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ORIGINAL RESEARCH

Ecofriendly green biosynthesis of bacterial cellulose by *Komagataeibacter xylinus* B2-1 using the shell extract of *Sapindus mukorossi* Gaertn. as culture medium

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Abstract Ecofriendly green biosynthesis of bacterial cellulose (BC) using a low-cost carbon source from the shell extract of *Sapindus mukorossi* was studied by *Komagataeibacter xylinus* B2-1. After 7 d of incubation, strain B2-1 produced 1.31 g L^{-1} BC, which had similar micro-morphology and structural properties to that from Hestrin–Schramm medium based on scanning electron microscopy, X-ray diffraction and Fourier transform infrared analyses. While strain B2-1 grew well and produced BC efficiently at pHs ranging from 4.0 to 6.0, the considerable BC production was only found at temperature of 30 °C. The present investigation can provide a new low-cost carbon source for BC preparation and lead towards commercialization and industrial scale up BC.

Yong-He Han and Hai-Long Mao have contributed equally to this work.

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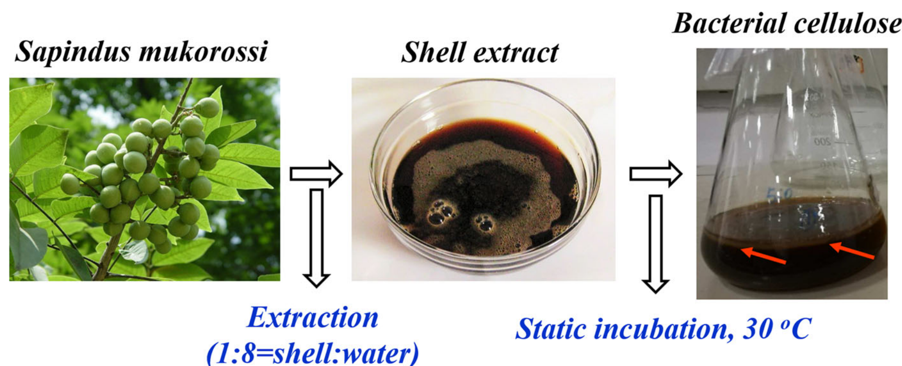
Graphic abstract



Keywords Green chemistry · Bacterial cellulose · Low-cost carbon source · *Sapindus mukorossi* · *Komagataeibacter xylinus* · Bioresource technology

Introduction

Cellulose is the most widely distributed polysaccharide in nature and mainly originated from plants, accounting for more than 50% of the plant carbon content. Besides plants, microorganisms including bacteria, fungi and algae are also good candidates for cellulose production. Due to its high purity, high crystallinity, selective porosity, high water-capacity, low production cost, lack of toxicity, easy sterilization, good biocompatibility and renewable properties, the cellulose produced by bacteria (i.e., bacterial cellulose, BC) has attracted great concerns and widely used for preparation of the high-strength recycled paper, cosmetic moisturizers, food hydrocolloids, and medical materials (Klemm et al. 2005; Basta and El-Saied 2009; Gallegos et al. 2016; Khosravi-Darani et al. 2016; Wang et al. 2017, 2018; Ling et al. 2018; Hussain et al. 2019). The first intermediate during BC biosynthesis is sub-elementary fibril with the diameter of 1–2 nm, which can form microfibrils with the diameter of 3–4 nm via a horizontal hydrogen bond linking (Yamanaka et al. 1989; Amano et al. 2005). After that, a cellulose ribbon with 30–100 nm in width and 3–8 nm in height is produced, and finally form the visible BC membranes (Yamanaka et al. 1989; Amano et al. 2005).

Since BC is an unbranched β-1,4-D-glucan composed of glucose, searching more low-cost glucose sources is a key step for efficient preparation of BC at a large-scale. To date, attempts to obtain alternative glucose for BC production have been made by several studies (Hussain et al. 2019). One promising way is to collect the carbon sources that contain high concentrations of glucose or those sources that can be easily biotransformed to glucose (Velásquez-Riaño and Bojacá 2017). Studies have also reported several chemical methods to obtain glucose from various byproducts produced in agricultural or industrial activities (Bae and Shoda 2004; Hong and Qiu 2008; Hong et al. 2012; Ye et al. 2019). While the biotransformation of various carbon sources to glucose is achieved by enzymes, the chemical transformation is often catalyzed by ionic liquors such as 1-allyl-3-methylimidazolium chloride ([AMIM]Cl) (Bentivoglio et al. 2006; Hong and Han 2011; Shill et al. 2011; Hong et al. 2012; Chen et al. 2013), 1-butyl-3-methylimidazolium cations ($[C_4mim]^+$) with a range of anions, from small, hydrogen-bond acceptors (Cl^-) to large, noncoordinating anions ($[PF_6]^-$) also including Br^-, SCN^-, and $[BF4]^-$ (Swatloski et al. 2002; Bentivoglio et al. 2006; Remsing et al. 2006; Kim et al. 2010), or the acids such as sulphuric acid and hydrochloric acid (Uraki et al. 2002; Hong and Qiu 2008; Yang et al. 2013). Due to the potential secondary pollution and relative high-cost of ionic liquors, using low-cost carbon sources containing high concentration of carbohydrates can be more ideal for BC production.

One popular low-cost carbon source used for BC production is fruit juice. For example, Lestari et al. (2014) showed that the pineapple juice from agricultural wastes was able to produce BC by 2.5 g L^{-1} d^{-1}. Other reported fruit juices include watermelon,

pawpaw, orange, apple, pear, grape, pomegranate, muskmelon, tomato, coconut and litchi (Kurosumi et al. 2009; Castro et al. 2011; Hungund et al. 2013; Lestari et al. 2014; Yang et al. 2016; Adebayo-Tayo et al. 2017). Among which, the muskmelon had the highest production ability of BC by 0.58 g L^{-1} d^{-1} (Hungund et al. 2013). Beside fruit juices, molasses or syrup (Bae and Shoda 2004; Keshk and Sameshima 2006; Moosavi-Nasab and Yousefi 2010; Li et al. 2015; Mohammadkazemi et al. 2015; Tyagi and Suresh 2016; Salari et al. 2019), tobacco waste extract (Ye et al. 2019), and other sources such as sisal juice (Lima et al. 2017) and waste glycerol (Kose et al. 2013) are also good candidates for BC production.

Sapindus mukorossi Gaertn. is a well-known tropical tree having pulpy fruit that lathers like soap. It is reported that the dry fruit of *S. mukorossi* contains 11.5% saponin (Kamra et al. 2006), making it possible for soap production. Besides saponin, *S. mukorossi* fruit also contains up to 10% of carbohydrate (Kamra et al. 2006), which should be removed before the use of saponin extract in soap production as the solution is very sticky. The most promising extractant for saponin extraction is ethanol (Wu et al. 2014). However, some carbohydrates can be dissolved in ethanol during the extraction process. As described, several bacteria can transform low-cost carbon sources to BC efficiently. We hypothesize that *S. mukorossi* extract may be a good carbon source for BC production, and the fermented solution without or with little carbohydrate can be used to produce the high-quality saponin. The full aims of this study were preliminary to (1) evaluate the potential use of the shell extract of *S. mukorossi* (SES) in BC production, (2) evaluate the effects of solution pH and incubation temperature on BC production ability in SES. Our study can provide a practice for ecofriendly green biosynthesis of BC by low-cost carbon source and a practice for pretreatment of *S. mukorossi* shell before its use in saponin extraction.

