

Biomass waste dissolution with imidazole ionic liquids for biohydrogen production

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Abstract

Cellulosic biomass waste from municipal solid and agricultural biomass residue are Second Generation energy source, mainly contain glucose and xylose monomers, were extensively studied in present research for fermentable sugar hydrolysate in biohydrogen production. Scrap paper cup (SPC), bamboo stem wall (BSW), sugar cane bagasse (SCB) and oil palm empty fruit bunch (OPEFB) were dissolved in laboratory prepared imidazole ionic liquids; 1-allyl-3-methylimidazolium chloride ([Amim]Cl) and 1-butyl-3-methylimidazolium chloride ([Bmim]Cl). A comparative study on biomass composition was presented by Van Soest and thermalgravimetric analysis (TGA) method. TGA was proved as comparative, cheaper and faster method in measuring the lignocellulose composition. Experimental result show that the ionic liquids were completely dissolved the SPC, BSW, SCB and OPEFB with high cellulose recovery; 96.00%, 91.34% 87.16% and 99.51% respectively. The used ionic liquids were highly recovered from the mixture at 94% to 99% recovery rate and FTIR analysis proofed that the recycled ionic liquid is principally consistent with the original. The regenerated cellulose was undergo acid hydrolysis to reducing sugars (glucose/xylose) hydrolysate to be used as feedstock fermentation for biohydrogen production. Acid hydrolysis of the recovered cellulose resulted up to 96% sugar conversion. IL-SPC hydrolysate reported higher total sugar conversion compare to SPC (control) due to higher surface area and disintegration of the cellulosic fibril structure resulted from the dissolution process. IL-SPC, IL-BSW, IL-SCB and IL-OPEFB hydrolysate contained higher total sugar compared to SPC hydrolysate even though their cellulose recovery are lower than the SPC (control). Biohydrogen fermentability test of this hydrolysate was carried out using biohydrogen producing bacterium *Clostridia* sp. Almost 85% of biomass waste hydrolysate substrate was utilized by the bacteria. Up to 196 ml H₂ / 100 ml cumulative biohydrogen production was collected for fermentation using the biomass hydrolysate while 174.91 ml H₂ / 100 ml was produced from the control.

Introduction

Circular economy is an international trend, and how to utilize waste to high-value products is a big-profile issue. Biohydrogen is one of attractive future energy carrier compared to other biofuel due to its high energy density, low energy input and higher conversion efficiency to usable power including its non-polluting nature (Dutra et al., 2017). Scrap paper cup (SPC) was widely used all over the world and become a threat to the environment. It composed of 90% high strength paper with 5% thin coating of polyethylene to make the paper containers waterproof but make it very complicated to recycle (Arumugam et al., 2017). Agriculture waste which is plentiful and readily available, is categorized as lignocellulosic biomass. It has complex molecular structure with tangled chain of cellulose, hemicellulose and lignin (Kim, 2018). Hydrolysis of the biomass and SPC can be used as source of fermentable sugar for biohydrogen production (Harun et al., 2013, Kim, 2018). However, the complex lignocellulose biomass structure with intra and intermolecular hydrogen bonds make the hydrolysis of the biomass and SPC become a major challenge (Xing et al., 2014). Therefore, this paper exploring an environment friendly preprocessing step of the SPC and agricultural biomass, to provide efficient process of cellulose conversion to fermentable sugar hydrolysate for biohydrogen production.

The difference composition of chemical components in biomass waste is directly influence their chemical reactivity. This is why determination of the total amount of each lignocellulose components is crucial to foresee the efficiency of a biomass conversion process (Carrier et al., 2011). There are several established composition analysis standard methods (Ioelovich, 2015) that applied at lignocellulose based research area such as National Renewable Energy Laboratory (Sluiter et al., 2008), Technical Association of the Pulp and Paper Industry, Scandinavian Pulp, Paper and Board (Scandinavian, 2009), Van Soest method (Van Soest et al., 1991) and Thermogravimetry analysis (TGA) that primarily introduced by American Society for Testing and Materials (Earnest, 1988). Since different methods may give different results, this study presented comparison of lignocellulose biomass composition determined by using Van

Soest and TGA method. Van Soest also known as detergent fiber analysis where the lignocellulose fiber fractionated into neutral detergent fiber (NDF), acid detergent fiber (ADF) and acid detergent lignin (ADL). In this method, NDF is the raw materials removed protein, fat and other extracts, which mainly includes cellulose, hemicellulose, lignin and ash. ADF mainly containing cellulose, lignin and ash while ADL mainly consists of lignin and ash. By difference, hemicellulose (NDF-ADF), cellulose (ADF-ADL) and lignin (ADL-Ash) are calculated (Hindrichsen et al., 2006). Compare to wet chemical method by Van Seost, TGA is an analytical technique, monitoring fraction material weight degradation that occurs as sample is heated at constant rate. TGA can quantitatively measure the components of complex mixtures because of ability to determine characteristic thermal decomposition each one of them. The composition of cellulose, hemicellulose, lignin and others from the TGA were calculated as the literature on pyrolysis of lignocellulose components as reported by Yang et al. The research report on pyrolysis of individual and mixtures components of cellulose, hemicellulose and lignin. Simplex-lattice approach was used for analysis of both approach and linear relationship occurred between the weight losses of each lignocellulose components during pyrolysis. From the study, first weight loss of biomass occurred at room temperature to 100 °C, indicate water content, hemicellulose started the weight loss at temperature 220 to 315 °C, follow by cellulose that degraded at temperature from 315 to 400 °C, while lignin have widest temperature range of degradation which at 100 °C to 900 °C (Yang et al., 2006). Numerous researches have been done in utilizing TGA as alternative method for biomass composition analysis as it can provide faster, easier and less expensive (Binti et al., 2014; Carrier et al., 2011; Earnest, 1988; Wang et al., 2019; Yang et al., 2007). However, comparative study of the TGA with Van Soest method in biomass composition analysis is new presenting in this paper.

Ionic liquids have received extensive attention as a promising solvent in dissolving cellulose under unpressurised condition. Ionic liquids are organic salt composed of anions and cations. They usually melt below 100 °C and also recognized as “green” solvent. Application of ionic liquids have several advantages such as low vapor pressure, high polarity, good dissolving and extracting ability, wide liquid range, good thermal stability and excellent design ability (Xia et al., 2018). Ionic liquids provide excellent characteristics in chemical processes which able to dissolve polar and non-polar organic, inorganic and polymeric compounds. The presence of anion in the ionic liquid can extensively disrupt the hydrogen bonding interactions in the three dimensional network of lignocellulose, leading to dissolution of biomass components. Besides that, the ionic liquids can be recovered after the dissolution process (Singh et al., 2018). Formerly, several literatures were reported utilization of variety ionic liquids for the dissolution of full lignocellulose biomass followed by cellulose hydrolysis with acid or enzymes (Jeong et al., 2018; Lammens et al., 2017; Moyer et al., 2018; Navarra et al., 2015; Tajuddin et al., 2019; Xing et al., 2014). However, dissolution of scrap paper cup by ionic liquids for preparation of fermentable sugar hydrolysate is a novel, presented in this paper.

There were many types of ionic liquid that was studied especially in cellulose dissolution. Imidazole cations ionic liquid has reported could dissolve lignocellulose biomass (Singh et al., 2018; Xia et al., 2018). Therefore, in this study two imidazole based ionic liquids were prepared in laboratory; 1-allyl-3-methylimidazolium chloride ([Amim]Cl) and 1-butyl-3-methylimidazolium chloride ([Bmim]Cl). The difference between the two ionic liquids are [Amim]Cl contained C = C structure on allyl chain of the imidazole ring while [Bmim]Cl consist of C-C butyl chain (Xing et al., 2014). The ionic liquids are analyzed by FTIR to confirm the target ionic liquid has synthesized. The ability and efficiency of prepared [Amim]Cl and [Bmim]Cl are determined by monitoring the regenerated cellulose/hemicellulose recovery and ionic liquid recycled recovery.

