts as a matrix to hold these NPs, but also affords sites for the nucleation of TiO_2 NPs from the medium. BC/TiO_2 composites demonstrated higher photocatalytic activity for a model pollutant (methyl orange) than that of the commercial photocatalyst, P25 (Sun, Yang, & Wang, 2010). Katepetch and Rujiravanit have prepared magnetically responsive BC sheets by synthesizing magnetic NPs (FeO) inside BC sheets using an ammonia gas enhancing in situ co-precipitation method. The method provided successful dispersion of magnetic NPs inside the BC sheets. The saturation magnetization of the magnetic nanoparticle-incorporated BC sheets ranged from 1.92 to 26.20 emu/g at 300 K and from 2.96 to 28.10 emu/g at 100 K (Katepetch & Rujiravanit, 2011).

Non aggregated ferromagnetic materials can be effectively used for targeted drug-delivery as well as high levels of information storage. The randomly organized nanofibers of BC offer an open porous structure that acts as a template for the magnetic inorganic NPs. The web shaped porous network not only stops particle aggregation, but also prevents inter-particle interaction. BC composites with FeO magnetic NPs (varying in size from 2 nm to 20 nm) produced supermagnetic behavior at room temperature (Olsson et al., 2010; Small & Johnston, 2009). Sourty et al. reported the generation of BC composite films with FeO NPs produced by in situ impregnation of FeO particles into the BC membrane. The dark green coating on the BC surface indicated the presence of FeO NPs, which could be detected with the naked eye and were confirmed by transmission electron microscopy (TEM), SEM and X-ray diffraction (XRD) (Sourty, Ryan, & Marchessault, 1998). The superparamagnetic behavior of all BC-FeO composites was observed by room temperature magnetization. The inclusion of magnetic properties in the BC membrane could be vital in many sectors, especially in electrical and magnetic devices (Sourty et al., 1998).

5.3. BC composites with solid materials

In addition to metals and metallic oxides, attempts have been made to combine various micro to nanosized particles with BC to accelerate its efficiency and increase its applications. Among these, clay materials, silica (Si) particles and carbonates have produced valuable effects. Some of these composites are described briefly below.

Biomedical applications of various clay materials have received attention owing to their potential utilization in different composites and biomedical materials. Clay particles are micro to nanometer in size and are strikingly enriched in minerals (Ul-Islam et al., 2012b). Among these, members of the smectite family, which includes bentonite and montmorrilonite (MMT), have become popular owing to their biomedical applications and polymer reinforcing properties (Meng, Zhou, Zhang, & Shen, 2009; Ul-Islam et al., 2012b). Accordingly, various clays have been utilized as reinforcing materials in polymer clay composites (Leszczyńska, Njuguna, Pielichowski, & Banerjee, 2007; Li, Jia, Zhu, Ma, & Sun, 2010; Meng et al., 2009). Studies have shown up to ten fold increases in mechanical properties following impregnation of the polymer matrix with clay particles (Liu, Chaudhary, Yusa, & Tade, 2010). In addition to reinforcing polymers, these clays and their products have a broad spectrum of applications in medical fields with no side effects, such as skin protection, cleansing and antibacterial activity (Haydel, Remenih, & Williams, 2008), immobilization of cell toxins produced by various bacteria (Meng et al., 2009), and excellent wound healing and blood clotting capabilities (Emami-Razavi et al., 2006). Furthermore, clay polymer composites can be synthesized without the use of any toxic or expensive organic solvents (Giannelis, Krishnamoorti, & Manias, 1999).

We prepared BC-MMT composites through ex situ impregnation of MMT particles inside MMT sheets with the goal of combining the desired properties of BC and MMT to obtain a biomaterial with enhanced physico-mechanical properties for various applications (Ul-Islam et al., 2012b). The composites produced much higher mechanical and thermal properties than pristine BC (Ul-Islam et al., 2012b). Considering the ion exchange properties of MMT, we further extended the work to the synthesis of BC composites with modified MMTs. In that study, BC composites with Ca-MMT, Na-MMT and Cu-MMT were prepared and their antimicrobial activities were investigated. The BC composites with modified MMTs produced significant antibacterial effects (Ul-Islam et al., 2013). These BC-MMT composites are of tremendous interest considering their role in biomedical and other industrial fields.