Materials and methods

Microorganism, culture media and cultivation

The microorganism used in this study was *Komagataeibacter xylinus* B2-1 (SRA accession number: PRJNA579797), a strain capable of BC production isolated from Kombucha. Hestrin–Schramm (HS) medium consisting of 2% glucose, 0.5% yeast extract, 0.5% peptone, 0.68% $Na_2HPO_4 \cdot 12H_2O$, 0.115% $C_6H_8O_7 \cdot H_2O$ and 0.051% $MgSO_4 \cdot 7H_2O$ was used as basic medium (Hestrin and Schramm 1954). The medium pH was adjusted to 6.0 using 1.0 M NaOH or HCl. The established medium was autoclaved at 115 °C for 30 min before use.

To obtain enough biomass, strain B2-1 was first incubated in HS medium at 30 °C for 24 h under agitated condition (180 rpm) and at 30 °C for 7 d under static condition (Wang et al. 2018). After that, the established suspension was centrifuged at 8000 × *g* for 5 min and re-suspended in 5 mL sterile Milli-Q water. This procedure was repeated three times to remove the residual glucose in the medium.

As to SES preparation, the procedures are detailed as below. Firstly, the fresh *S. mukorossi* fruits were collected from Fujian Sanqing *Sapindus* Technology Co., LTD and oven-dried to a balance weight. The dry fruits were separated into shells and seeds. The shells were dipped in water (w/w, 1:8) overnight at room temperature and boiled for 3 h. Following a centrifugation at 10,000 × *g* for 10 min to remove the shell residues, the supernatants were collected and autoclaved at 115 °C for 30 min after pH adjustment as previously described. To evaluate the potential ability of strain B2-1 in BC production by using SES as sole culture medium, established biomass (initial OD_{600} of 1.0) was transferred to a 250 mL Erlenmeyer flask, which contained 100 mL SES medium with 5% bacterial inoculation. All setups were incubated at 30 °C for 7 d under static condition.

BC purification and yield calculation

After 7 d of incubation, the medium pH was determined by a pH meter (PHS-3C, Shanghai Yidian Scientific Instruments Co., Ltd., China). The initial and final concentrations of total carbohydrate and glucose were also determined to evaluate the transformation rate of carbon sources to BC. Specifically, aliquots of SES were collected and centrifuged at 8000 × *g* for 5 min, followed by dilution by Milli-Q water as required. For total carbohydrate determination, 1 mL of supernatant was mixed with Milli-Q water to a total volume of 2 mL and subsequently mixed fully with 0.05 mL of 80% phenol. Finally, 5.0 mL of H_2SO_4 was rapidly added to the above

mixture and mixed again on a Vortex test tube mixer. While cooled to room temperature, the mixture was placed in a 25 °C bath for 10 min and mixed again before reading the absorbance at 490 nm (Nielsen 2010). For glucose analysis, 25 μL of established supernatant was determined on a biosensor analyzer (SBA-40E, Biology Institute of Shangdong Academy of Sciences, China) according to the manufacturer's instructions.

The BC membranes were collected and boiled at 100 °C for 2 h in a 0.1 M NaOH bath to eliminate bacterial cells and anther 2 h in a Milli-Q water bath to remove all residual chemicals (Wang et al. 2018). To remove the water held by BC membranes, the samples were oven-dried at 50 °C to a balance weight. The dried weight was recorded and used to calculate the BC yield by Eq. (1):

$$\text{Yield} = \frac{m_{ce}}{V} \tag{1}$$

where m_{ce} is the dry weight of BC (g) produced by strain B2-1 and V is the medium volume (L) used for BC production.

To have a full comparison with the reported BC productivity of representative carbon sources, the relative BC yield was also calculated and recorded as g L^{-1} d^{-1}.

BC characterization

A visible membrane on medium surface is a direct evidence of BC production. However, full understanding of BC properties needs further characterizations by physical and chemical methods. In our study, scanning electron microscopy (SEM, Quanta^{TM} 250 FEG, FEI, Hillsboro, OR, USA), X-ray diffraction (XRD, Bruker D8 ADVANCE, Karlsruhe, Germany) and Fourier transform infrared (FTIR, Thermo Scientific Nicolet iS5, Waltham, MA, USA) spectroscopy were used for BC characterization.

To have an in situ observation of BC, the samples were gold-sprayed for 45 s and mounted on an insulating tape. The established BC was observed by SEM with a spot of 3.0, high voltage of 15 keV and magnification of × 20,000. Besides microscopic morphology, the diameter distribution of BC nanofibrils was also calculated by using a Nano Measurer 1.2 (Fudan University, Shanghai, China). A total of 100 randomly-selected nanofibrils was grouped by each 10 nm and the normal distribution was calculated based on statistical histogram program in OriginPro 9.0 (OriginLab Corporation, Northampton, MA, USA).

Crystal analysis is very important for understanding the cellulose properties. X-ray diffraction has been widely used to characterize the crystal structure of BC (Terinte et al. 2011). In this study, the XRD analysis was conducted at a voltage of 40 kV and a filament emission of 40 mA, with 0.1° step, from 5° to 70° (2θ, angle) by using nickel filtered copper K_a radiation (λ = 0.15406 nm) (Wang et al. 2018). To subtract the background signal, a XRD analysis of the holder was also run as control. While the d-spacing between the crystal planes was determined using Bragg's law by Eq. (2), an apparent crystal size (ACS) approximation was determined using Scherrer's formula by Eq. (3) (Molina-Ramírez et al. 2017):

$$d = \frac{\lambda}{2\sin\theta} \tag{2}$$

$$\text{ACS} = \frac{0.9\lambda}{\text{FWHM} \ \cos\theta} \tag{3}$$

where λ is the X-ray wavelength, θ is the angle between the plane and the diffracted or incident beam (i.e., Bragg's angle), and FWHM is the width of the peak at half the maximum height. The multiple peak fit and FWHM calculation were performed by Gaussian function and Integrated Peaks analysis based on the Peaks and Baseline module in OriginPro 9.0.

To further evaluate the crystal property of BC, the crystallinity index (C.I.) was also calculated by Eq. (4) (Wang et al. 2018):

$$\text{C.I.} = \frac{I_{ma} - I_{am}}{I_{ma}} \tag{4}$$

where I_{ma} is the maximum diffraction intensity of the lattice peak between 2θ angle of 22° to 24° and I_{am} is the diffraction intensity of the amorphous phase at around 2θ angle of 18° to 19°.