Most organisms cannot directly consume cellulose/hemicellulose because of its complex structure and high molecular weight. In order to convert cellulose/hemicellulose into a carbon source that can be utilized by organisms, hydrolysis is an indispensable step in bioenergy. Acid hydrolysis is one of promising method for degradation of cellulose/hemicellulose into monosaccharides or destroy the fiber structure by cut the glycosidic bond composed of

cellulose/hemicellulose (Xiang et al., 2003). The biohydrogen production was carried out by dark fermentation, and used the mixed consortium of *Clostridia* sp. The experiments show that green processes is a feasible way to make the biomass waste to energy.

Experimental Method

The experiments were design by several steps to work out a green process of recovering cellulose/hemicellulosse from municipal solid waste SPC; and plant biomass: BSW, SCB, and OPEFB, to be used as sugar feedstock in biohydrogen production as shown at Fig. 1.

- I. A control step was established to distinguish the effect of ionic liquids on the acid hydrolysis and biohydrogen production by directly hydrolyzed the biomass waste without ionic liquid treatment. SPC was hydrolysed with 10M sulfuric acid at 65 °C for 50 min, then the obtained hydrolysate was used as the feedstock for hydrogen production.
- II. The other route, involved the utilization of ionic liquids [Amim]Cl and [Bmim]Cl to dissolved the biomass waste before acid hydrolysis. The biomass waste that pretreated with ionic liquid named as IL-SPC, IL-BSW, IL-SCB and IL- Then, the hydrolysate of the IL-SPC, IL-BSW, IL-SCB and IL-OPEFB were used as sugar feedstock in dark fermentation for biohydrogen production.

Biomass composition analysis by Van Soest and TGA method

Biomass composition was analyzed by analytical method develop by Van Soest et al. (1991) and compared with TGA method. Two different biomass were used in the study; BSW and SCB. In this study, the biomass were analyzed by Van Soest standard method as reported at Van Soest et al., (1991). Analysis of the cellulose, hemicellulose, and lignin contents of the biomass were determined synchronously. It involved preparation of natural detergent fiber (NDF), acid detergent fiber (ADF), acid detergent lignin (ADL) and ash content determination. NDF is the raw materials removed protein, fat and other extracts, which mainly contained cellulose, hemicellulose, lignin and ash. ADF is the acid detergent fiber that mainly containing cellulose, lignin and ash. Thus, hemicellulose content can be determined by minus the NDF and ADF while the cellulose composition calculate as ADF – ADL. As ADL is mainly consists of lignin and ash, lignin composition can be found by minus the ash amount from the ADL. Table 1 shown the simplification of the hemicellulose, cellulose and lignin component by Van Soest method.

Table 1
Determination of lignocellulose component composition by Van Soest method

Lignocellulose components	Component determination
Moisture content	Moisture content analysis of raw biomass
Hemicellulose	NDF - ADF
Cellulose	ADF - ADL
Lignin	ADL - Ash
*Method adapted from (Van Soest et al., 1991)	

TGA analysis of the biomass was carried out using TA Instrument TGA2950 with the presence of nitrogen gas (N₂) at the flowing rate of 150 ml/min. Biomass waste samples at size 3 mm x 3 mm x 1 mm between 5 to 10 mg were pyrolysis to a maximum temperature of 600 °C. The sample was first heated to 110 °C and kept at that temperature for

30 minutes to remove any moisture. After that, the samples were individually heated at 50 °C/min until 600 °C. The composition of cellulose, hemicellulose, lignin and others from the TGA were calculated as the literature on pyrolysis lignocellulose components; the first weight loss occurred at room temperature (RT) to 100 °C, indicate the water content, hemicellulose started the weight loss at temperature 220–315 °C, follow by cellulose that degraded at temperature from 315 to 400 °C, while lignin have widest temperature range of degradation which at 100 °C to 900 °C (Yang et al., 2006)

Synthesis of ionic liquids

The preparation of ionic liquids are a modified method of Xing et al., (2014). For the synthesis of [Amim]Cl, 1-methylimidazole and allyl chloride were added to a three-necked flask, the temperature was controlled at 55 °C for 8 hours, and then the excess allyl chloride and impurities were distilled off to give pure [Amim]Cl. [Bmim]Cl was synthesized with combination of 1-methylimidazole and 1-chlorobutane in three-necked flask and kept the temperature at 85 °C for 24 h. After that, it was cooled to room temperature before wash with ethyl acetate and concentrated. Then, the pure final product was obtained by lyophilization. Schematic diagram of preparation of the ionic liquid was presented at Fig. 2.

Dissolution of biomass with ionic liquids

The ionic liquids were heated to 120 °C while stirring, and 10 wt% of biomass waste SPC, BSW, SCB and OPEFB were added gradually into the ionic liquids individually. The SPC and BSW were dissolved in [Bmim]Cl while SCB and OPEFB were dissolved in [Amim]Cl. The mixtures were agitated and the mixture's temperature maintained at 120 °C until all biomass waste were completely dissolved. The mixtures are turned to dark brown color and touch light was used to check any presence of undissolved particles in the solution.

Cellulose recovery and ionic liquids recycle

After completed the dissolution process, water was added to the mixture to precipitate the cellulose/hemicellulose. Two apparent layer was formed and the upper layer was separated from the mixture for further lignin and ionic liquid recovery. The precipitated cellulose/hemicellulose was further washing with deionized water on pre-weighed stainless steel filter paper by suction filtration. While washing, the precipitate cellulose/hemicellulose turned to clear solution. The obtained cellulose/hemicellulose was further washed with hot water to completely remove the ionic liquid. The cellulose/hemicellulose recovery was calculated as Equation 1. The filtered cellulose/hemicellulose was then undergo acid hydrolysis. Water and ionic liquids were separated by vacuum distillation. At 160 mmHg pressure and 80°C, water was distilled while remained the ionic liquid in collector flask. The moisture content of the ionic liquid was measured and FTIR analysis was done to check on any changes of ionic liquid compositions. The recycled ionic liquid recovery was determined as Equation 2.

$$\text{cellulose/hemicellulose recovery (\%)} = \frac{\text{regenerated cellulose/hemicellulose (g)}}{\text{cellulose/hemicellulose in raw biomass (g)}} \times 100 \quad (1)$$

$$\text{ionic liquid recovery (\%)} = \frac{\text{recovered ionic liquid (g)}}{\text{initial ionic liquid (g)}} \times 100 \quad (2)$$

Acid hydrolysis

10 M sulphuric acid has been choose for the acid hydrolysis process, based on preliminary study of selection inorganic acid for hydrolysis of biomass waste. In the study, four different acids which are sulfuric acid, hydrochloric acid, nitric acid and phosphoric acid at concentration 3, 5, 8 and 10 M were tested to hydrolyze SPC at 65 °C for 55 min. The

experimental result show that, 10 M sulphuric acid gave the highest conversion of reducing sugar (98.72% conversion). Therefore, sulfuric acid was selected for further hydrolysis of the SPC (without ionic liquid dissolution), ILs-SPC, ILs-BSW, ILs-SCB and ILs-OPEFB at maintained temperature of 65 °C for 55 min.

Biohydrogen production

Batch fermentations were carried out in a 200 ml serum bottle. Hydrolyzed sugar of recovered cellulose from SPC, IL-SPC, IL-BSW, IL-SCB and IL-OPEFB were used individually as carbon source for fermentation. The initial total concentration of carbohydrates in the enzymatic hydrolysate was adjusted to 20 g/L. Urea was used as the nitrogen source at concentration 4.4 g/L. Additional nutrients for bacterial growth consisted of NaHCO_3 (6.72 g /L), CaCl_2 (0.1 g /L), K_2HPO_4 (0.125 g/L), $\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$ (0.0001 g /L), $\text{CuSO}_4 \cdot 7\text{H}_2\text{O}$ (0.005 g /L), $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ (0.025 g/L), $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$ (0.1 g/L), $\text{MnSO}_4 \cdot 4\text{H}_2\text{O}$ (0.015 g/L). All samples were adjusted to an initial pH of 7, and 90 mL of media was dispensed into the bottle. Argon gas was sparged through the media for 30 seconds to create anaerobic conditions for the fermentation. 10 mL of the activated hydrogen producing bacterium *Clostridia* sp. was added into the fermentation media and make the total working volume to 100 ml. Bottles were capped with a soft silicon caps and clamped with an aluminium cap. The bottles were incubated for 12 hours at 37 °C and 150 rpm in an incubated shaker throughout the experiments. The biogas produced was sampled using disposable syringes for further compositional analysis at designated sampling time up to 200 hours (Li et al., 2011; Liu et al., 2013; Wu et al., 2003).

