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Atmospheric cold plasma-assisted pineapple peel waste hydrolysate detoxification for the production of bacterial cellulose

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ABSTRACT

Toxic compounds in pineapple peel waste hydrolysate (PPWH), namely formic acid, 5-hydroxymethylfurfural (HMF), and furfural, are the major predicament in its utilization as a carbon source for bacterial cellulose (BC) fermentation. A rapid detoxification procedures using atmospheric cold plasma (ACP) technique were employed to reduce the toxic compounds. ACP treatment allows the breakdown of toxic compounds without causing excessive breakdown of sugars. Herein, the performance of two available laboratory ACP reactors for PPWH detoxification was being demonstrated. ACP-reactor-1 (R1) runs on plasma power of 80–200 W with argon (Ar) plasma source, while ACP-reactor-2 (R2) runs at 500–600 W with air plasma source. Treatment in R1, at 200 W for 15 min, results in 74.06%, 51.38%, and 21.81% reduction of furfural, HMF, and formic acid. Treatment in R2 at 600 W gives 45.05%, 32.59%, and 60.41% reductions of furfural, HMF, and formic acid. The BC yield from the fermentation of *Komagateibacter xylinus* in the R1-treated PPWH, R2-treated PPWH, and untreated-PPWH is 2.82, 3.82, and 2.97 g/L, respectively. The results show that ACP treatment provides a novel detoxified strategy in achieving agricultural waste hydrolysate reuse in fermentation. Furthermore, the results also imply that untreated PPWH can be an inexpensive and sustainable resource for fermentation media supplementation.

1. Introduction

Agro-industrial waste is an atypical renewable resource that has the potential to be processed into various valuable materials. The utilization and conversion of waste into value-added products is one smart solution in overcoming waste accumulation [1]. Fruit peel waste, such as pineapple peel waste (PPW), is one of the many agro-industrial wastes generated from open markets and other food-related industries [2]. The use of agro-industrial waste as a source of nutrients for bacterial cellulose (BC) fermentation can be a great solution to relieve PPW accumulation. The major predicament that inhibits the direct-utilization of PPW in BC fermentation is the high lignocellulose content. Lignocellulosic material is mainly composed of the carbohydrate-polymers (i.e., cellulose and hemicellulose) and layers of lignin. This material is responsible for the rigidity of the biomass cell walls. These lignin layers cover and protect the cellulose and hemicellulose thus makes it inaccessible for microbial degradation [3].

Only a small number of bacterial species had been reportedly able to produce lignocellulose-degrading enzymes (e.g., cellulase, ligninase, and hemicellulases), which allow them to digest the lignocellulose and convert it into (fermentable) sugars [4]. Unfortunately, none of the BC-producing bacteria exhibited the ability to produce these enzymes [5]. In addition, the degradation of lignocellulose by lignocellulose-degrading bacteria requires a considerably-long time

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which may not be suitable for many industrial applications [6]. To overcome this problem, acid hydrolysis had been adopted as an effective method to break down the lignocellulose into simple sugars due to its fast and straightforward process [7]. The use of acid in the hydrolysis process allows an immediate breakdown of lignocellulosic biomass into sugars through hydrogen bonding disruption of lignocellulose. However, the byproduct of this conversion (carboxylic acids and furan aldehydes) can be toxic to the bacteria and may inhibit bacterial growth and fermentation [8]. Formic acid is one of the carboxylic acids in high amounts resulting from hydrolysis, whereas furfural and 5-hydroxymethylfurfural are the two commonly produced furan aldehydes [9]. Thus, a detoxification step is necessary to reduce the toxic compounds in the hydrolysate resulting from acid hydrolysis to promote bacterial growth and fermentation.

Several detoxification techniques, such as adsorption, ion-resin exchange, heat-treatment, irradiation treatment, peroxidase treatment, vacuum evaporation, and other biological treatments, can be utilized to reduce or eliminate the occurrence of toxic compounds [2,10–14]. However, most of the methods are not suitable for biomass detoxification because of the high degradation rates of essential compounds in biomass (e.g., fermentable sugars). For example, the heat, UV, microwave, and peroxidase treatment cause deformation of the physical state and degrade some of the essential compounds in biomass [15]. The use of peroxidase compound in the treatment also causes additional chemical waste generation, which can be harmful to the environment. Other techniques such as adsorption, ion-resin exchange, and biological treatment may be environmentally friendly, but they may involve a complicated and lengthy process [16].

A novel technique, namely atmospheric cold plasma (ACP), has been referred to as a breakthrough for biomass detoxification technique. ACP technique allows detoxification of biomass without any undesirable physicochemical properties change [15,17–19]. Furthermore, the ACP technique is also postulated to be potentially applied on an industrial scale due to its tunable and scalable operational parameters, including the voltage, type of feed gas, and treatment time [3]. ACP treatment has been popularly used to detoxify mycotoxins from food, where these compounds are un-removable using the conventional treatments [18,20–22]. ACP treatment works by inducing the generation of reactive species, which can inhibit microbial growth, thereby increasing the shelf-life of the food products. Owing to this work mechanism, ACP treatment can be applied to detoxify biomass from various toxic compounds [3]. Herein, ACP treatment was utilized to reduce the toxic compounds in hydrolysate from PPW prior to its use as the carbon source in BC fermentation. In this work, the effect of ACP treatment in the compositional change of PPWH was evaluated, especially the composition of the toxic compounds and fermentable sugars in PPWH. Two ACP reactors were used for the detoxification of PPWH, where the reactors operate at different specific plasma power range and gas sources. Thus, the performance of the two ACP reactors was demonstrated separately. The detoxificated-PPWH was then utilized as an inexpensive carbon source to supplement the fermentation media for BC production by *Komagateibacter xylinus* (*K. xylinus*).