In addition to BC crystallinity, the typical functional groups of BC were also analyzed by FTIR. Since the BC is difficult to be powdered, the attenuated total reflection (ATR) mode with 32 scans per measurement and a resolution of 0.5 cm^{-1} ranging from 4000 to 500 cm^{-1} was used in this study (Wang et al. 2017). After baselines normalization, the cellulose I_{α} content was calculated by Eq. (5) (Yamamoto et al. 1996):

$$f_{\alpha}^{IR} = \frac{A_{\alpha}}{A_{\alpha} + A_{\beta}} \quad (5)$$

where A_{α} and A_{β} are the integrated intensities of celluloses I_{α} and I_{β} at 750 and 710 cm^{-1}, respectively.

Effects of pH and temperature on BC production

To evaluate the potential roles of pH and temperature in BC production, 5% inoculation of established biomass (initial OD_{600} of 1.0) was transferred to SES medium. The pHs used in this study were in range of 4–8, while the temperatures were 25, 30 and 35 °C. After 7 d of incubation, the medium pH and carbohydrate concentrations were determined, and the BC yields were calculated as previously described.

Statistical analysis

All experiments were conducted in triplicate. The data are presented as the mean value of the triplicate with standard error. Significant differences were determined according to two-way analysis of variance (ANOVA) by Tukey's multiple comparisons test at $p \leq 0.05$ using GraphPad Prism (Release 6.0, La Jolla, CA, USA).

Results and discussions

BC production by using SES as sole culture medium

It is well-known that BC has an unbranched β-1,4-D-glucan structure consisting of glucose (Reiniati et al. 2017; Krasteva et al. 2018). Besides glucose, the precursor for BC biosynthesis, other carbohydrates such as fructose, lactose, maltitol, sucralose, xylitol, glycerol, sucrose and galactose can also be transformed to glucose and then to form BC (Wang et al. 2018). There has so far been lots of attempts to utilize various low-cost carbon sources as substrates for BC production (Velásquez-Riaño and Bojacá 2017). Due to the high content of carbohydrates in *S. mukorossi* shell, it can also serve as an important candidate for BC biosynthesis, thereby improving the saponin quality by a ecofriendly way.

In our study, the carbohydrates of *S. mukorossi* were extracted by boiling water. As shown in Fig. 1A, the initial carbohydrate concentration in SES was 17.3 mg L^{-1}, with 2.0 mg L^{-1} of which being glucose. After 7 d of incubation, 28% of total carbohydrate reduced (Fig. 1A). However, the glucose concentration decreased by 60%, indicating the preferrence of glucose utilization although strain B2-1 also utilized other carbohydrates (Fig. 1A). Similar to the case of HS medium, an apparent membrane on SES surface was observed (Fig. 1B). Strain B2-1 is a typical acetic acid bacterium often used for acetic fermentation (Valera et al. 2015; Barja et al. 2016), explaying why the medium pH of SES reduced from 6.0 to 5.1 (Fig. 1C). Moreover, the BC yield was up to 1.31 g L^{-1} (Fig. 1D), lower than the yield obtained in HS medium (~ 1.8 g L^{-1}, data not shown). However, the data was comparable to Komagataeibacter sp. W1 (~ 1.5 g L^{-1}) after 14 d of incubation in HS meidum in our previous study (Wang et al. 2018). It was also worthy to note that some unknown carbohydrate loss was found based on mass balance (Fig. 1A, D), probably due to the consumption of carbohydrates for bacterial cell growth and multiplication (Saxena and Brown Jr 2013).

To have a full understanding of BC productivity in strain B2-1 by using SES as sole culture medium, more than 35 low-cost carbon sources used for BC production and the corresponding BC yields were summarized. These carbon sources can be classified into broad categories as fruit juices or wastes, date sugars or sugar wastes, biomass hydrolysates, biomass extracts, and other carbon wastes (Table 1). Among reported fruit juices or wastes, muskmelon, watermelon and grape juice obtained the top three yield of BC by 0.58, 0.56 and 0.50 g L^{-1} d^{-1}, respectively (Table 1). Molasses is a viscous and dark liquid by-product of sugar beet (*Beta vulgaris* var. *saccharifera*) or sugar cane (*Saccharum* L.) containing considerable carbohydrates (e.g., sucrose, glucose and fructose), organic matters such as betaine and amino acids, minerals and trace elements, and vitamins (Varaee et al. 2019). All these components can benefit BC production (Noro et al. 2004; Keshk 2014), which is supported by the fact that molasses have shown considerable BC yields up to 1.78 g L^{-1} d^{-1} (Table 1). However, other date sugars or sugar wastes only resulted in 0.17–0.45 g L^{-1} d^{-1} of BC yields (Table 1). Unlike the above two types of carbon sources, biomass hydrolysates and extracts are obtained after chemical and water pretreatment.