Although we discussed BC composite with different classes of materials, composites with some individual solid particles prepared through diverse strategies have also been reported. Yano et al. (2008) prepared BC composites with silica particles through both in situ and ex situ impregnation strategies. Both methods ensured the formation of composites and the presence of silica particles inside the BC fibers. However, the effects on structural features and mechanical properties were different for each type of composite. The BC-Si composite produced through in situ impregnation of silica particles had lower mechanical properties than the control, while those of ex situ synthesized BC-Si composite were higher (Yano et al., 2008). Ashori et al. recently reported BC-Si composites prepared by immersing solid BC gels in an aqueous solution of tetraethoxysilane and then pressing the treated BC matrices at 120 °C and 2 MPa. The mechanical properties of the BC-Si matrices were significantly increased with the 7% loading of Si particles (Ashori et al., 2012).

Serafica et al. (2002) developed a bioreactor equipped with rotating discs for inclusion of various solid particles inside BC. The major problem associated with incorporation of solid particles is that they precipitate quickly and do not penetrate the gel produced at the surface. The rotating discs kept the particles suspended, providing better opportunities to embed inside the BC produced on the discs. However, in these systems, the particle penetration rate is size and geometry dependent and the produced composites are not entirely homogenous. The various particles used in the BC composites included aluminum, iron, glass beads, $CaCO_3$, and paper fibers (Serafica et al., 2002). In a similar study, very small paper (cellulose) particles were suspended in BC culture media and incorporated into formed gelatinous BC mats. BC production was then carried out in a rotating disk bioreactor (Mormino & Bungay, 2003) and the BC-paper composites were found to be much stronger than the control BC mats. Later, Kuure-Kinsey et al. (2005) suggested an improved model for the production of BC composites with solid particles. Their produced bioreactor could synthesize composites with solid particles in different ratios, controlled concentrations, stripes, gradients, and mixtures of different particles (Kuure-Kinsey, Weber, Bungay, Plawsky, & Bequette, 2005).

6. Applications of BC composites

As described above, BC composites are synthesized to enhance existing properties and impart some additional properties for certain specific applications. A variety of composite synthetic approaches and a number of reinforcement materials have resulted in a broad range of composite designs for different applications. Being a multifunctional material, BC can combine with different materials for specific applications. Here, we summarize some of the