Analysis

Composition of the SPC and agricultural biomass were analyzed by thermogravimetric analysis (TGA) brand TA Instrument TGA2950. The physical morphology of biomass waste, before and after dissolution by ionic liquid were figured out by scanning electron microscope (SEM) Hitachi S-3400H analysis. The synthesized ionic liquid [Amim]Cl and [Bmim]Cl were analysed by fourier transform infrared spectroscopy (FTIR) (JASCO FTIR 460 Plus) to confirm the target ionic liquids were produced. After the acid hydrolysis, the composition of hydrolysate was measured by high performance liquid chromatography (HPLC) Brand Hitachi with ICESep Coregel 87H3 column to determine the amount of primary monosaccharides (glucose, xylose, arabinose, sucrose), the metabolites (butyric acid and acetic acid) and other fermentation inhibitor (formic acid, propionic acid, and furfural). Biogas constituents were analysed using gas chromatography (GC) with thermal conductivity detector (SHIMADZU GC-14B). Phenol-sulfuric acid method by UV/VIS spectrophotometer (HITACHI U-5100).

Results And Discussion

Characterization of ionic liquid

The laboratory synthesized ionic liquids were analyzed with FTIR to characterize the target products. The important FTIR spectrum peaks for [Bmim]Cl and [Amim]Cl were: 3150 cm^{-1} (C-H of imidazole ring), 3110 cm^{-1} (= C-H of imidazole ring), 1430 cm^{-1} (C-H of = CH₂ structure), 1570 cm^{-1} (C-N of imidazole ring) and 621 cm^{-1} (C-Cl). Additional of peaks 1650 cm^{-1} (C = C, allyl) and 951 cm^{-1} (R-CH = CH-H) for [Amim]Cl (Xing et al., 2014).

[Bmim]Cl spectrum at Fig. 3(a) presented wave number 3110 cm^{-1} , 1570 cm^{-1} and 1461 cm^{-1} were the imidazole ring skeletons vibration absorption peaks. The characteristic bands at 1168 cm^{-1} was assigned to the methyl hydrogen's deformation absorption while the skeleton vibration of C-C was at 1569 cm^{-1} . Peak at 1168 cm^{-1} and 620 cm^{-1} were the inner and outer bending vibration of C-H and C-Cl respectively. The peaks of wave number 3460 cm^{-1} and 3360 cm^{-1}

1 which were attribute to the absorption of n-butyl hydrogen. Determination of the peaks were indicated that the [Bmim]Cl was synthesized accordingly.

The functional groups and their wavelengths of [Amim]Cl were showed in Fig. 3(b). Imidazole ring skeletons vibration absorption peaks indicated at peak 3150 cm^{-1} (alkyl group), 1430 cm^{-1} and 1570 cm^{-1} (C = C stretching vibration) and 1165 cm^{-1} (HCC). 1639 cm^{-1} was the peak for¹ C = C, allyl group while peak at 951 cm^{-1} was the R-CH = CH-H. According to the analysis results, the specific functional groups are consistent with the results analyzed by and the results proved that the target product was successfully synthesized.

Comparison of biomass composition by Van Soest and TGA method

The biomass composition analysis by Van Soest and TGA were compared and showed at Table 2. The result was described as composition of moisture, cellulose/hemicellulose and lignin, ash and other. Cellulose and hemicellulose contents were presented together as sugar sources is come from both of them. The experiment result showed that, among the other biomass components, cellulose/hemicellulose content was the largest composition for both BSW and SCB. Cellulose/hemicellulose content of SCB, showed 2% significant difference which were 74.03% by Van Soest and 71.99% by TGA method. Lower cellulose/hemicellulose content from Van Soest of SCB may because that certain percentage of soluble hemicellulose might have been dissolved in neutral detergent solution (Hindrichsen et al., 2006). Nevertheless, BSW shown very near cellulose/hemicellulose content by both Van Soest and TGA method. BSW contained 66.12% and 65.13% of cellulose/hemicellulose as identified by Van Soest and TGA method respectively. The finding showed that TGA was a comparative method with Van Soest on quantifying cellulose/hemicellulose content of lignocellulose biomass even though there were a slight different. Moisture content determination by both methods were consistent as the results were very close for Van Soest and TGA method. Biomass composition analysis by TGA method seems to be quite reliable because the value where close to the data from Van Soest method.

Table 2
Biomass waste compositions by Van Soest and TGA measurement

Composition (%)	Bamboo stem wall		Sugar cane bagasse	
	Van Soest	TGA	Van Soest	TGA
Moisture content	5.69 ± 0.39	5.66 ± 0.02	7.16 ± 0.10	6.49 ± 0.43
Cellulose & hemicellulose	66.12 ± 1.45	65.13 ± 1.86	74.03 ± 1.77	77.63 ± 1.07
Lignin, ash & others	28.20 ± 0.62	26.33 ± 0.71	18.81 ± 0.62	21.53 ± 0.43
Total	100	100	100	100

Biomass waste composition by TGA method

All the biomass waste’s chemical compositions were analyzed with TGA method. According to the literature on pyrolysis lignocellulose components, hemicellulose started the weight loss at temperature 220–315 °C, follow by cellulose that degraded at temperature from 315 to 400 °C, while lignin have widest temperature range of degradation which at 100 °C to 900 °C. Figure 4(a) shows the pyrolysis curve of scrap paper cups (SPC) in TGA analysis. The first weight loss of SPC occurred at room temperature (RT) to 100 °C, it accounts for 7.52% of total weight, which indicate the water content in the SPC. From the pyrolysis curve, it was determined the hemicellulose and cellulose composition of SPC which are 58.50% and 17.17% respectively. There might be some weight of lignin together with the composition of cellulose and hemicellulose because of the wide degradation of lignin. Nonetheless, as the degradation of lignin is very low mass loss rate (< 0.14 wt%/°C) the amount can be negligible. The SPC pyrolysis curve has comparable study

of Dikobe and Luyt (2010) which reported on the properties of PP/LLDPE/wood composites polymer. There were identified on the SPC pyrolysis curve, 11.46 wt% weight loss at 400–500 °C and this signal is visibly known as linear low-density polyethylene (LLDPE) material. Above 500 °C, some residual was decomposed and identified as ash. The composition of scrap paper cup, rice husk, sugar cane bagasse and oil palm empty fruit bunch were shown in Table 3. From the result analyzed by TGA method, bamboo stem wall contains 27.35% cellulose, 43.14% hemicellulose, 19.39% lignin and the rest of 6.94% was the ash and others. The cellulose, hemicellulose and lignin contents were congruent with chemical composition of bamboo studied by (Sharma et al., 2018) and (Sulaiman et al., 2016) which the analysis was performed in TAPPI and NREL method respectively. The studies found that the SCB chemically composed of 27.35%, cellulose, 28.71% hemicellulose, 26.29% lignin and others. The cellulose content was low if compare to study did by Rocha et al., (2012), while the hemicellulose and lignin content is quite similar with the research finding even though the analysis was performed by NREL method. Oil palm empty fruit bunch contains cellulose 16.11%, hemicellulose 45.36%, lignin 22.61% and 7.55% of ash or others. The cellulose content was also low compared to OPEB chemical composition found by (Nur Farahin et al., 2018)(Abdul et al., 2016) (Tajuddin et al., 2019) which the cellulose content is around 31–42% of total composition. The hemicellulose composition in this study was determined high compare to other findings. Nevertheless, proximate analysis of OPEFB pyrolysis by TGA performed by Alias et al. (2014) shown similar result with this findings. The difference may due to the influence of lignin, because the pyrolysis range of lignin covers the entire pyrolysis process.