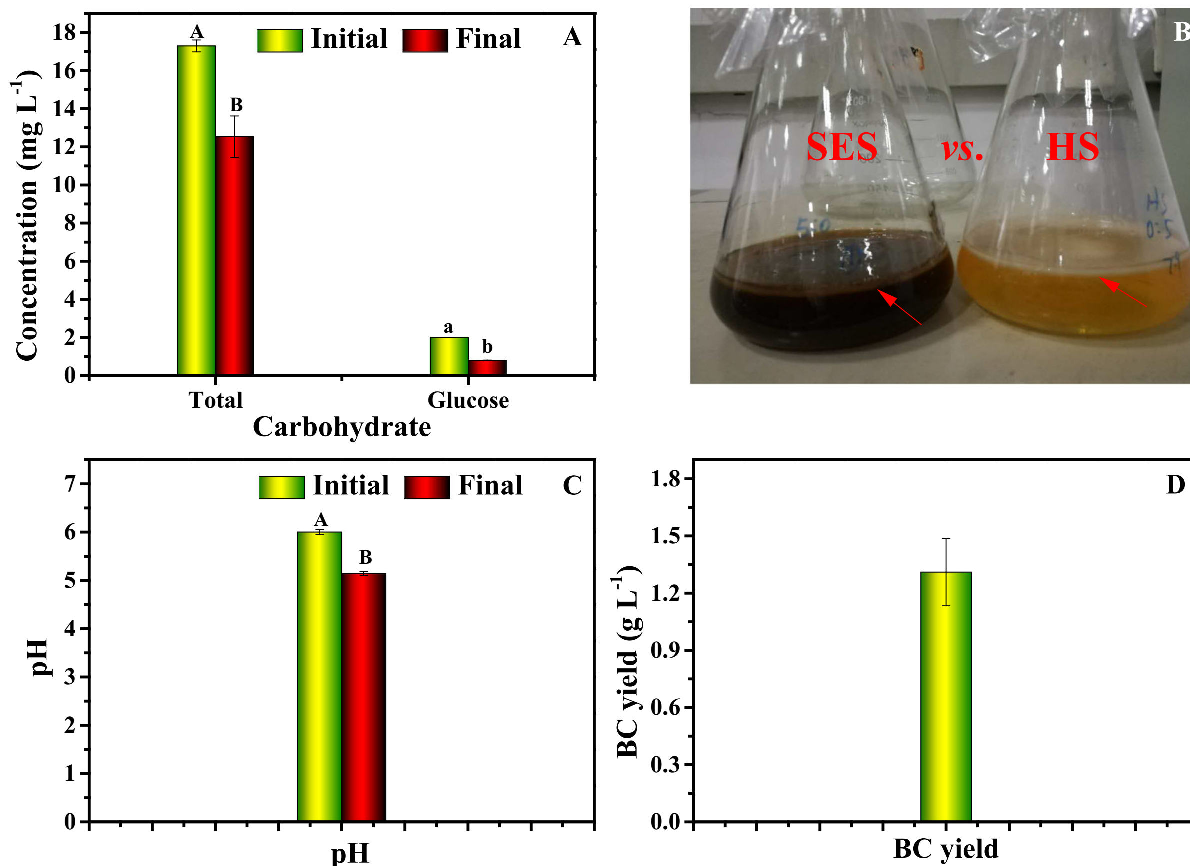


Fig. 1 The total carbohydrate and glucose concentrations **A** in SES before and after 7 days incubation with *K. xylinus* B2-1. **B** visible BC on SES and HS media surface. **C** SES medium pH before and after bacterial incubation. **D** BC yield. Different letters indicate no significant difference between the setups according to Limited Slip Differential (LSD) test ($p \leq 0.05$)

Correspondingly, the BC yields are up to 1.9 and 0.74 g L^{-1} d^{-1}, respectively (Table 1). The data indicated that biomass extracts were also good carbon sources for BC production.

As described, strain B2-1 efficiently utilized the carbohydrates in SES for BC biosynthesis without any additives (Fig. 1). The relative BC yield was 0.21 g L^{-1} d^{-1}, which was double as that of pecan nutshell extract spiked with lots of organic matters and that of dry olive mill residue (Table 1). This was likely associated with the high contents of amino acids and fatty acids in *Sapindus* (Yin et al. 2011; Lovato et al. 2014), which warranted further investigations. Although the BC yield in SES medium was lower than tobacco waste extract and coffee cherry husk extract, the incubation time in our study was much shorter than other studies. Our data indicated that *S. mukorossi* shell extract could serve as a good alternative carbon source for ecofriendly BC production.

BC characterization by SEM, XRD and FTIR

Ribbon (30–100 nm in width and 3–8 nm in height), microfibril (diameter of 3–4 nm) and sub-elementary fibril (diameter of 1–2 nm) are three substructures of BC (Yamanaka et al. 1989; Amano et al. 2005). To evaluate the morphology and microstructure of BC produced from SES, a SEM observation was conducted, followed by a further calculation of the diameter distribution based on statistical histogram program analysis in OriginPro 9.0. As can be seen in Fig. 2A, the BC produced from SES consisted of pellicles by a layer-by-layer assembly manner. The data also showed that most observed nanofibrils were below 100 nm, with an average diameter of 40–50 nm

Table 1 The low-cost carbon sources used for BC production and BC productivity in representative bacterial strains

Bacterial names[a]	Carbon sources	Conditions	Substrate concentrations (g L^{-1})	BC yields		References
				g L^{-1}	g L^{-1} d^{-1}	
Fruit juices or wastes						
Acetobacter pasteurianus PW1	Watermelon juice medium	Static, 28–30 °C, 15 d	N.M.[b]	8.41	0.56	Adebayo-Tayo et al. (2017)
Acetobacter pasteurianus RSV-4	Tomato juice and orange pulp	Static, 30 °C 7 d	20	7.8 and 2.8	0.39 and 0.14	Kumar et al. (2019)
Acinetobacter sp. BAN1	Pawpaw juice medium	Static, 28–30 °C, 15 d	N.M.	6.48	0.43	Adebayo-Tayo et al. (2017)
Acetobacter xylinum	Coconut water, pineapple juice	Static, 28, 12 d	N.M.	up to 50[c]	up to 4.2	Lestari et al. (2014)
Acetobacter xylinum LKN6	Sago liquid waste	Static, 30 °C, 14 d	N.M.	4.12	0.29	Yanti et al. (2017)
Acetobacter xylinum NBRC 13693	Orange, apple, pineapple, Japanese pear, grape	30 °C, 14 d	62–103 of total carbohydrate, 13–42 of Glu, 12–59 of Fru, 2–49 of Suc[d]	up to 5.9	up to 0.42	Kurosumi et al. (2009)
Acetobacter xylinum TISTR975	Pineapple peels juice	28 °C, 7 d	N.M.	N.M.	N.M.	Saowapark et al. (2017)
Gluconacetobacter persimmonis GH-2	Fruit juices including pineapple, pomegranate, muskmelon, water melon, tomato, orange, molasses, starch hydrolyzate, sugarcane juice, coconut water, coconut milk	30 °C, 14 d	20 of total carbohydrate (after normalization)	up to 8.08	up to 0.58	Hungund et al. (2013)
Gluconacetobacter sp.	Grape juice (mixed with 5% Suc.)	Static, room temperature, 14 d	17.6 of total carbohydrate, 13 of reducing sugar	7.47	0.50	Rani et al. (2011b)
Gluconacetobacter sp. gel_SEA623-2	Citrus fruit juice (unshiu, orange, grape, apple, pear)					Kim et al. (2017)
Gluconacetobacter swingsii	Pineapple peel juice	Static, 28 °C, 8 d	21.4 of Glu, 24 of Fru, 21 of Suc	2.8	0.35	Castro et al. (2011)
Gluconacetobacter xylinus ATCC 53582	Milk whey, rotten fruit (plums, green grapes, pineapples, and apples)	Static, 30 °C, 4 d	10–20 of total carbohydrate	up to 60[c]	up to 15	Jozala et al. (2015)
Gluconacetobacter xylinus CH001	Litchi extract	Static, 28 °C, 14 d	N.M.	2.53	–[f]	Yang et al. (2016)
Date sugars or sugar wastes						