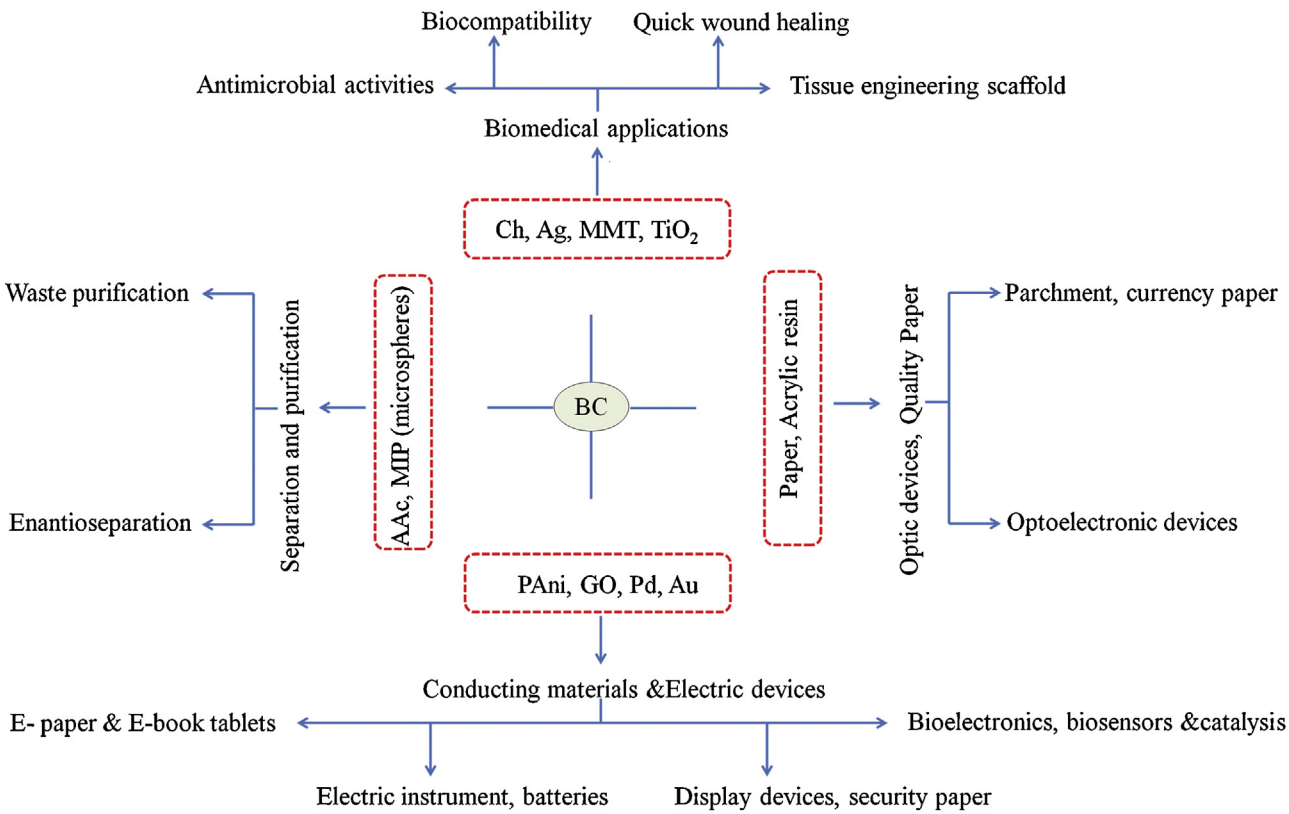

Fig. 3. Illustration of BC composites with various materials for specified applications in different fields.

important applications of BC composites, which are also illustrated in Fig. 3.

6.1. Biomedical applications

BC is widely used as a dressing material in medical fields. To impart antimicrobial, antifungal, tissue regeneration, and biocompatible properties in BC, its composites with polymeric and non polymeric compounds were prepared. BC-Ch composites increased the biocompatibility, cell adhesion and proliferation properties. The composites produced better results as wound dressing materials and in tissue engineering scaffolds (Kim et al., 2010). Accordingly, BC-Ch can be applied to the treatment of hard to heal wounds, skin ulcers, bedsores, burns, and wounds needing frequent dressing changes (Ciechanska, 2004). Ul-Islam et al. (2011) found that BC-Ch have higher water holding capacities and the potential to retain water for a long time, with very slow water release, which is of the utmost importance for wound healing purposes. BC-Ch composites were also found to have both bactericidal and bacteriostatic activities against gram positive and gram negative bacteria (Ciechanska, 2004). When brought into contact with human fluid containing lysozyme, these composites are degraded and produce glucosamine and N-acetylglucosamine units, which accelerate the wound healing process (Ciechanska, 2004). Lin et al. recently reported the skin wound healing efficacy of BC-Ch composites in experiments assessed with rat models (Lin, Lien, Yeh, Yu, & Hsu, 2013). They found that the composite did not produce any toxic effects on animal cells. Moreover, examination of the tissue regeneration process revealed that wounds treated with BC–Ch composites epithelialized and regenerated earlier than those treated with BC or commercially available dressing materials (Lin et al., 2013). BC-gelatin composites produced similar effects during cell adhesion and proliferation. Fibroblast cells showed definite adhesion and proliferation when incubated with BC-gelatin composite for 48 h. The composite biocompatibility was much better than that of pure BC. Accordingly, the prepared BC-gelatin scaffolds are bioactive, indicating that they can be used for wound dressing and as tissue engineering scaffolds (Kim et al., 2010).

Tissue replacement through newly developed biomaterials is an important aspect of the present advances in biomedical fields. However, the replacement materials should possess mechanical properties equivalent to those of body tissues. BC composites with polyvinyl alcohol (PVA) were prepared for development of such biomaterial devices that could replace the tissue. The mechanical and anisotropic behavior was found to be controllable by the relative amounts of BC and PVA. It has been proposed that these composites could be shaped for devices that could replace cardiovascular and other connective tissues (Millon, Guhados, & Wan, 2008). BC-COL composites were synthesized for potential tissue engineering applications through an in situ composite synthesis strategy. The high biodegradability, low antigenicity and cell-binding properties of COL are important characteristics for a biomaterial from a medical point of view. However, these applications are hampered by the low mechanical properties of BC-COL composite enhances the mechanical strength of COL and the cell adhesion properties of BC; thus, these composites have the potential for use in biomedical fields (Luo et al., 2008). BC composite materials with cotton have been specifically developed for biomedical applications. These composites showed much higher water absorption potential, which is an important prospect in wound dressing and healing applications (Meftahi et al., 2010).