Biomass waste dissolution and recovery

The experiment result showed that the laboratory prepared ionic liquids [Amim]Cl and [Bmim]Cl were able to dissolved completely the SPC, BSW, SCB and OPEFB biomass waste at temperature 120 °C. The complete dissolution of the biomass waste was confirmed though scanning electron micrograph (SEM) images as shown at Fig. 5. Dissolution mechanism of cellulose in ionic liquids is based on capability of ionic liquid’s anions to effectively break the extensive intra and inter molecular hydrogen bonding network in cellulose. [Amim]Cl ionic liquid is composed of [Amim]⁺ cation and Cl[−] anion. While, [Bmim]Cl has [Bmim]⁺ cation and also Cl[−] anion. Cl[−] anion of the ionic liquid acts as the hydrogen bond acceptor in dissolution where it interacts specifically with the hydroxyl protons of the cellulosic materials and facilitates the formation of hydrogen bonds between cellulose and ionic liquid (Gogoi & Hazarika, 2017; Gupta & Jiang, 2015; Hou et al., 2017; Liu et al., 2019). Ionic liquid with strong hydrogen bonds are effective in weakening the hydrogen bonding network of the polymer chain. The cation of ionic liquid also can indirectly influence the dissolving ability by impacting their physical properties, such as melting point, density and viscosity. The researcher also found that weak hydrogen bond present between the oxygen atom of cellulose and the acidic hydrogen ring of cation. Small van der Waals interaction formation between of glucose and cations.

Table 3
Biomass waste compositions by TGA measurement

Composition (%)	Scrap paper cup	Bamboo stem wall	Sugar cane bagasse	Oil palm empty fruit bunch
Moisture content	7.52	5.66 ± 0.02	6.49 ± 0.43	8.37 ± 0.23
Cellulose & hemicellulose	75.67	65.13 ± 1.86	71.99 ± 1.07	61.47 ± 2.50
LLDPE	11.46	-	-	-
Lignin, Ash and others	4.47	29.21 ± 1.43	21.53 ± 0.43	30.16 ± 0.97
Total	100	100	100	100

After complete dissolution process of biomass waste, the mixture was appeared dark brown in color which is imparted by the dissolved lignin of the lignocellulosic matrix in the ionic liquid. Researchers were reported that the ionic liquid [Bmim]Cl and [Amim]Cl also capable to dissolve lignin (Hou et al., 2017; Moniruzzaman & Goto, 2018). Some amount of water was added for the fractionation of the lignocellulosic components. Two layer of mixtures were form as shown at Fig. 6.

Lignin has well-known properties as water impermeability, hydrophobicity and film-forming ability. Once the lignocellulose biomass was dissolved, it can be fractionated to its principle component by adding anti-solvent. In this experiment, water was used as the anti-solvent. The dissolution of lignin is more tolerant to water compared to the dissolution of cellulose. High solubility of lignin can be achieved in the mixture of ionic liquids and water, while the presence of water has negative impact on the dissolution ability of cellulose in ionic liquid. They also found that addition of applicable amount of water to ionic liquid can significantly increase the lignin solubility. This is due to the increased interaction probability between lignin and free ions, which the amount and mobility were remarkably increased after the addition of water (Hou et al., 2017). Thus, addition of water in a complete dissolution of lignocellulose with ionic liquid is applicable for fractionation of cellulose, lignin and the ionic liquid recyclability. The lignin separated from lignocellulose biomass is not only a desirable for preparation of lignin-free renewable feedstock for the production of bioenergy, but the extracted lignin also can be a promising starting material for production of novel materials. Therefore, the extraction of lignin from biomass has greatly improve the biorefinery productivity and profitability.

Further washing of cellulose solution resulted a smooth and transparent layer of regenerated cellulose as shown at Fig. 5 (a). Theoretically, the regeneration of cellulose was started when water is added in the dissolve cellulose with ionic liquid. This is because, the hydrogen bonds that formed during dissolution process, between the hydroxyl group of cellulose and Cl^- anions are dismissed by adding more H_2O . The regenerated cellulose was converted to cellulose II from cellulose I in original raw biomass. Addition of water was promoted formation of hydrogen bond of water with anions Cl^- and resulted the hydrogen bonds between cellulose are connected again leading to precipitation (Gupta & Jiang, 2015). It was observed that the surface morphology and cellulose structure of the cellulose was change significantly. Subsequently, relative homogeneous textures were displayed as shown in SEM images (Fig. 5b, d, f, h).

Table 4 showed the results of cellulose recovery from the ionic liquid and the ionic liquid recovery. The cellulose recovery percentage of SPC, BSW, SCB and OPEFB were 96.00 wt%, 91.34 wt%, 87.16 wt%, and 91.14 wt% respectively. SPC recorder the highest cellulose recovery compare to other biomass because it contained lesser amount of lignin component (less than 5 wt%) as reported previously at Table 1. It was supported by Hou et al., that dissolution of polysaccharide which are not bonded with lignin or fewer lignin composition is easier than the dissolution of supermolecular network of complex entanglement lignocellulose compounds (Hou et al., 2017).

As much as ionic liquid cost is important, the ionic liquid recycling rate is a key variable for the process economics perspective. In this experiment, the recycled ionic liquids were highly recovered by vacuum distillation. The ionic liquid recovery rate after dissolution of SPC, BSW, SCB and OPEFB were 94.0%, 95.04%, 97.12% and 98.15% individually. The purity of the recycle ionic liquids were proof by FTIR analysis at Fig. 7. The result shows that the FTIR spectra of recycled ionic is principally consistent with the original.

Table 4
The cellulose/hemicellulose and ionic liquid recovery percentage

Biomass waste	Ionic liquids	Cellulose/hemicellulose recovery (wt%)	Ionic liquids recovery (wt%)
Scrap paper cups	[Bmim]Cl	96.00	94.00
Bamboo stem wall	[Bmim]Cl	94.34	95.04
Sugar cane bagasse	[Amim]Cl	87.16	97.12
Oil palm empty fruit bunch	[Amim]Cl	91.14	98.15

Acid Hydrolysis Of Biomass Waste By H₂SO₄

The SPC, IL-SPC, IL-BSW, IL-SCB and IL-OPEFB were hydrolyzed by 10 M sulfuric acid at temperature 65 °C to produce fermentable reducing sugar. In the acid hydrolysis process, glycosidic bonds between the cellulose molecules are break to form monosaccharides. Literatures reported that, compare to other method of hydrolysis, acid hydrolysis promising higher sugar yield and good reproducibility. However, the method will produce large amount of degradation products from the monosaccharids such as furfural generated from pentose and hydroxymethylfurfural produced from hexose which further degrade into formic acid. The composition of the biomass waste hydrolysate including the sugars and degradation products were shown at Table 5. From the experimental data, IL-SPC hydrolysate contained higher glucose compare to SPC. It is because, IL-SPC cellulose has undergo ionic liquid dissolution which resulted higher surface area and disintegration of the cellulosic fibril structure then provide better accessibility for the hydrolysis process (Jeong et al., 2018; Loow et al., 2016). Almost negligible amount of xylose (0.29 g/L) was found and neither furfural nor HMF were detected in the hydrolysate of IL-SPC compare to SPC's hydrolysate. It shows that, the ionic liquid dissolution was removed most of hemicellulose components from the SPC which also contribute to high glucose conversion from IL-SPC (Kim, 2018). This result also supported by the result of biomass hydrolysis (IL-BSW, IL-SCB and IL-OPEFB) where their total sugar are higher that the SPC even though, their cellulose content are lower that the SPC. Besides glucose, sucrose, arabinose and xylose, biomass waste hydrolysate contained other unknown soluble sugar. The unknown soluble sugar may consist of other monomers of hemicellulose such as galactose and mannose. Besides monomer, the soluble sugar can also be cello-oligomer and xylo-oligomers. Oligomer is a molecule that consists of a few monomers units which can be dimer, trimer, and tetramer, depends on the number of monomers, two, three or four. Dimer can be digest by the bacteria but trimer oligomers are cannot (Nivea et al., 2006).