Table 1 continued

Bacterial names[a]	Carbon sources	Conditions	Substrate concentrations (g L^{-1})	BC yields		References
				g L^{-1}	g L^{-1} d^{-1}	
Acetobacter lovaniensis HBB5	Beet molasses	Static, 30 °C, 7 d	N.M.	0.021	0.003	Çoban and Biyik (2011)
Acetobacter pasteurianus HBB6	Beet molasses	Static, 30 °C, 7 d	N.M.	0.029	0.004	Çoban and Biyik (2011)
Acetobacter pasteurianus RSV-4	Cane molasses	Static, 30 °C 7 d	20	3.6	0.18	Kumar et al. (2019)
Acetobacter xylinum ATCC 10244, IFO 13693, 13772, 13773, 14815, and 15237	Sugar cane molasses	Static, 28 °C, 7 d	N.M.	1.24–5.99	0.18–0.86	Keshk and Sameshima (2006)
Acetobacter xylinum BPR2001	Molasses	Agitated, 30 °C, 3 d	56 of Fru, 48 of Glu	up to 5.3	up to 1.78	Bae and Shoda (2004)
Gluconacetobacter intermedius SNT-1	Sugarcane molasses	Static, 30 °C, 10 d	45.8	12.6	1.26	Tyagi and Suresh (2016)
Glucanoacetobacter xylinum ATCC 23768	Black strap molasses	Agitated, 30 °C, 9 d	N.M.	2.9	0.32	Khattak et al. (2015)
Glucanoacetobacter xylinum ATCC 23768	Brewery molasses	Agitated, 30 °C, 9 d	N.M.	1.74	0.19	Khattak et al. (2015)
Gluconacetobacter xylinum CGMCC No. 2955	Candied jujube wastewater	Static, 30 °C, 6 d	38 of Glu in hydrolysate	2.25 (1.5)[g]	0.45	Li et al. (2015)
Gluconacetobacter xylinus CH001	Acetone-butanol-ethanol fermentation wastewater (5% of sugars before fermentation)	Static, 28 °C, 8 d	Glu, Xyl	1.34	0.19[e]	Huang et al. (2015)
Gluconacetobacter xylinus FC01	Molasses	Static, 30 °C, 6 d	N.M.	0.57	0.095	Çakar et al. (2014)
Gluconacetobacter xylinus PTCC 1734	Date syrup	Agitated, 28 °C, 7 d	N.M.	up to 1.2	up to 0.17	Mohammadkazemi et al. (2015)
Gluconacetobacter xylinus PTCC 1734	Date syrup	Static, 28 °C, 15 d	N.M.	3.02	0.22[e]	Moosavi-Nasab and Yousefi (2010, 2011)
Gluconacetobacter xylinus PTCC 1734	Sugar beet molasses and cheese whey media	Static, 28 °C, 14 d	20–40 of total carbohydrate	4.56 and 3.55	0.33 and 0.25	Salari et al. (2019)
Biomass hydrolysates ((bio-)chemical extraction)						
Acetobacter aceti subsp. *xylinus* ATCC 23770	Konjac powder hydrolysate	Static, 30 °C, 8 d	>20 of total carbohydrate	up to 2.12	up to 0.27	Hong and Qiu (2008)
Acetobacter xylinum KJ1	Enzymatic extract of food wastes	Agitated, 30 °C, 3 d	N.M.	5.7	1.9	Song et al. (2009)

Table 1 continued

Bacterial names[a]	Carbon sources	Conditions	Substrate concentrations (g L^{-1})	BC yields		References
				g L^{-1}	g L^{-1} d^{-1}	
Gluconacetobacter hansenii CGMCC 391	Waste beer yeast	Static, 30 °C, 14 d	43.8 of total carbohydrate	up to 7.02	0.7[e]	Lin et al. (2014)
Gluconacetobacter xylinus	Lipid fermentation wastewater (containing corncob acid hydrolysate and 5.5% of sugars before fermentation)	Static, 28 °C, 10 d	N.M.	0.66	0.13[e]	Huang et al. (2016)
Gluconacetobacter xylinum ATCC 10245	Potato peel wastes acid hydrolysate	Static, 30 °C, 2–6 d	N.M.	4.7	0.78	Abdelraof et al. (2019)
Gluconacetobacter xylinum ATCC 23770	Cotton cloth hydrolysate	Static, 30 °C, 7–14 d	17 (normalized to Glu)	10.8	–[f]	Hong et al. (2012)
Gluconacetobacter xylinus CH001	Elephant grass (*Pennisetum purpureum*) acid hydrolysate	Static, 28 °C, 14 d	12 of Glu, 20 of Xyl, 2.3 of Ara	6.4	0.46	Yang et al. (2013)
Komagataeibacter rhaeticus	Cashew tree residues	Static, 28 °C, 7 d	N.M.	2.8	0.4	Pacheco et al. (2017)
Komagataeibacter sucrofermentans DSM 15973	Crude glycerol and sunflower meal hydrolysates	Static, 30 °C, 15 d	0.8 of Suc, 18.6 of Glu, 7.5 of Fru (sunflower meal)	13–13.3	0.87–0.89	Tsouko et al. (2015)
Komagataeibacter xylinus CH001	Biomass acid hydrolysate	Static, 28 °C, 10 d	N.M.	2.9	0.73[e]	Luo et al. (2017)
Medusomyces gisevii Sa-12	Enzymatic hydrolysates of oat hulls	Static, 27 °C, 9 d	20 (normalized to Glu)	N.M.	N.M.	Aleshina et al. (2018)
Biomass extracts (water extraction)						
Acetobacter xylinum 23769	Red Maple (Acer rubrum) strands	Static, 28 °C, 28 d	Glu., Xyl.	0.15	0.0054	Kiziltas et al. (2015)
Acetobacter xylinum ATCC 23767	Tobacco waste extract	Agitated, 30 °C, 7 d	up to 22.4 of total carbohydrate, 10.7 of Glu, 4.29 of Fru, 2.94 of Suc, 1.8 of Man_o, 0.32 of Xyl, 0.23 of Gal, 0.16 of Ara	up to 5.2	0.74	Ye et al. (2019)
Gluconacetobacter entanii	Pecan (*Carya illinoinensis*) nutshell	Static, 30 °C, 28 d	40 (reduced carbohydrate)	2.82	0.1	Dórame-Miranda et al. (2019)
Gluconacetobacter hansenii UAC09	Coffee cherry husk	Static, 27 °C, 14 d	N.M.	5.6–8.2	0.4–0.59	Rani et al. (2011a); Rani and Appaiah (2013)_ENREF_58

Table 1 continued

Bacterial names[a]	Carbon sources	Conditions	Substrate concentrations (g L^{-1})	BC yields		References
				g L^{-1}	g L^{-1} d^{-1}	
Gluconacetobacter sacchari	Dry olive mill residue	Static, 30 °C, 8 d	11.9 of total carbohydrate, 10 of Glu, 1.65 of Fru, 0.25 of Xyl	0.85	0.11	Gomes et al. (2013)
Komagataeibacter xylinus B2-1	Shell extract of *Sapindus mukorossi* Gaertn.	Static, 30 °C, 7 d	17.3 of total carbohydrate, 2 of Glu	1.47	0.21	**This study**
Other carbon wastes						
Consortium of acetic acid bacteria and yeasts	Brew of tea leaves	Static, 30 °C, 2–20 d	20–100 of Glu (spiked)	up to 13.3	0.64–2.4	Sharma and Bhardwaj (2019)
Gluconacetobacter intermedius NEDO-01 (NITEP-1495)	Waste glycerol	Agitated, 30 °C, 4 d	N.M.	3.4	0.85	Kose et al. (2013)
Gluconacetobacter oboediens MTCC 5610	Crude distillery effluent	Static, 30 °C, 8 d	N.M.	8.5	1.1	Jahan et al. (2018)
Komagataeibacter hansenii ATCC 23769	Sisal juice	Static, 30 °C, 2–22 d	15.1 of total carbohydrate with 8.2 of Glu, 1 of Suc, 0.8 of Fru	up to 3.38	0.34[e]	Lima et al. (2017)
Komagataeibacter saccharivorans BC1	Crude distillery effluent	Static, 30 °C, 8 d	20 of Man_i	1.24	0.16	Gayathri and Srinikethan (2019)
Komagataeibacter sp. PAP1	Soya bean whey	Static, 30 °C, 7 d	2.6 of total carbohydrate with 0.2 of Lac	4.35	0.62	Suwanposri et al. (2014)