In addition to biocompatibility, the second major shortcoming associated with the medical application of BC is its non-bactericidal nature. As a result, several bactericidal elements have been attached to BC to enhance its antimicrobial activities. BC-Ag nanocomposites were prepared through a variety of routes for the same purpose. BC-Ag composites were found to be effective against many bacterial and fungal species, thereby reducing

the chances of wound infection when utilized as dressing materials. The ultimate result of such composites is rapid and safe wound healing (Maneerung et al., 2007; Maria et al., 2010). BC-Au nanocomposite accomplished similar biomedical applications with specific usage in biosensors and enzyme immobilization processes (Zhang et al., 2010). A novel H_2O_2 biosensor was prepared using BC-Au nanocomposites as outstanding supports for horseradish peroxidase (HRP) immobilization. The nanocomposites could be further processed for the immobilization of many other enzymes, thus enabling their application in bioelectroanalysis and bioelectrocatalysis (Zhang et al., 2010). Composites with metallic oxide (BC-TiO_2) also impart antibacterial and conducting properties to BC and therefore enhance its biomedical applications (Gutierrez et al., 2012).

BC composites with clay materials have shown tremendous potential as biomedical materials. BC-MMT composites not only enhance the mechanical properties of BC, but also impart unique antibacterial properties and wound healing potentials to the BC (Ul-Islam et al., 2012a, 2012b, 2013). Moreover, the composites are completely safe for wound dressing purposes and utilize the effective role of both BC and MMT for such applications. The ion exchange and drug carrying capabilities of MMTs further enhance their applicability (Ul-Islam et al., 2012a, 2012b, 2013).

6.2. Conducting materials and electrical devices

BC consists of a three dimensional fibril network that looks similar to paper upon drying. The potential of BC as substrates for flexible optoelectronic and photonic devices has recently been the subject of various investigations (Okahisa, Yoshida, Miyagushi, & Yano, 2009). Being non-conducting in nature, BC is initially converted to electrically conductive sheets by incorporation of some conductive materials. The conductive or semi conductive BC sheets are then immobilized with electrochromic dyes and attached to electrodes. Subsequent application of potential to the electrodes results in a reversible color change (Shah & Brown, 2005). These display devices are highly flexible, biodegradable having contrast and high reflectivity. Organic light emitting diode (OLED) devices based on flexible bacterial cellulose membranes have already been established (Legnini et al., 2008). Moreover, Okahisa et al. (2009) successfully fabricated OLED displays on cellulose and acrylic resin nanocomposite. These devices are useful in numerous applications including e-newspapers, e-book tablets, rewritable maps, dynamic wall papers, and learning tools (Shah & Brown, 2005). Several attempts to synthesize BC composites containing various conductive materials have also been made to achieve the final goal of preparing electrical devices, including attempts to synthesize BC composites with conducting polymers as well as the conducting metals. Brief descriptions of a few of these efforts are provided below.

The BC-PAni composite produced a high degree of conductivity and was therefore suggested for application in biosensors, flexible electrodes, flexible displays, platform substrates to evaluate the effects of electrical signals on cell activity, and to guide desired cell function for tissue engineering applications (Hu et al., 2011; Shi et al., 2012). BC composite with graphene oxide (BC-GO) greatly enhanced the conducting properties while imparting increased mechanical properties. The flexible and electrically conductive BC-GO composite film with striking mechanical properties is a candidate for advanced biochemical and electrochemical devices (Feng et al., 2012). The incorporation of carbon nanotubes into the BC matrix improves its material properties and conductivity. Indeed, the electrical conductivity of the BC-MWCNT composite was found to be approximately 1.4×10^{-1} S/cm. These high conductivities are helpful in designing bio-electrodes, biosensors and electronic devices (Yan et al., 2008; Yoon et al., 2006). BC-Pd composites were also found to have high conductivity. BC-Pd generated an electrical current from hydrogen in membrane electrode assemblies (Evans et al., 2003). These composites have potential applications in electrical instruments, batteries, and display devices. Similarly, the conducting properties of BC-Pt can be efficiently utilized in synthesis biosensors (Yang et al., 2009). An important prospect of BC-metal NPs composites is their use in proton exchange membranes in fuel cells. BC composites with Pd and Pt NPs have shown the potential for oxidation of hydrogen in fuel cells. The ionic conductivity of pristine BC is further increased by doping, which facilitates its utility as a proton exchange membrane in fuel cells (Vitta & Thiruvengadam, 2012). Au NPs are used in electronics, sensors, catalysis, and several related fields. Moreover, Au composites prepared using biomolecules and microorganisms as templates have been employed as biosensors (Berry et al., 2010). The major problem associated with such biotemplates is their low chemical and mechanical stability. Polymers and sol–gel matrices have been shown to be better substitutes that can provide a network structure for Au immobilization and have higher stability (Zhang et al., 2010). Zhang et al. utilized BC nanofibers as robust biotemplates for BC-Au nanocomposite synthesis, and these Au-BC nanocomposites were used for the construction of H_2O_2 biosensors. The composites were also found to have the potential for use in enzyme immobilization and in bioelectronics systems (Zhang et al., 2010). Pinto et al. reported photocatalytic applications of BC-Au nanocomposites. Specifically, they illustrated the long term stability of BC-Au nanocomposites with impressive applications in optical and catalytic fields. These nanocomposites are of interest to security paper applications (Pinto, Marques, Martins, Neto, & Trindade, 2007).