Table 5 Hydrolysate composition					
Biomass waste	SPC	IL-SPC	IL-BSW	IL-SCB	IL-OPEFB
	control	[Bmim]Cl	[Bmim]Cl	[Amim]Cl	[Amim]Cl
Soluble component	Liquid acid hydrolysate (g/L)				
Glucose	6.54	6.68	8.71	10.22	8.08
Xylose	2.88	0.29	1.81	1.34	1.25
Arabinose	N.D.	0.25	N.D.	0.06	0.02
Sucrose	2.06	1.69	2.30	2.20	1.25
Unknown soluble sugar	1.85	11.06	7.39	6.23	6.12
Formic acid	N.D.	N.D.	N.D.	N.D.	0.02
Acetic acid	N.D.	N.D.	N.D.	N.D.	0.01
Propionic acid	N.D.	N.D.	N.D.	0.88	N.D.
Butyric acid	N.D.	N.D.	N.D.	N.D.	N.D.
Furfural	N.D.	N.D.	N.D.	N.D.	N.D.
T-sugar	13.33	19.97	20.21	20.93	17.05
N.D. : Not Detectable					

Fermentation for biohydrogen production

Biohydrogen producing bacterium *Clostridia* sp. was used to examine the suitability of sugar feedstock from acid hydrolysis of SPC, IL-SPC, IL-BSW and IL-SCB for biohydrogen production. Control set was established in the experiment which the fermentation was fed with pure glucose as carbon source. The biohydrogen production profile are illustrated in Fig. 8. As shows at Table 6, the final measured biogas volume using SPC, IL-SPC, IL-BSW and IL-SCB hydrolysate were 196.67, 174.82, 117.21 and 106.32 ml H₂/100 ml respectively. While 174.91 ml H₂/100 ml was produced from the control. Almost 95% of substrate (pure glucose) was utilized by the bacteria while 85.44, 80.37, 83.68 and 80.41% substrate was utilized by the bacteria from the SPC, IL-SPC, IL-BSW and IL-SCB hydrolysate individually. Lower sugar utilization rate of biomass waste hydrolysate may be due to the complexity of the sugar composition; some of which have larger molecular weights, which may result in the inefficient use by the organisms. Table 7 shows that the ethanol content of SPC, IL-SPC, IL-BSW and IL-SCB which are 4.68, 10.9, 20.41, 17.35% respectively. The presence of ethanol in fermentation inhibited some hydrogen-producing bacteria, this can be the reason of lower hydrogen production of IL-SPC, IL-BSW and IL-SCB compare to SPC. During fermentation, the sugars were broken down to pyruvate. The pyruvate then diverted towards either the acidogenesis pathway or solvotogenesis pathway. Acidogenesis leads to formation of butyric acid and acetic acid; producing biohydrogen and carbon dioxide gas. Formation of other reduced fermentation metabolites such as ethanol and other alcohols (solvotogenesis) lower biohydrogen yield. Thus for maximum biohydrogen yield, fermentation parameters must be manipulated to divert the metabolism pathway towards producing butyric acid and acetic acid as the final fermentation product (Abdul et al., 2016). This report is the first of a trial of biohydrogen production from SPC and IL-SPC hydrolysates. Optimisation of the fermentation process will bring higher yields of biohydrogen.

Table 6 Kinetic parameters for hydrogen production with different hydrolysates				
Substrate	P (mL/100 mL)	R _m (mL/h/100 mL)	λ (h)	R ²
Glucose	174.91	3.04	31.67	0.9973
SPC	196.67	3.44	21.83	0.9968
ILs-SPC	174.82	3.98	34.79	0.9989
ILs-BSW	117.21	1.64	30.07	0.9924
ILs-SCB	106.32	6.37	39.00	0.9941
*Kinetic parameters calculated from Gompertz fitting; P : Cumulative hydrogen productions, R _m : hydrogen production rate, λ : later period				

Table 7
Liquid metabolites of biohydrogen production

Substrate	T	Substrate (g/L R-sugar/L)	pH		HLA (%)	HAc (%)	HPr (%)	EtOH (%)	HBu (%)	H ₂ yield (mol H ₂ /mol T-sugar)	Substrate Utilization (%)
			Initial	final							
Glucose	37	20	7.00	4.58	26.25	6.1	0	9.14	50.74	0.68	94.81
SPC	37	20	6.98	5.74	7.05	46.05	3.23	4.68	38.98	0.95	85.44
ILs-SPC	37	20	7.00	6.32	2.09	38.37	0.75	10.9	47.79	0.85	80.37
ILs-BSW	37	20	7.00	5.50	5.64	37.08	11.23	20.41	25.64	0.51	83.68
ILs-SCB	37	20	7.00	5.31	9.33	32.78	9.1	17.35	26.89	0.49	80.41
T: temperature (°C), HLA : lactic acid, HAc : acetic acid, HPr : propionic acid, HBu : butyric acid, EtOH : ethanol, SPC : Scrap paper cup											

Conclusion

This study provides the new concept in utilization municipal solid waste and agricultural plant residues by using green solvent ionic liquid to dissolve the cellulose. Biomass composition analysis by TGA method seems to be quite reliable because the value where close to the data from Van Soest method. The experiments show that the laboratory prepared imidazole ionic liquid [Amim]Cl and [Bmim]Cl were successfully dissolved the SPC, BSW, SCB and OPEFB. Efficiency of prepared ionic liquids were proved with high cellulose recovery and reusability of the ionic liquids. To prepare fermentable sugar that can be consumed by the biohydrogen producing bacteria, acid hydrolysis of the regenerated cellulose was hydrolysed by 10% sulfuric acid which resulted up to 96% sugar conversion. IL-SPC hydrolysate reported higher total sugar conversion compare to SPC (control) due to higher surface area and disintegration of the cellulosic fibril structure resulted from the dissolution process. IL-SPC, IL-BSW, IL-SCB and IL-OPEFB hydrolysate contained higher total sugar compared to SPC hydrolysate even though their cellulose content are lower that the SPC (control). In the fermentation process, almost 85% of biomass waste hydrolysate substrate was utilized by the bacteria and producing up to 196 ml H₂/100 ml cumulative biohydrogen. As compare to control fermentation, the bacteria utilized

94.81% substrate, but producing lower cumulative biohydrogen which is 174.91 ml H₂/100 ml. The biohydrogen fermentability results show that the recovered cellulose from the biomass waste is compatible to be used as a source of sugar in producing bio-hydrogen. This method of recycling cellulose from municipal solid waste and agriculture biomass was a new alternative for renewable energy resources, and provide new solutions to environmental pollution problems.

Abbreviations

Scrap paper cup: SPC; bamboo stem wall: BSW; sugar cane bagasse: SCB; oil palm empty fruit bunch: OPEFB; 1-allyl-3-methylimidazolium chloride: [Amim]Cl; 1-butyl-3-methylimidazolium chloride: [Bmim]Cl; ionic liquid treated scrap paper cup: IL-SPC; ionic liquid treated bamboo stem wall: IL-BSW; ionic liquid treated sugar cane bagasse: IL-SCB; ionic liquid treated oil palm empty fruit bunch: IL-OPEFB; thermal gravimetric analysis: TGA; neutral detergent fiber: NDF; acid detergent fiber: ADF; acid detergent lignin: ADL; temperature: T; lactic acid: HLa; acetic acid: HAc; propionic acid: HPr; butyric acid: HBu; ethanol: EtOH

Declarations

Ethics approval and consent to participate

Not applicable. The study does not involve the use of any animal or human data or tissue.

Consent for publication

Not applicable. The manuscript does not contain any other individual person's data.

Availability of data and materials

All data generated or analyzed during this study are included in the main manuscript file.

Competing interests

The authors declare that they have no competing interests.

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Author's contributions

Shu-Yii Wu: Conceptualization, experiments design, research methodology and analysis, original manuscript draft, review and editing.

Mastura Abd Manaf: Preparation of manuscript draft, visualization, writing and editing.

Sheng-Kai Lin, Ji Gang Chen, and Kai-Hsiang Yang: Conduct most of experiments and analysis, report the research progress.

Wei-Lun Zeng, Yi-Feng Lin: Data analysis

Shuhaida Harun: Review the prepared manuscript draft.