2. Material and methods

2.1. Materials

PPW was collected from a local open market in Taipei, Taiwan, as a discarded biomass waste. Glucose and cellulose enzymes were purchased from Sigma-Aldrich, MO, USA. Fructose and xylose were purchased from Himedia, Mumbai, India. Peptone was obtained from Bioshop, Ontario, Canada. Yeast extract was obtained from Lab M, Heywood, UK. Citric acid, formic acid, HMF, and furfural were purchased from JT Baker Chemical Co., NJ, USA. Sodium bicarbonate was supplied by Showa Chemical Co., Tokyo, Japan. *K. xylinus* ATCC 23769 was obtained from Bioresource Collection and Research Center (Hsinchu, Taiwan).

2.2. Hydrolysis of PPW

The PPW was washed, crushed, and dried in a 60 °C oven. The dried pulp was ground and sieved to obtain uniformly sized particles of 30 mesh (± 0.6 mm). The lignocellulose content in PPW was determined using the Van Soest method [23]; the lignocellulosic content is 29.4% cellulose, 23.7% lignin, and 15.4% hemicellulose. Two-stage hydrolysis was employed to maximize the sugars content in the hydrolysate. In a typical hydrolysis procedure, the dried PPW was added to the 2.5%v of H_2SO_4 solution at a solid-to-liquid ratio of 1:10. The hydrolysis was conducted at 130 °C for 60 min. The hydrolysate was then separated from the solid residue by means of centrifugation. The solid residue was washed and dried in a 60 °C oven for 8 h, and then subjected for the 2nd-stage hydrolysis at the same conditions. The hydrolysate was pooled together and referred to as PPWH.

The composition of the sugars (i.e., glucose, xylose, and fructose) and toxic or inhibitory compounds (i.e., formic acid, HMF, and furfural) in PPWH was determined using high-pressure liquid chromatography (HPLC, JASCO, USA) equipped with Repro-Gel Ca column (Dr. Maisch GmbH, Germany). The chromatogram is provided in Supplementary data Fig. S1. 5 mM H_2SO_4 solution was used as the mobile phase at a flow rate of 0.6 mL/min. The hydrolysis rate was determined based on the amount of glucose extracted from PPWH per amount cellulose contained in dry PPW, according to Eq. (1):

$$\text{Hydrolysis rate } (\%) = \frac{\text{Amount of glucose in PPWH}}{\text{Amount of cellulose in dry PPW}} \times 100\% \quad (1)$$

2.3. Detoxification of PPWH using plasma treatment

Two types of ACP reactors (Industrial Technology Research Institute, Taiwan) were utilized to detoxify PPWH. Each reactor has a specific gas source and plasma power that can be achieved. Reactor 1 (R1) works at the power of 80–200 W with an Ar gas plasma source, while reactor 2 (R2) works at 500–600 W with air as the plasma source. The detailed specifications of the two reactors are provided in Supplementary data Table S1. The two reactors were used to detoxified PPWH, and the treated-PPWH was used as the carbon source to supplement the fermentation media. The effect of the plasma power of each reactor to detoxify PPWH was evaluated. The plasma power of 80, 120, 160, and 200 W was used in R1; meanwhile, plasma power of 500 and 600 W was used in R2. All plasma treatments were performed for 15 min, which is the best treatment time resulting in a minimal reduction in sugar and a moderately high reduction in inhibiting compounds (data not shown).

The detoxification procedures using each reactor were carried out by putting 50 mL of PPWH in an open stainless-steel Petri dish. Subsequently, the dish was placed into the discharge chamber at 1 cm distance from the jet head, which was designated to exert gas at a flow rate of 14 mL/min. The detoxification effect from both reactors was reported, where the effect was evaluated based on the alteration in toxic compounds and fermentable sugars of PPWH. The degradation rate of each compound is calculated according to Eq. (2):

$$\text{Degradation rate } (\%) = \frac{C_0 - C_f}{C_0} \times 100\% \quad (2)$$

where, C_0 and C_f are the concentration of the compound before and after treatment.

Table 1
Composition of glucose (Glu), fructose (Fru), xylose (Xyl), formic acid (FA), hydroxymethylfurfural (HMF), and furfural (Fur) in the fermentation media.

Media[a]	Concentration (g/L)					
	Glu	Xyl	Fru	FA	HMF	Fur
PPWH	16.49	8.86	14.56	0.26	0.26	0.18
R1-treated and its control group						
ARP-PPWH[b]	11.38	6.58	10.77	0.57	0.10	0.02
ARP-PPWH-c1	11.33	6.53	10.72	0.56	0.09	0.03
ARP-PPWH-c2	11.35	6.61	10.86	–	–	–
R2-treated and its control group						
AP-PPWH[c]	15.71	8.53	13.99	0.28	0.22	0.02
AP-PPWH-c1	15.59	8.84	13.88	0.26	0.21	0.03
AP-PPWH-c2	15.73	8.57	14.06	–	–	–

[a] Abbreviation for media naming: AP, air plasma-treated; ARP, Ar plasma-treated; c1, control media full composition; c2, control media without formic acid, HMF, and furfural.
[b] The media was prepared mixed with the R1-treated PPWH at 200 W for 15 min.
[c] The media was prepared mixed with the R2-treated PPWH at 600 W for 15 min.

2.4. Preparation of the culture medium

Seven culture media were prepared at different concentrations of sugars and inhibitory compounds, as given in Table 1. The PPWH was derived from the non-plasma-treated PPWH. ARP-PPWH was derived from the plasma-treated PPWH using R1. ARP-PPWH-c1 and ARP-PPWH-c2 were the control media prepared by modifying the composition of HS media to mimic the composition of ARP-PPWH. Meanwhile, AP-PPWH was derived from the plasma-treated PPWH using R2. AP-PPWH-c1 and AP-PPWH-c2 were the control media prepared by modifying the composition of HS media to mimic the composition of AP-PPWH. The pH of the culture media was adjusted to a pH value of 6.8 by the addition of 0.1 M NaOH. Then, the culture media was sterilized at 121 °C in an autoclave for 30 min. Besides the composition listed in Table 1, the media were supplemented with 1.15 g/L citric acid, 5 g/L peptones, 5 g/L yeast extract, and 2.7 g/L sodium hydrogen phosphate.