6.3. Separation and waste purifications

BC has the potential to be utilized as a purification/separation membrane if modified with certain materials. Composite membranes of BC were prepared by modification using acrylic acid (AAc) with ion exchange capacity. The BC-AAc composite membranes possessed electrochemical properties comparable to commercial membranes, as well as excellent absorption capability for heavy metals, binding with metal ions, and separation of trace metals. Use of BC composite with biodegradable materials for such purposes is effective since their disposal does not cause environmental problems. Finally, these composites could be effectively utilized for waste water treatment (Choi et al., 2004).

A promising approach toward utilization of BC as a porous substrate for homogeneous distribution of molecularly imprinted polymers (MIP) was developed several years ago. Composites of BC with MIP particles act as a separating membrane, and MIP NPs are well dispersed in BC, resulting in their being plenty of binding sites available for attachment of specified enantiomers. MIP microspheres selective for S-propranolol were successfully distributed in porous BC matrix with controlled allocation and availability of the molecule recognition sites (Jantarat, Tangthong, Songkro, Martin, & Suedee, 2008). The proposed MIP (microspheres)-BC composite membrane system could be applicable for manufacturing new membranes with controllable permeability corresponding to the presence of target solutes (Jantarat et al., 2008). Similarly, BC-MIP composite membranes were developed for the separation of S-propranolol enantiomer. The composites were successfully utilized for enantioselective separation. BC and MIP composite membrane have great potential for application as a transdermal enantioselective system for racemic propranolol (Bodhibukkana et al., 2006).

6.4. Composites with high mechanical strength for industrial applications

BC-composites with high strength have potential applications in a variety of industries. Addition of materials to BC producing media results in incorporation of such materials into growing BC fibrils, which subsequently enhances the mechanical properties of BC. Scrap paper particles added to BC growth media were found to be embedded in the growing cellulose. Moreover, the quantities of the added paper were controllable, and the produced composites were stronger than paper. This type of paper could be used as parchment or for currency (Mormino & Bungay, 2003). In a related study, BC was prepared by adding glucose phosphate along with glucose to the culture media. In this system, the phosphate became incorporated inside the BC gels. These BC containing phosphates were then used for synthesis of composites with wood pulp during paper sheet formation. BC in composite with pulp significantly enhanced the strength and fire resistance properties of the paper sheets. Thus, these composites will likely play an important role in the synthesis of quality paper (Basta & El-Saied, 2009).

Optoelectronic devices are prepared from optically transparent polymers; however, the low mechanical properties of such polymeric materials create a few shortcomings in their applications. BC composites with such materials possess very high mechanical properties. Moreover, in these composites, BC acts as a reinforcement material and does not disturb the optical properties of the polymer (Nogi, Handa, Nakagaito, & Yano, 2005). Nogi et al. prepared BC nanocomposites with acrylic resins by impregnating BC sheets in acrylic resin solution. The composites were transparent and had great potential for utilization in optoelectronic devices. Moreover, the composites were much stronger and showed stable transmittance upon temperature variation (Nogi et al., 2005).

7. Concluding remarks

BC composites have become important to the development of BC hydrogels for numerous applications. Besides improving the existing properties, composite materials have imparted BC with new features. Developmental approaches to synthetic strategies have led to novel methods of composite synthesis. Physical, chemical and biological properties of BC have been improved, ultimately enhancing their biomedical, electronic and environmental applications. Antimicrobial, conducting, separating, purifying, biocompatibility, biosensing and many other characteristics have also been imparted in the BC composite. The synthetic approaches, classification, and applications of numerous BC composites compiled in the present study will provide insight into future developments and applications of BC composites.

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