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Figures

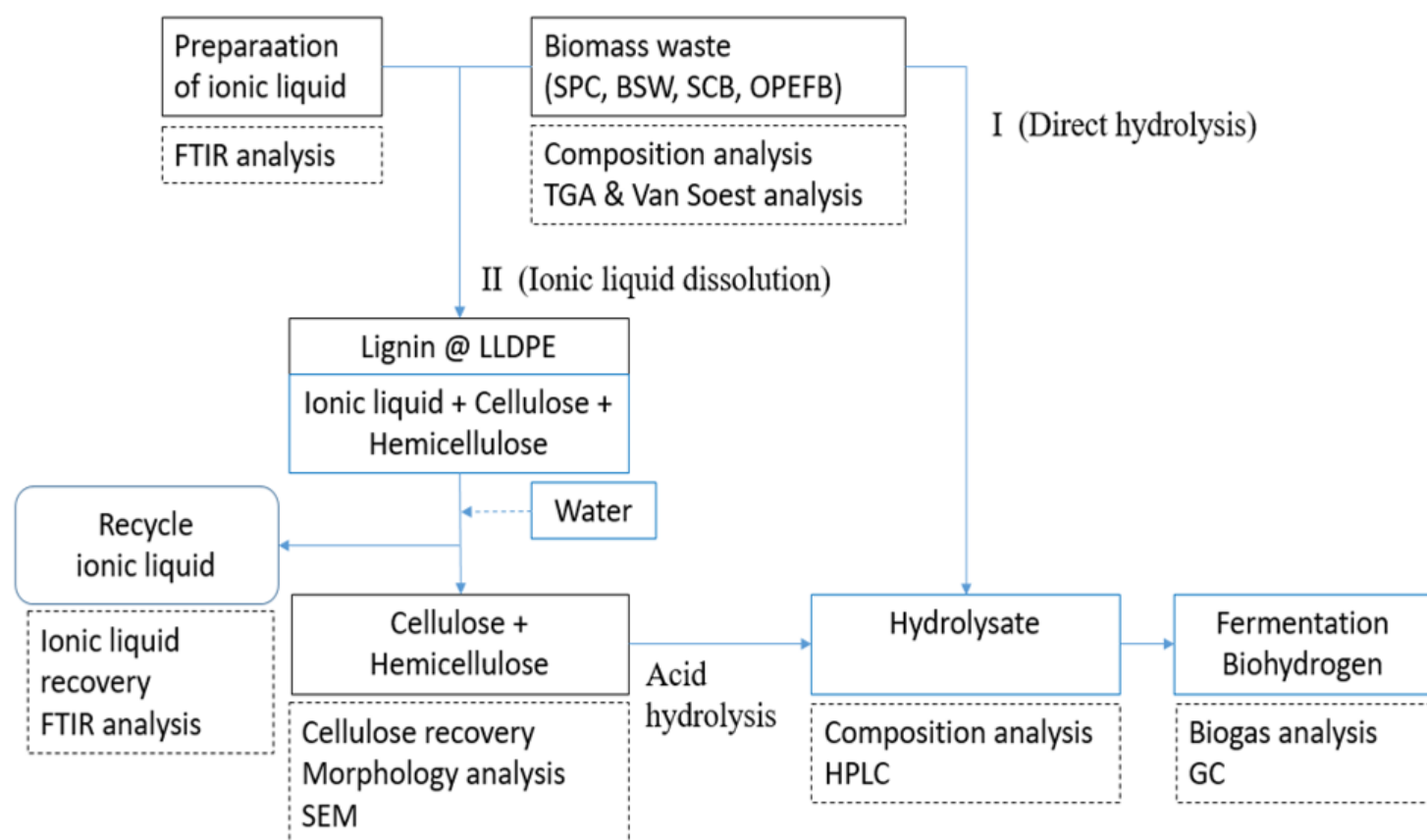


Figure 1

Experiment flow chart

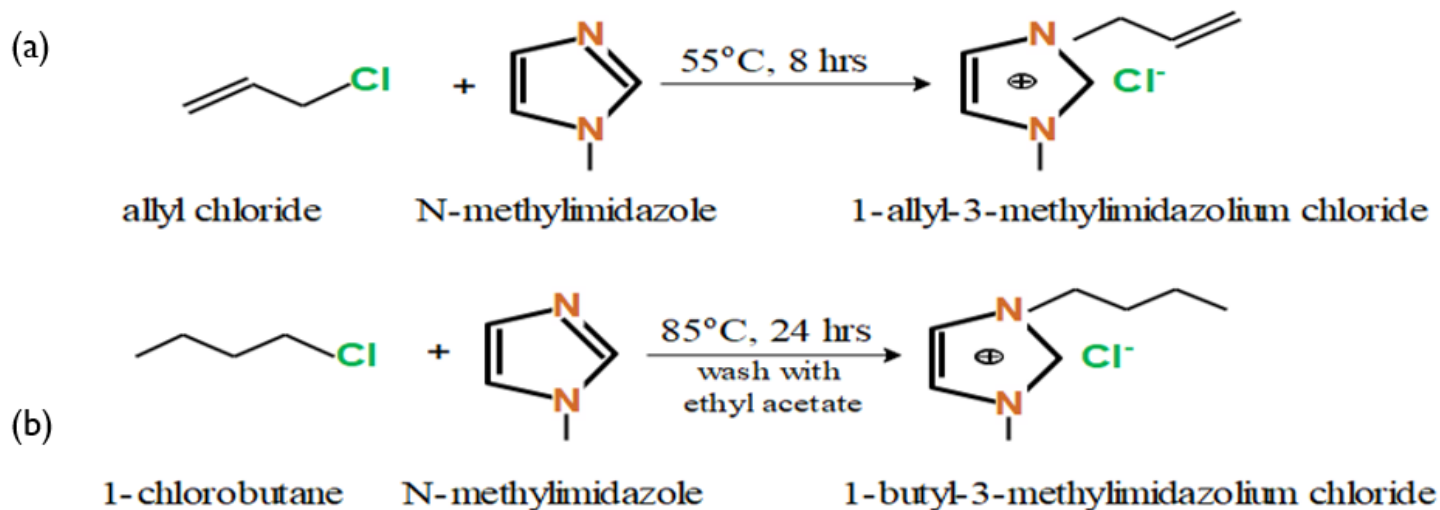


Figure 2

Synthesis of ionic liquids. (a) [Amim]Cl (b) [Bmim]Cl (Xing et al., 2014)