2.5. Culture of K. xylinus and BC production

The detailed preparation of the frozen stock culture and the propagation method of *K. xylinus* can be seen elsewhere [24,25]. The BC pellicles from the propagation step were aseptically collected and pureed using a Waring Blender 7011 HS. Then, 5 mL of the puree was inoculated in 50 mL of the modified media (Table 1) and incubated at 28 °C for 9 days. After fermentation, the BC pellicles were collected and then boiled at 90 °C in 100 mL of 0.1 N NaOH solution or 30 min. The obtained translucent BC pellicles were dried using a freeze dryer, the difference in weight before and after drying was measured to determine the water content.

2.6. Cell biomass measurement

The obtained BC pellicles were added into a fresh culture media, then 500 μL of cellulase from *Trichoderma reesei* (Sigma-Aldrich, USA) was added. The enzymatic degradation was allowed to proceed for 60 min at 50 °C. The obtained supernatant containing *K. xylinus* cells was centrifuged (Centrifuge-Universal 320R, Hettich Zentrifugen, Germany) at 5000 ×*g* at 4 °C for 10 min. Subsequently, the *K. xylinus* cells pellet was collected and washed with deionized water, then dried at 60 °C for 24 h. The dry cells were weighed and recorded as the dry cell weight.

2.7. Characterization of BC pellicles

The freeze-dried BC was characterized for their dynamic weight loss using a thermogravimetric analyzer (TGA) (TA Instrument Q500, DE, USA). The morphology was observed using a scanning electron microscope (SEM) (JEOL JSM-7800F, Tokyo, Japan); the samples were first coated with thin Pt-film using a coater JEOL JEC-300FC Quick coater (Tokyo, Japan). To determine the thickness of BC pellicles, image processing software ImageJ was used. The mechanical strength was determined using a texture analyzer (TXA) (TA-XT2 model, NY, USA). The

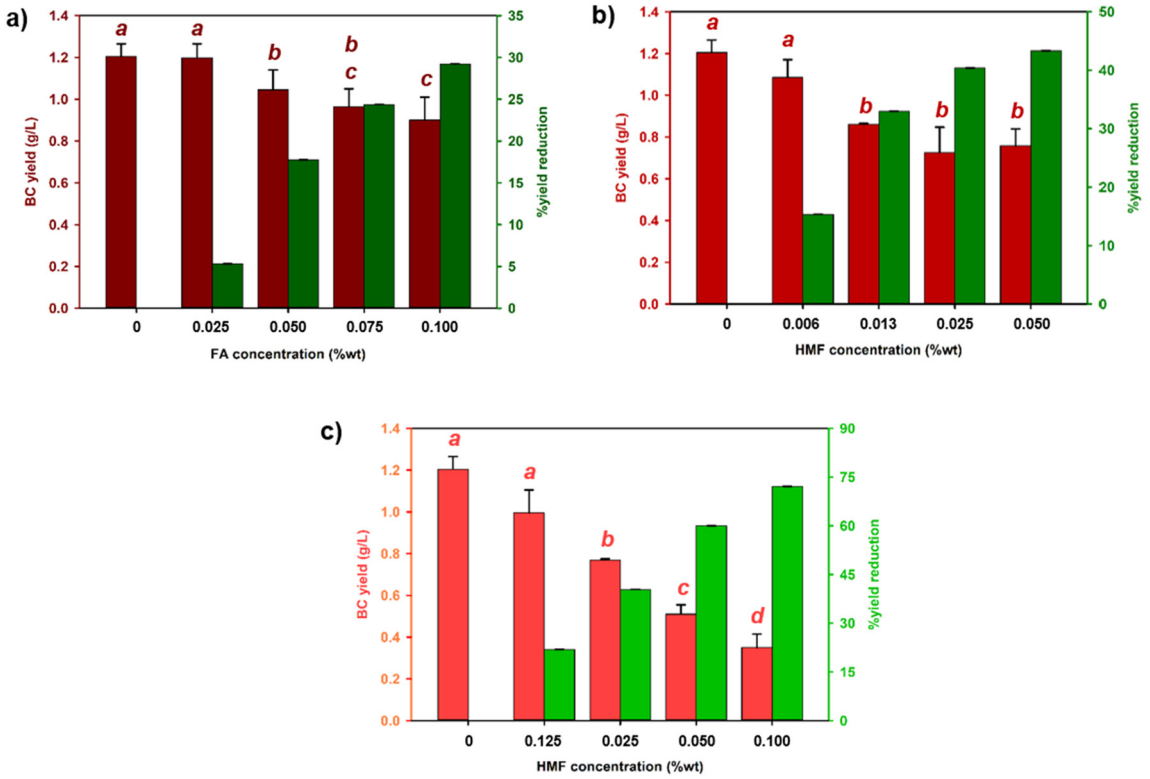

Fig. 1. Inhibitory effect of formic acid (FA), HMF, and furfural (Fur) on BC production by *K. xylinus*. The inhibitory compound was added at a specific concentration into the normal HS medium.