[a]It is worthy to note that the bacterial names *A. xylinum* and *G. xylinus* have been reclassified as *K. xylinus* now

[b]Not mentioned

[c]Whether the data was dry weight or fresh weight was not given

[d]Glu, glucose; Fru, fructose; Xyl, xylose; Gal, Galactose; Man_o, Mannose; Suc, sucrose; Ara, arabinose; Man_i, mannitol; Lac, lactose

[e]The maximum yield calculated before the final day

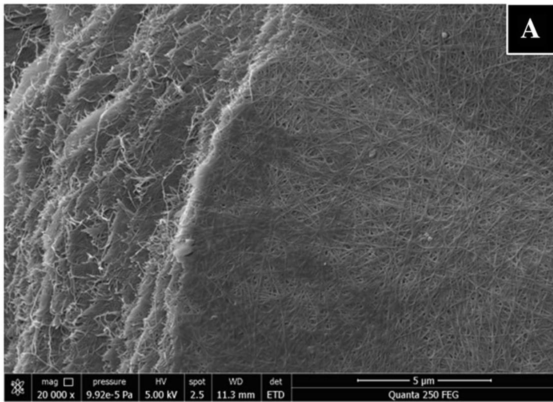
[f]Because the total volume was not mentioned or the incubation time was unclear, the relative yields could not be calculated successfully

[g]The BC yield was 1.5 if the candied jujube wastewater was not hydrolyzed before use

(Fig. 2B). It was apparent that the fibrils observed by SEM were ribbons, similar to the BC produced from HS medium (Fig. 2C–D).

As a typical cellulose, BC also has ordered crystalline and less ordered amorphous regions to form a homogeneous polycrystalline structure (Bi et al. 2014). While the ordered crystalline often forms three diffraction peaks at around 2θ of 14.5°, 16.6° and 22.7°, the amorphous peak can be found at around 2θ

Fig. 2 Representative characteristics of the BC produced by *K. xylinus* B2-1 grown in SES (**A–B**) and HS medium (**C–D**) after 7 d incubation. **A**, **C** the BC morphology observed with SEM with a spot of 3.0, high voltage of 15 keV, and magnification of × 20,000. **B–D** the BC diameter calculated by Nano Measurer 1.2 with 100 random selection of the BC nanofibrils on SEM images. **E** The XRD analysis performed by using nickel filtered copper K_α radiation, with 0.1° step, from 5° to 70° (2θ). **F** The FTIR analysis performed on using a Nicolet iS5 in the ATR mode with 32 scans per measurement between 4000 and 500 cm^{-1} ▶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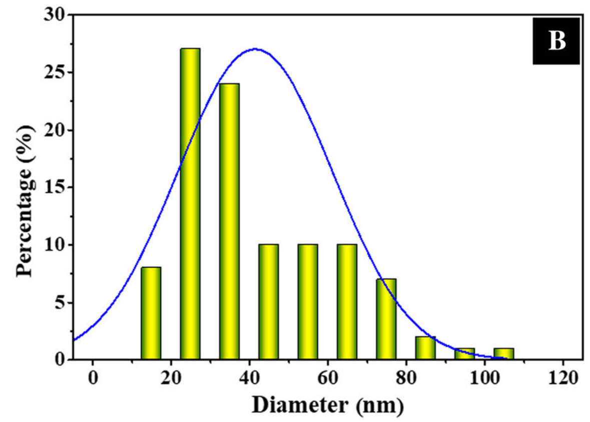
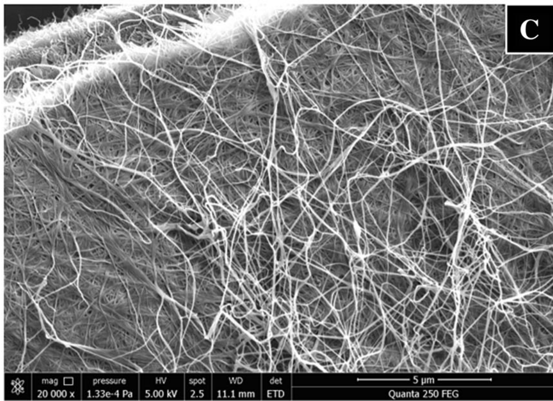

B
Percentage (%)
Diameter (nm)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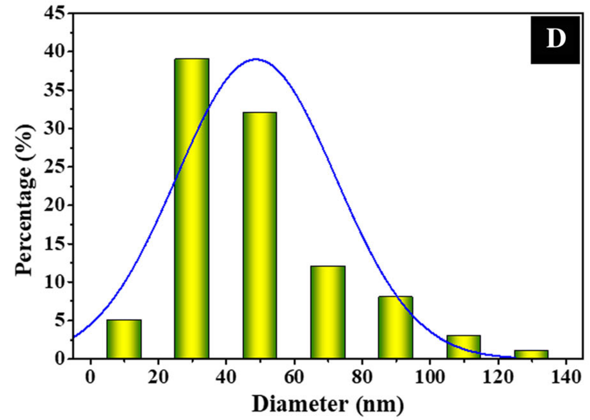
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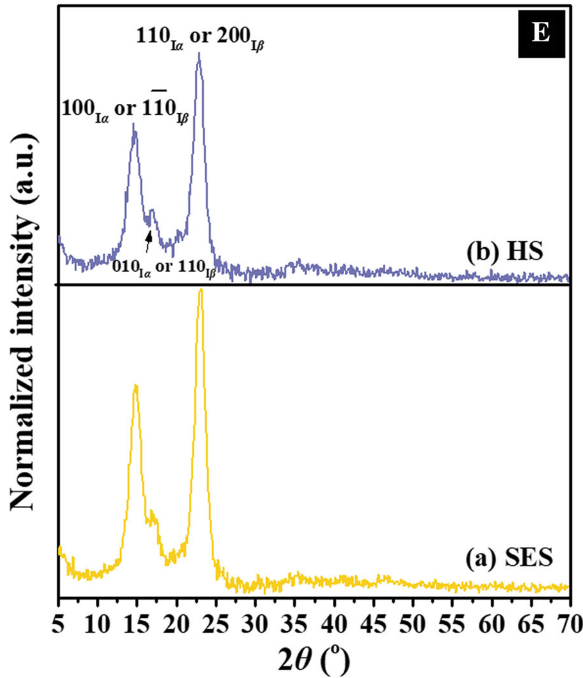
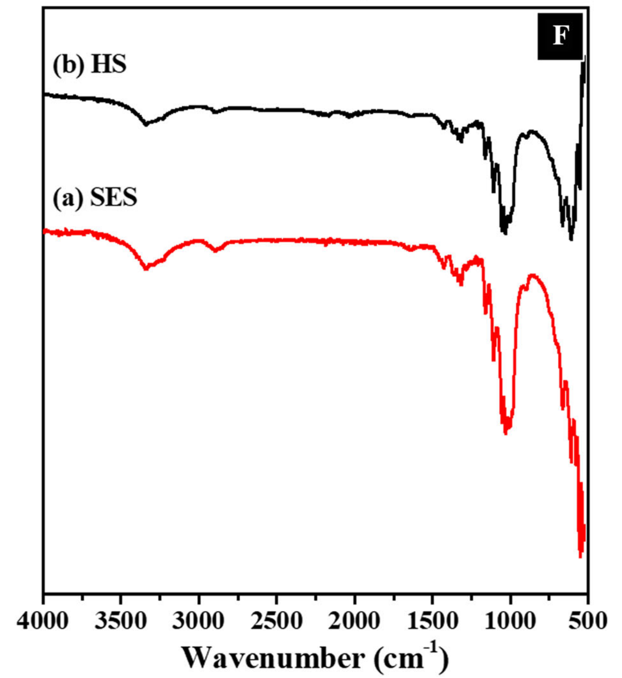