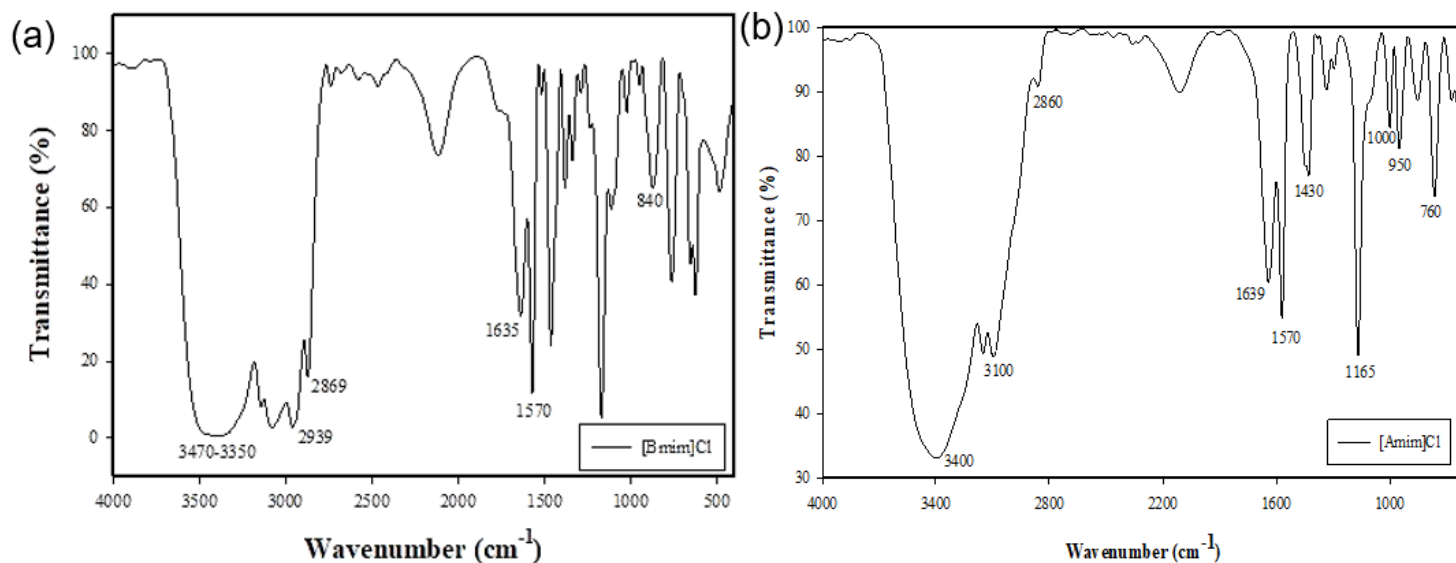


Figure 3

FTIR spectrum of (a) [Bmim]Cl (b) [Amim]Cl

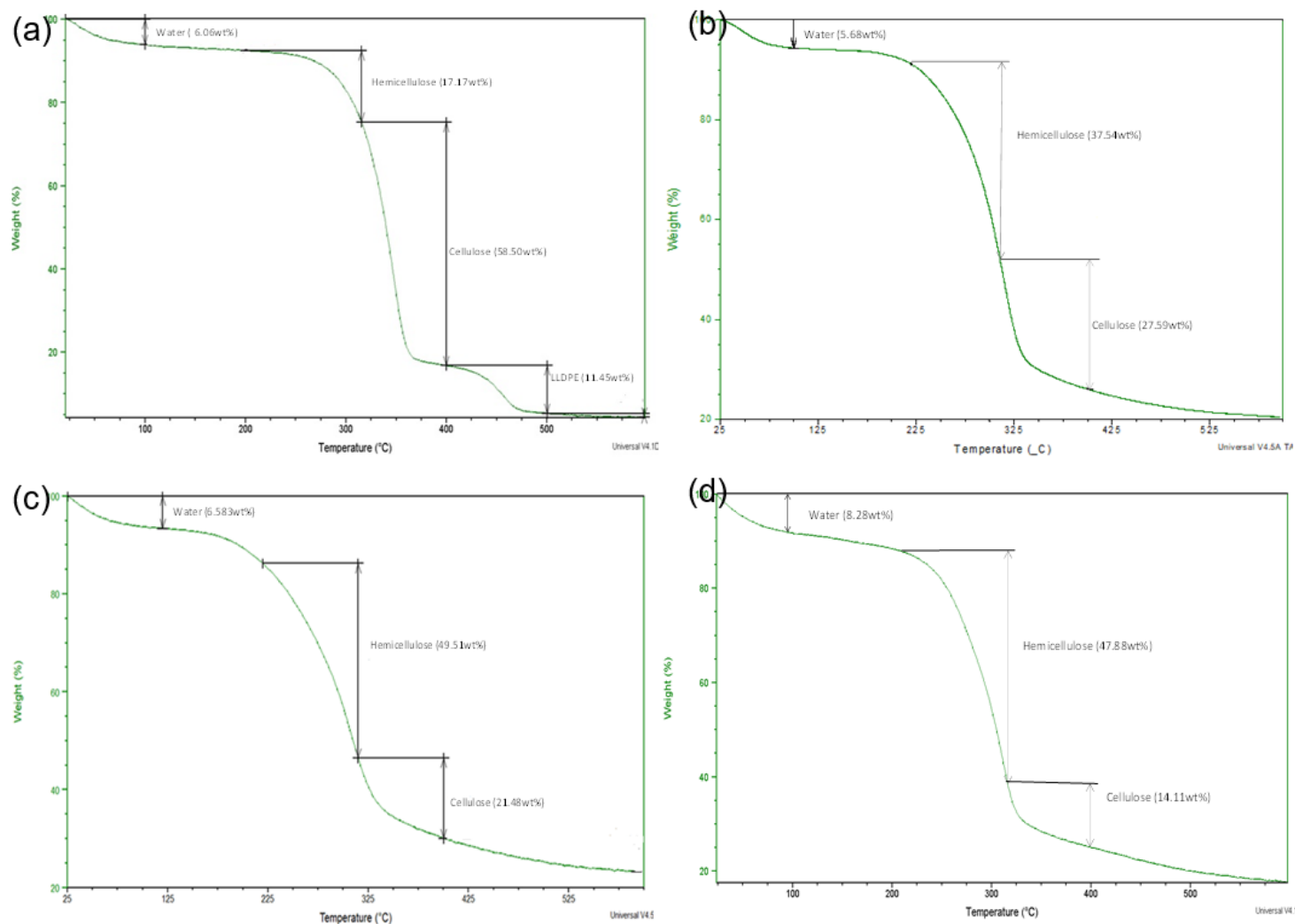


Figure 4

TGA analysis for (a) scrap paper cups; (b) bamboo stem wall; (c) sugar cane bagasse; (d) oil palm empty fruit bunch

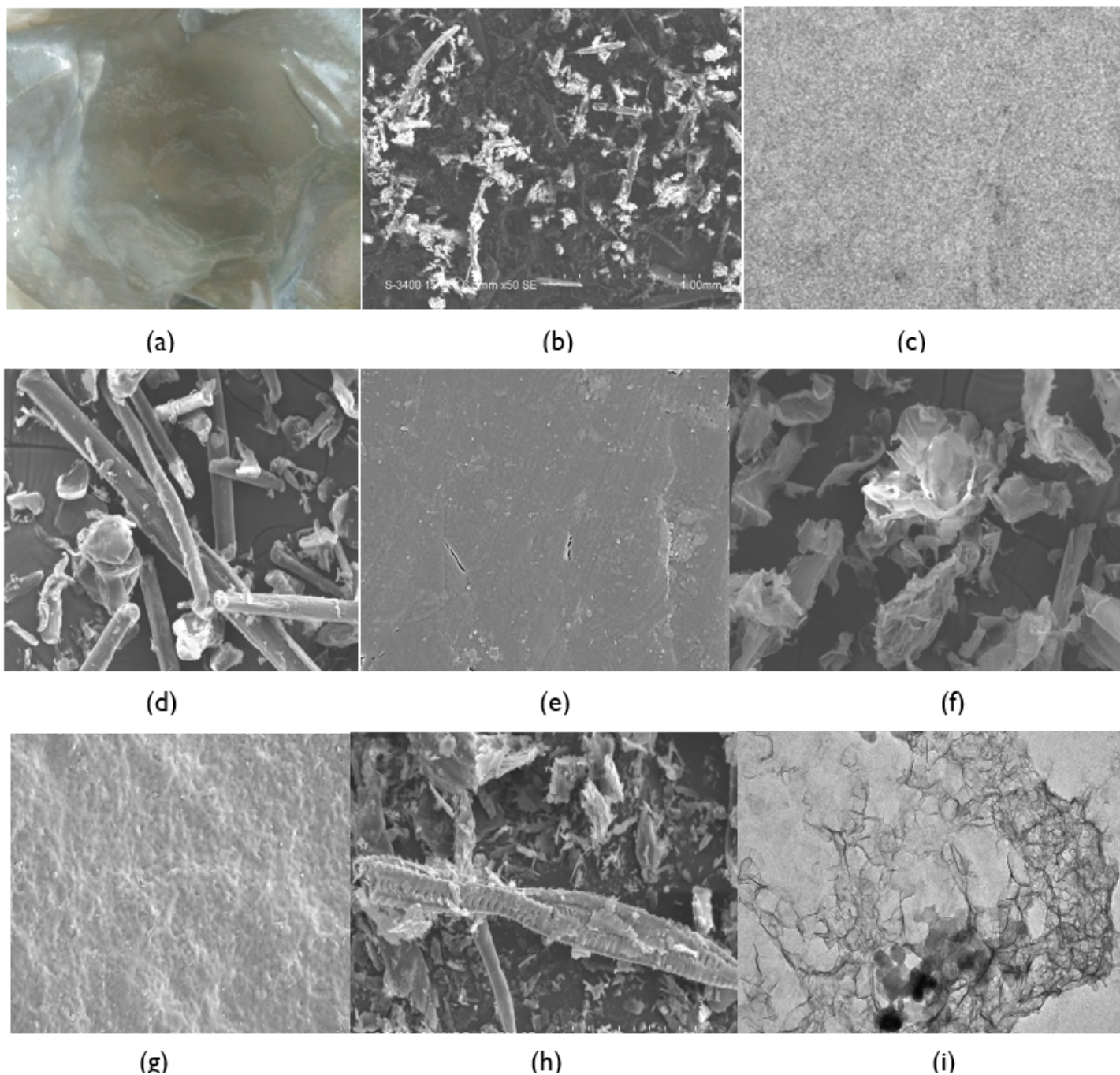


Figure 5

(a) transparent layer of regenerated cellulose after washing with water. SEM images of biomass waste before and after ionic liquid dissolution (b) raw SPC 50x (c) SPC regenerated cellulose/hemicellulose 200x optical microscope (d) raw BSW 500x, (e) BSW regenerated cellulose/hemicellulose 3000x, (f) raw SCB 500x (g) SCB regenerated cellulose/hemicellulose 3000x, (h) raw OPEFB 500x (i) OPEFB regenerated cellulose/hemicellulose 10000x