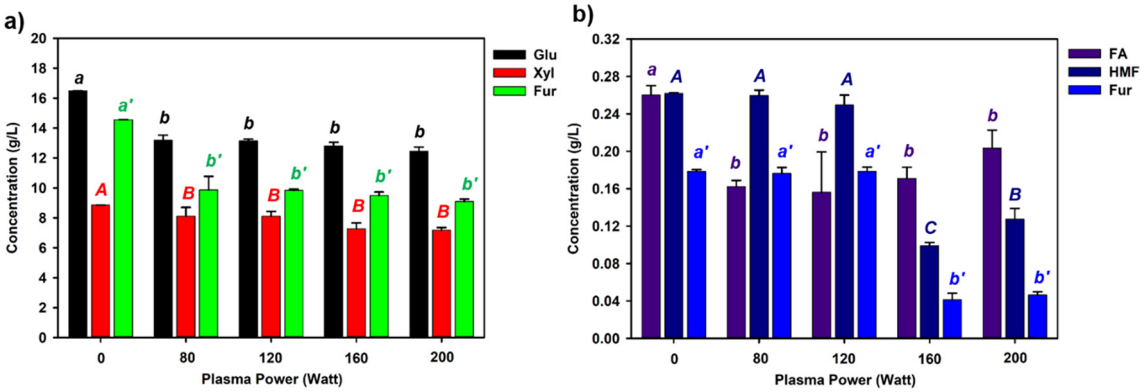

Fig. 2. The composition of sugars (a) and inhibitory compounds (b) in PPWH after 15 min ACP treatment using R1. Different letters on the error bars represent a significant difference ($P < 0.05$) relative to the control at 0 watt plasma power.

functional groups were determined using Fourier-transform infrared spectroscopy (FTIR) spectroscopy analysis using Shimadzu IRTracer-100 with KBr background. The crystallinity was determined based on X-ray diffraction analysis (XRD) using X'Pert PRO X-ray diffractometer, Philips, Netherlands, with Ni-filtered Cu Kα (1.540562 Å) radiation.

3. Results and discussion

3.1. Effect of the inhibitory compounds on BC production

The inhibitory effects of formic acid (FA), 5-hydroxymethylfurfural (HMF), and furfural (Fur) on the BC production by *K. xylinus* were studied using the one-factor-at-a-time (OFAT) method. Fig. 1a–c shows the demoting effect of these three inhibitors on BC yield and the percent reduction on BC yield. The quantitative data are provided in Supplementary data Tables S2 and S3. It can be noted that the presence of these compounds, even in small amounts, had a hugely detrimental impact on the formation of BC. The presence of 0.1%wt of Fur results in 72.1% BC yield reduction; meanwhile, the same concentration FA gave a 29.2% reduction. HMF shows more significant inhibitory activity to the BC formation, where the presence of this compound at concentrations 2.7 times smaller than Fur and FA resulted in a 43.3% reduction. This result is in well-convergence with a previous report describing the inhibitory effect of HMF on some enzyme functions, which caused a prolonged lag phase of the bacteria [3,26]. Disparate from HMF inhibitory effect, Fur is reported to affect bacterial cell viability due to its toxic effect [5].

3.2. Evaluation of the ACP treatment in the detoxification of PPWH

3.2.1. Effect of plasma power

ACP R1 works in the plasma power range from 80 to 200 W with an Ar gas source. The effect of different plasma power on the composition of inhibitory compounds and sugars in PPWH are presented in Fig. 2; the detailed quantitative values are presented in Supplementary data Table S4. As shown in Fig. 2b, the FA, Fur, and HMF are significantly decreased by treatment using a high plasma power. However, a significant decrease in the sugars content was also observed after high plasma power treatment (Fig. 3a).

Separate plasma treatments to the PPWH were conducted using ACP R2, which is equipped with air gas plasma and power of 500 W and 600 W. Interestingly, although ACP R2 has much higher plasma power, it has less impact on the sugars content in PPWH compared to ACP R1. As presented in Fig. 3b, no significant reduction of sugars occurs after plasma treatment using R2 at 500 and 600 W. Interestingly, as shown in Fig. 3a, the inhibitory compounds were significantly reduced after treatment using the highest plasma power of R2.

The degradation rate of each investigated compound in PPWH after treatment using R1 and R2 are presented in Table 2. It can be observed, both for R1 and R2, that a significant decrease in inhibitory compounds occurs at higher plasma power. This is since an increased plasma power allows faster plasma mobility and consequently promotes the interaction between the plasma and the inhibitory compounds, thus accelerating their degradation [3]. Furthermore, the inhibitory compounds are more susceptible to degradation since they have smaller molecular sizes than the sugars [17,18,27]. Based on the analysis of the

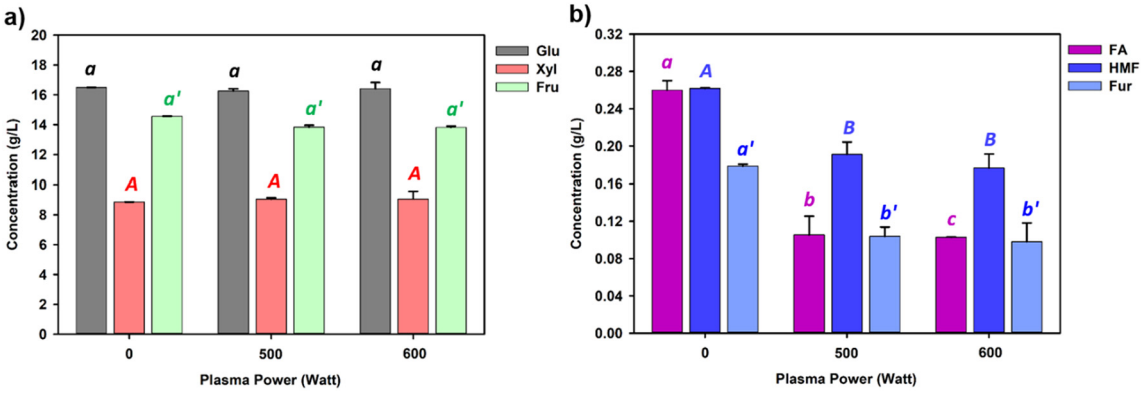

Fig. 3. The composition of sugars (a) and inhibitory compounds (b) in PPWH after 15 min ACP treatment using R2. Different letters on the error bars represent a significant difference ($P < 0.05$) relative to the control at 0 watt plasma power.