E
110Iα or 200Iβ
100Iα or 1̄10Iβ
010Iα or 110Iβ
(b) HS
(a) SES
Normalized intensity (a.u.)
2θ (°)


F
(b) HS
(a) SES
Wavenumber (cm-1)

of 18° to 19° (Wang et al. 2018). All the information can be obtained by XRD analysis. As expected, the BC produced from SES in our study displayed two apparent diffraction peaks at 2θ of 14.5° and 23.2° with strong intensity and a weak peak at 2θ of 16.8° (Fig. 2E-a). This indicates the presence of type I crystalline cellulose rather than type II cellulose as the latter one has two typical peaks at around 12° and 20° (Chen et al. 2011; de Marco Lima et al. 2011). By comparing to typical BC produced from HS medium, all the peaks are overlapped (Fig. 2E-b), again showing the good potential of SES to produce high-quality BC. As shown in Fig. 2E, the peaks at 2θ of 14.5° and 23.2° can be assigned to (100) and (110) planes of cellulose I_α or (1 $\bar{1}$ 0) and (200) planes of cellulose I_β, while the peak at 2θ of 16.8° is probably associated with (010) plane of cellulose I_α or (110) plane of cellulose I_β (French, 2014; Wang et al. 2018). However, the relative content of I_α and I_β cannot be calculated based on XRD data due to the second dimension of BC crystallites and the overlap of cellulose I_α and I_β reflections (Tokoh et al. 1998; Bi et al. 2014).

To have a further analysis of BC crystalline structure and types, we also determined the *d*-spacing, ACS and C.I. based on XRD data. As shown in Table 2, the *d*-spacing corresponding to each peak was same for both SES- and HS-produced BCs, similar to previous studies (Castro et al. 2011; Tyagi and Suresh 2016; Wang et al. 2018), implying the same I_α contents between the two samples (Kiziltas et al. 2015). Unlike *d*-spacing, however, the ACSs of two samples were different (Table 2), probably due to the complex components of SES and the subsequent attachment of the components to BC. It was also interesting to note that the lower ACS corresponded to the higher BC crystallinity (Table 2), which was supported by Meza-Contreras et al. (2018). Our data showed that SES was a good source for preparation of BC with high crystallinity.

Besides SEM and XRD analyses, we also used FTIR to characterize BC properties. The FTIR spectra exhibited several vibration bands as reported by previous studies (Fig. 2F). For example, the typical adsorptions at around 3345 cm^{-1} (O–H stretching), 2900 cm^{-1} (C − H stretching), 1430 and 1335 cm^{-1} (O–H in-plane bending), 1360, 1280 and 1205 cm^{-1} (O–H bending), 1160 cm^{-1} (C − O–H antisymmetric bridge stretching of 1, 4-β-glucoside), 1108, 1055 and 1031 cm^{-1} (C − O bending), 900 cm^{-1} (antisymmetric out-of-phase ring stretching of β-glucosidic linkages between glucose units) and below 660 cm^{-1} (O–H out-of-phase bending) were observed (Fig. 2F). Among of which, the vibration bands at 1430, 1335 and 1108 cm^{-1} can also be assigned to CH_2 symmetric bending, C − H deformation and C–C bonds of the monomer units of polysaccharide, respectively, while the adsorptions at 1055 and 1031 cm^{-1} are also associated with C–O–C pyranose ring skeletal vibration (Wang et al. 2017, 2018). We hypothesized that the BC produced from SES mainly composed of cellulose I due to the presence of adsorptions at around 3345, 1430, 1160 and 900 cm^{-1} (Wang et al. 2017). As noted previously, due to the overlap of cellulose I_α and I_β reflections, XRD analysis cannot differentiate the two allomorphs, i.e., around 3240 and 750 cm^{-1} for I_α allomorph and around 3270 and 710 cm^{-1} for I_β allomorph (Molina-Ramírez et al. 2017). By FTIR analysis, the I_α fractions of BCs produced from HS and SES were 0.41 and 0.45, respectively, in line with the studies of Keshk and Sameshima (2006) and Kiziltas et al. (2015). Our data concluded that the BC produced from SES was mainly composed of cellulose I, being 45% of cellulose I_α.

Effects of pH and temperature on BC production

Studies have shown that several factors such as carbon source, ethanol addition, temperature, pH and substrate mass transfer are important for BC biosynthesis

Table 2 *D*-spacing, ACS and C.I. of BC samples produced from the SES and HS media

Media	Peak 1 ($100_{I\alpha}$ or $1\bar{1}0_{I\beta}$)		Peak 2 ($010_{I\alpha}$ or $110_{I\beta}$)		Peak 3 ($110_{I\alpha}$ or $200_{I\beta}$)		Diffraction intensities at 2θ scale		C.I.
	d-spacing (nm)	ACS (nm)	*d*-spacing (nm)	ACS (nm)	*d*-spacing (nm)	ACS (nm)	I_{am}	I_{ma}	
SES	0.60	7.39	0.53	11.0	0.38	8.64	123	1646	0.93
HS	0.61	6.54	0.52	7.89	0.39	8.32	142	1242	0.89

(Hornung et al. 2006; Hutchens et al. 2007; Çakar et al. 2014; Penttilä et al. 2016). In general, the carbon sources containing high content of carbohydrate, the higher ethanol addition and substrate mass transfer rate can result in more BC production. However, the solution pH and incubation temperature are more strict conditions during bacterial growth and BC production. In this study, the effects of medium pH and incubation temperature on BC production are evaluated.