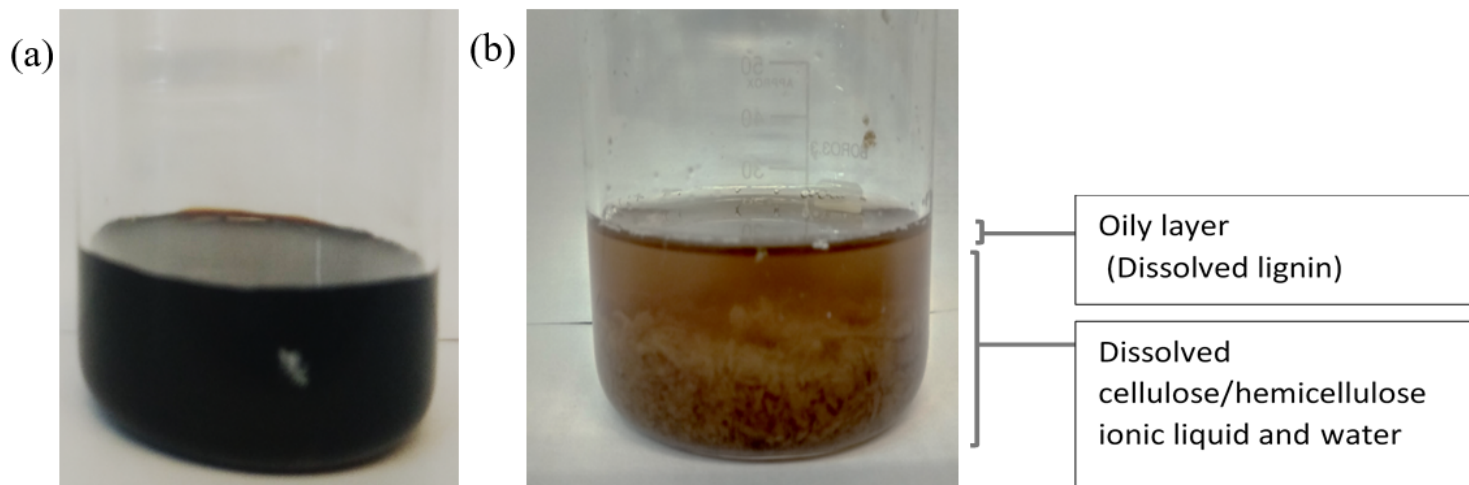


Figure 6

(a) Dissolved biomass waste in ionic liquid (b) Fractionation of dissolved biomass waste with ionic liquid and water

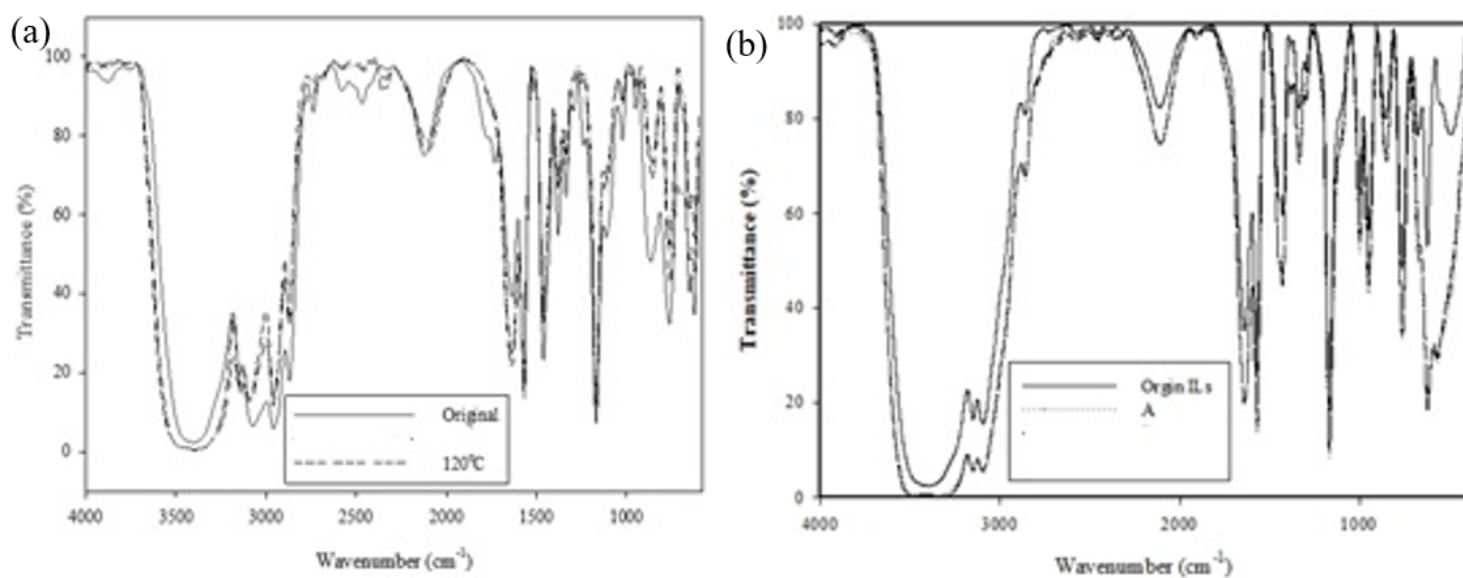


Figure 7

FTIR spectrum of fresh and recycle ionic liquid (a) [Bmim]Cl (b) [Amim]Cl

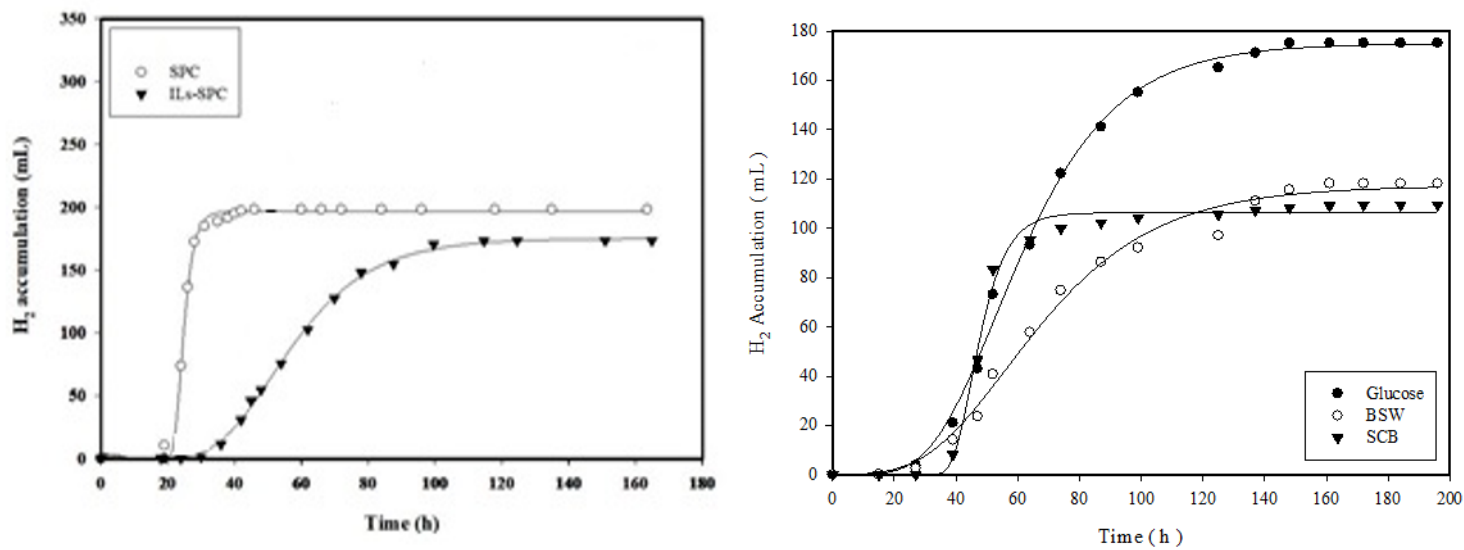


Figure 8

Biohydrogen production curve of different matrix carbon sources

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