Table 2
Degradation rate of each compound in PPWH after 15 min plasma treatment using R1 and R2 at different plasma power.

Power (W)	Degradation rate (%)					
	Glu	Xyl	Fru	FA	HMF	Fur
Plasma treatment in R1						
80	20.03 ± 0.01	8.46 ± 0.12	32.19 ± 0.30	37.61 ± 0.01	0.87 ± 0.01	1.23 ± 0.01
120	20.29 ± 0.14	8.46 ± 0.22	32.48 ± 0.10	39.92 ± 0.03	4.66 ± 0.01	−0.06 ± 0.00[a]
160	22.24 ± 0.13	17.87 ± 0.09	34.75 ± 0.21	34.75 ± 0.01	62.10 ± 0.00	76.91 ± 0.11
200	24.51 ± 0.18	18.99 ± 0.18	37.55 ± 0.17	31.55 ± 0.02	51.38 ± 0.01	74.06 ± 0.20
Plasma treatment in R2						
500	1.38 ± 0.01	−2.18 ± 0.02[a]	5.06 ± 0.02	59.47 ± 0.12	26.97 ± 0.11	41.80 ± 0.11
600	0.48 ± 0.01	−2.28 ± 0.05[a]	5.12 ± 0.02	32.59 ± 0.05	32.58 ± 0.31	45.04 ± 0.22

[a] The negative value shows that the compound was formed instead of degraded.

degradation rate (Table 2), it can also be noted that ACP-treatment of PPWH using R1 shows a higher sugar loss than the treatment using R2 —Even though the plasma power in R1 is much lower than R2. One possible scenario for this result is the difference in the type of gas used as the plasma source. As mentioned in several studies, treatment using Ar plasma can induce a higher ROS formation than treatment using air-plasma [28,29]. The increased formation of ROS promotes the degradation of the compounds contained in PPWH. Furthermore, in the plasma treatment using R1, the high loss of sugars also followed by the significant increase of FA, which can be accounted for the degradation reaction of the sugars. Based on the degradation rate (Table 2) evaluation, the best detoxification effect was produced using plasma treatment at 200 W for R1 and 600 W for R2.

3.3. BC production from fermentation of *K. xylinus* in PPWH detoxified media

To investigate the performance of the untreated and plasma-treated PPWH, these hydrolysates were supplemented to the growth media of *K. xylinus*. The composition of the media can be seen in Table 1. Fig. 4a

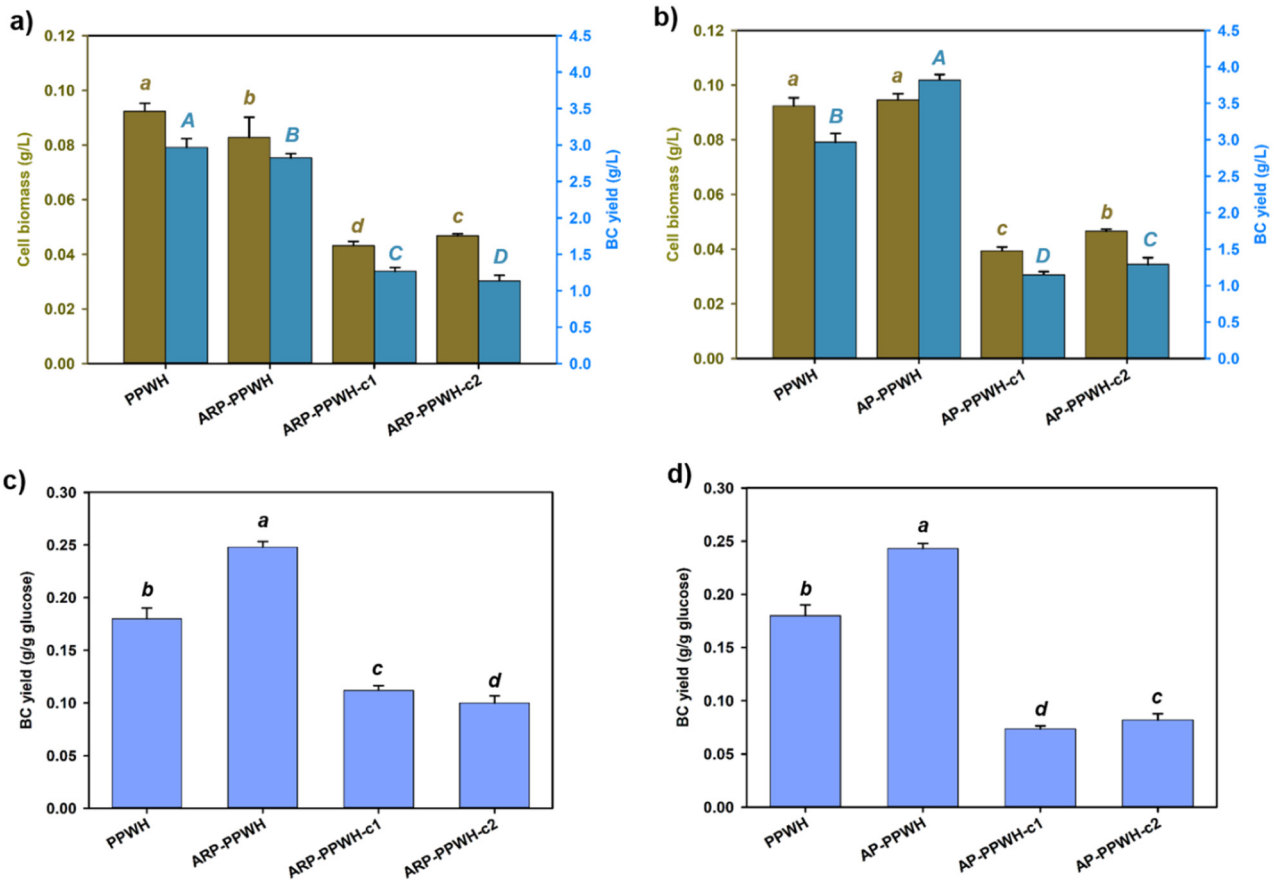

Fig. 4. a, b) BC yield (g/L) and cell biomass (g/L) after 9-days fermentation in R1-treated media and R2-treated media along with their control groups, respectively. c, d) BC yield in g/g of glucose after 9-days fermentation in R1-treated media and R2-treated media along with their control groups, respectively. Different letters on the error bars represent a significant difference ($P < 0.05$) relative to the BC yield and cell growth in PPWH media.