Figure 3A, B show the effects of medium pHs on carbohydrate utilization and BC production. Generally, strain B2-1 was able to utilize the carbohydrates at all pH gradients. The highest decrease of total carbohydrate and glucose, from 17.2 and 2.0 mg L^{-1} to 12.5 and 0.8 mg L^{-1}, respectively, was observed at pH 6.0 (Fig. 3A). However, the corresponding BC yield showed different trends from carbohydrate consumption. For example, pH 5.0 resulted in the highest BC yield of 1.35 g L^{-1}, followed by 1.31 g L^{-1} at pH 6.0 and 1.07 g L^{-1} at pH 4.0 (Fig. 3B). Our data was similar to Çoban and Biyik (2011) and Çakar et al. (2014) but different from Jagannath et al. (2008) in that *A. xylinum* NCIM 2526 preferred to grow and produce BC at pH of 4.0. According to the results from Fig. 3A and B, we hypothesized that the optimal pH for bacterial growth and BC production in strain B2-1 was between 5.0 and 6.0. Moreover, strain B2-1 could grow well at pH > 6.0 although the BC production was inhibited (Fig. 3). Apparently, medium pH bear important roles in BC production from SES in strain B2-1, in accordance with previous studies (Jagannath et al. 2008; Çoban and Biyik 2011).

In addition to medium pH, we also evaluated the effects of incubation temperature on BC production. As shown in Fig. 3C, the significant carbohydrate utilization occurred at temperature of 30 °C, by a decrease of 24% and 47% of total carbohydrate and glucose, respectively (Fig. 3C). Correspondingly, the BC yield was 1.47 g L^{-1} (Fig. 3D). At low temperature, it was probably that strain B2-1 preferred to utilize glucose as carbon source for BC production, but the transformation rate of carbohydrate to BC was very low (i.e., 0.42 g L^{-1}; Fig. 3C, D), which might

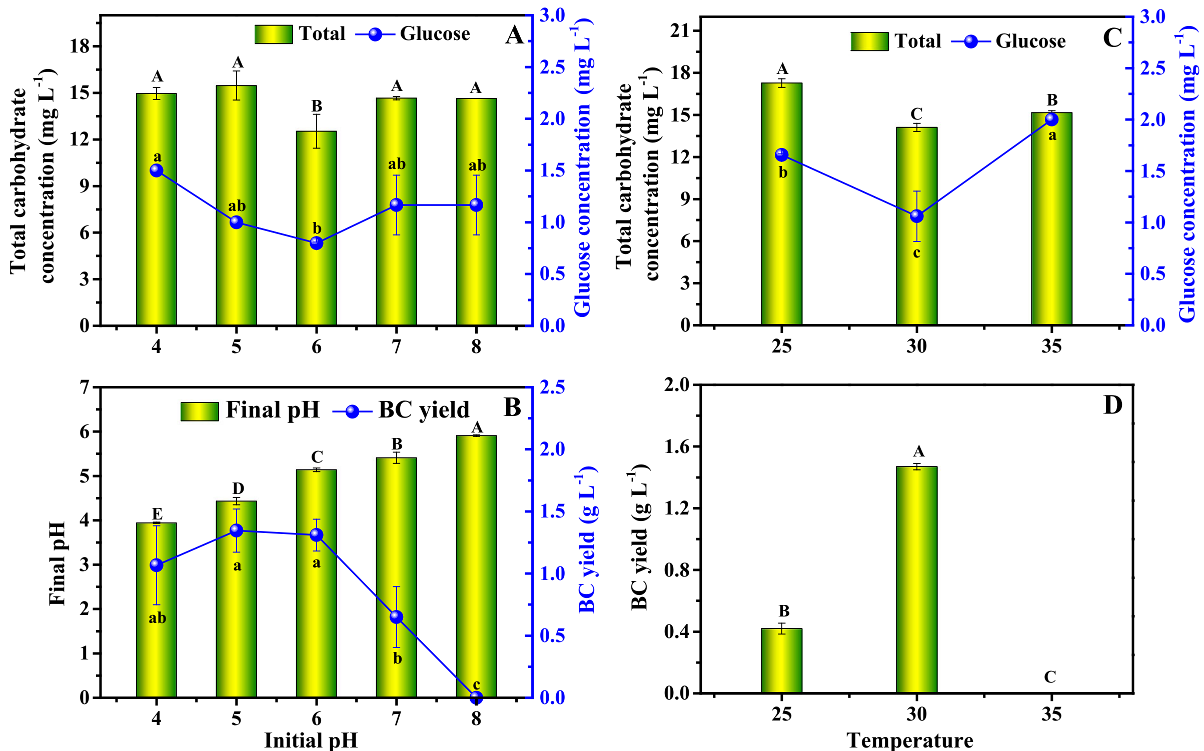


Fig. 3 Effects of initial pH (**A–B**) and temperature (**C–D**) on BC production by *K. xylinus* B2-1 grown in SES medium after 7 d incubation. **A**, **C** the total carbohydrate and glucose concentrations and **B**, **D** final pH and BC yield after 7 d incubation with different initial pH and temperature, respectively

be due to the slow synthesis rate influenced by decreased thermal motion (Penttilä et al. 2016). However, high temperature can result in protein denaturation and aggregation (Penttilä et al. 2016), explaining why no BC production was observed in our study at temperature of 35 °C (Fig. 3D). Unlike strain B2-1, strains *K. xylinus* ATCC 53524, *A. pasteurianus* HBB6 and *A. lovaniensis* HBB5 in previous studies grew well and produced BC efficiently at temperature of 35 °C or higher (Çoban and Biyik 2011; Penttilä et al. 2016), implying that strains have different sensitivity to environmental temperature during bacterial growth and BC production.

Taken together, our study has shown the good potentials of *S. mukorossi* shell as a good low-cost carbon source in extracting carbohydrate and preparing high-quality BC. The BC productivity in SES was similar to HS medium, so did the BC properties. Due to the large biomass of *S. mukorossi* shell, the high concentration of carbohydrates in SES, and the potential use of SES saponin in soap production, further study should pay more attentions to the integrated utilization of *S. mukorossi* fruits. One of the future attempts is to produce BC efficiently at a pilot- and large-scale. Moreover, a full evaluation of the saponin quality is very important as BC producing bacteria may have altered the byproducts during the long-term fermentation.

Conclusions

A new low-cost carbon source with high carbohydrates from *S. mukorossi* shell was successfully transformed to BC by *K. xylinus* B2-1. The prepared BC was of high purity and showed no differences from that from HS medium. The optimized pHs and temperature for BC production were from 4.0 to 5.0 and at 30 °C, respectively. It was apparent that the SES was a good alternative low-cost carbon source for ecofriendly green biosynthesis of BC. Our study also provided a good method to reduce carbohydrates in SES before its application in detergent development.

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Compliance with ethical standard

Conflict of interest The authors declare that they have no conflict of interests.

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