Table 3
Comparison of BC yield produced from the fermentation of several bacterial strain in different media.

Producing strain	Fermentation media	BC yield (g/L)	Ref.
Acetobacter xylinum[a]	Treated corn stalk	2.86	[35]
Gluconacetobacter xylinus[a]	Litchi extract	2.53	[14]
Acetobacter aceti	Optimized HS	1.73	[36]
Komagateibacter xylinus[a]	CPPE[b]	5.7	[11]
Komactobacter intermedius	Optimized HS	3.91	[25]
Komagateibacter xylinus[a]	AP-PPWH	3.82	This work
	ARP-PPWH	2.82	
	PPWH	2.97	

[a] *Acetobacter xylinum* and *Gluconacetobacter xylinus* are the former names of *K. xylinus*.
[b] CPPE: citrus peel and pomace enzymolysis.

and b shows the amount of BC yield and cell biomass growth after 9-days of fermentation of *K. xylinus* in PPWH and the plasma-treated PPWH containing media, along with the controls. A high BC yield can be obtained from the fermentation of *K. xylinus* in the plasma-treated PPWH compare to their control groups. The highest BC yield of 3.82 g/L is obtained from the fermentation of *K. xylinus* in the media containing R1-treated PPWH (ARP-PPWH). The second highest yield of 2.97 g/L is obtained from the fermentation in media containing untreated PPWH. Meanwhile, the fermentation in media containing R2-treated PPWH (AP-PPWH) resulted in a BC yield of 2.82 g/L.

The BC yield obtained from media containing AP-PPWH may seem lower than the BC yield obtained from PPWH in terms of g BC per L of media. However, when considering the BC yield in terms of g BC per g of Glu, the BC yield obtained from the AP-PPWH-containing media is much higher than the PPWH-containing media. As shown in Fig. 4d, BC yield of 0.24 g/g Glu is obtained after fermentation in AP-PPWH-containing media; meanwhile, 0.18 g BC/g Glu is obtained from PPWH-containing media. Interestingly, the fermentation in untreated PPWH also resulted in a significantly high BC yield; it implies that the PPWH alone can serve as the carbon source for BC fermentation. Investigation of cell biomass concentration of *K. xylinus* after 9-days fermentation (Fig. 4) indicates that the plasma-treated PPWH can significantly promote the growth of BC-producing bacteria.

It also can be noted from Fig. 4 that the formation of BC and the cell growth in PPWH supplemented media was higher than that of in ARP-PPWH-c2 and AP-PPWH-c2, where all of the media contained a specific concentration of the inhibitory compounds. This phenomenon can be attributed to the presence of other complex components in PPWH (i.e., plant phytochemicals) that may be promoted the growth of *K. xylinus* [30]. As postulated in other studies, organic compounds and some trace elements in fruit juices can facilitate the fermentation of BC [31–34].

The BC produced by *K. xylinus* in the Ar and air-plasma treated media has a comparable yield to BC produced from several other strains (Table 3). It is worth noting that the BC produced in fermentation using ACP plasma-treated media has a higher yield than several reported strains. Specifically, *A. xylinum*, which able to produce 2.86 g/L of BC using the acetic acid pre-hydrolysis liquor of corn stalk; *G. xylinus*, which able to produce 2.53 g/L of BC using litchi extract; and *A. aceti*, which only able to yield 1.73 g/L of BC after fermentation using optimized HS media [14,35,36]. The BC yield produced by *K. xylinus* (in this study) is still lower than several reported studies [11]. Even so, the utilization of PPW as the low-cost carbon source in BC fermentation can be a great solution in overcoming the high accumulation rate of this waste.

3.4. Characterization of the BC product

As shown in Fig. 5a, all BC samples share similar FTIR characteristic peaks. The peak corresponds to the O—H stretching vibration detected at the wavenumber 3358–3374 cm^{-1}. The C—H stretching of CH_2 and CH_3 peak is detected at wavenumber 2858–2936 cm^{-1}. Next, the peak corresponds to the H-O-H bending of the absorbed water is observed at a wavenumber range of 1644–1682 cm^{-1}. The peak displaying the CH_2 symmetric bending is observed at a wavenumber range of

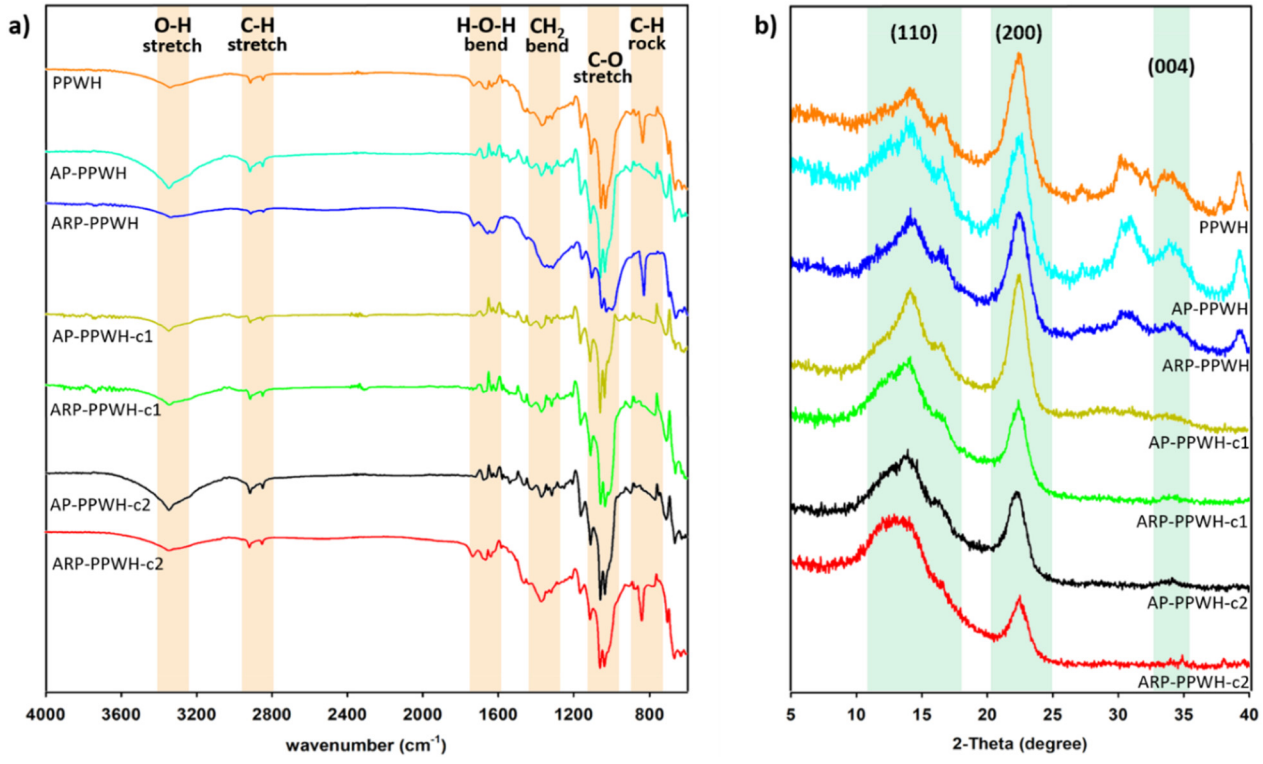

Fig. 5. Characterization on BC produced from the fermentation of *K. xylinus* in different media: (a) FTIR spectra and (b) XRD pattern.

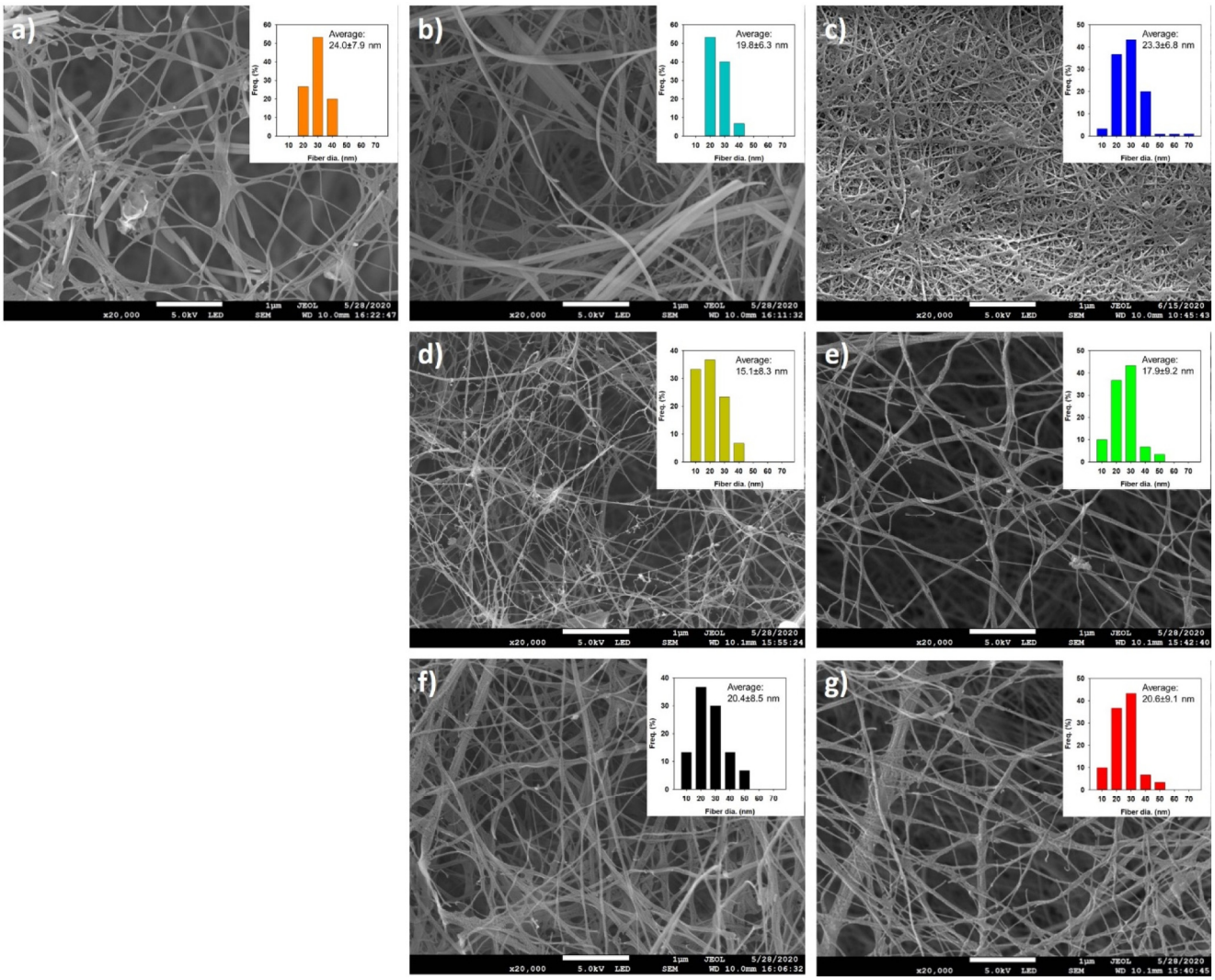

Fig. 6. The fiber morphology BC from *K. xylinus* fermented in different media. a) PPWH, b) AP-PPWH, c) ARP-PPWH, d) AP-PPWH-c1, e) ARP-PPWH-c1, f) AP-PPWH-c2, g) ARP-PPWH-c2.

1378–1474 cm^{-1}, commonly convoluted with the peak corresponding to O—H bending vibration. The C—O bending spectra are observed at a wavenumber range of 1040–1070 cm^{-1}. The FTIR spectra of BC samples are in good accordance with that of the reported literature [24].

XRD patterns and the corresponding crystal planes of the BC are presented in Fig. 5b. The typical pattern associated with the cellulose peaks is observed in all BC samples, which indicates the purity of the obtained BC [37]. Overall, the peak corresponds to cellulose Iα at the plane (110) is detected at a 2-Theta ranging from 13.1 to 14.2°, which is accompanied by the appearance of shoulder peak at near 16°. The respective peak associated with the cellulose Iβ, plane (200), is detected in the range of 22.1–22.4°. The plane (004) reflection appears obviously for BC fermented in PPWH, AP-PPWH, and ARP-PPWH at 33.4, 34.2, and 33.4°, respectively. This plane corresponds to the non-equatorial reflection of cellulose Iα [38].

Table 4
Measured properties of BC produced from the fermentation of *K. xylinus* in different media.

Fermentation media	Properties of BC				
	Water content (%)	Mechanical strength		Crystallinity (%)	Decomposition temp. (°C)
		Tensile stress (MPa)	Tensile strain (%)		
PPWH	98.91 ± 0.08^{a}	4.0 ± 1.07^{c}	0.48 ± 0.32^{c}	72.93	258.1
ARP-PPWH	98.68 ± 0.06^{a}	5.21 ± 1.48^{c}	1.05 ± 0.24^{bc}	73.46	311.3
ARP-PPWH-c1	97.82 ± 0.24^{b}	43.83 ± 14.59^{a}	0.59 ± 0.16^{bc}	83.53	342.7
ARP-PPWH-c2	98.86 ± 0.16^{a}	46.14 ± 13.15^{a}	0.95 ± 0.73^{bc}	84.40	316.5
AP-PPWH	98.70 ± 0.03^{a}	6.04 ± 0.94^{c}	0.52 ± 0.09^{bc}	70.54	264.5
AP-PPWH-c1	98.76 ± 0.13^{a}	47.25 ± 9.11^{a}	1.02 ± 0.18^{b}	85.18	335.5
AP-PPWH-c2	98.73 ± 0.01^{a}	33.55 ± 1.54^{b}	2.47 ± 0.92^{a}	86.11	338.7

Different superscript letters indicate a significant difference ($P < 0.05$).

The morphology of the BC produced in PPWH and plasma-treated PPWH containing media (Fig. 6a–c) are only slightly different from the control (Fig. 6d–g). All of the samples show similar fibrous morphology, suggesting that the BC structure mostly depends on culture condition rather than feedstock sources [1,12]. The most-dense BC fibers distribution is observed from the fermentation using the ARP-PPWH medium, followed by AP-PPWH and PPWH. The resultant BC fibers have a diameter ranging from 10 to 70 nm (inset Fig. 6), with an average width of 20 to 24 nm.

Other properties of the BC were also studied (Table 4). The crystallinity of the BC samples was determined based on the amorphous and crystalline area ratio, as detected in the XRD pattern. The BC obtained from the fermentation of *K. xylinus* using PPWH containing media has a lower crystallinity than the control groups. The decreased crystallinity of BC produced from PPWH containing media can be due to polysaccharides content in PPW, which induces aggregation of cellulose chains into microfibrils [10,39]. All BC samples possess equally high water content, indicating their excellent water retention ability. The thermostability of BC was assessed based on decomposition temperature, as measured using TGA. It was noted that the BC obtained via fermentation using PPWH containing media have lower thermal stability. This is due to the larger amorphous moiety of BC, which is more susceptible to thermal breakdown [10].

The mechanical strength of BC is expressed as tensile strength and tensile strain; both values depict the capability of the BC fiber to resist and elongate without cracking after given external forces. The BC produced from fermentation in PPWH containing media is susceptible to deformation than BC produced from their control groups. This can be attributed to the higher amorphous moiety of the BC from PPWH containing media, resulting in lower mechanical strength.

4. Conclusion

Detoxification of PPWH using atmospheric cold plasma (ACP) treatment with argon and air gas source has been demonstrated. Both treatments result in an adequate reduction of the toxic compounds (i.e., formic acid, HMF, and furfural) in PPWH. The argon plasma, which works at the plasma power of 80–200 W, has a more significant degradation effect on the sugars content in PPWH than air plasma, which operates at a much higher plasma power of 500–600 W. The use of ACP-treated PPWH, as well as the untreated PPWH, as the carbon source for BC production can result in a high yield of BC. The BC fermented in the ACP-treated PPWH and untreated PPWH do not show meaningful characteristics change, which indicates that the use of those bioresource does not cause a decrease in the quality of the BC. This also suggests that both treated and untreated PPWH can serve as a low-cost and sustainable carbon source to supplement the fermentation media.

CRediT authorship contribution statement

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Shin-Ping Lin: Data curation, Writing – Reviewing and editing
Tan-Ying Wang: Formal analysis, Writing – Original draft
Yuwen Ting: Project administration, Supervision
Chang-Wei Hsieh: Data analysis; Validation
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Artik Elisa Angkawijaya: Writing – Original draft, Writing – Reviewing and editing
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Kuan-Chen Cheng: Supervision, Conceptualization, Resources, Project administration, Funding acquisition.

Declaration of competing interest

The authors declare no conflict of interest.

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Appendix A. Supplementary data

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