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Combined pretreatment with hot compressed water and wet disk milling opened up oil palm biomass structure resulting in enhanced enzymatic digestibility

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HIGHLIGHTS

- OPEFB and OPFF were hydrothermally treated under isothermal and non-isothermal.
- Hemicellulose dissolution more pronounced at higher temperature.
- Subsequent WDM of HCW-pretreated oil palm biomass unravels fiber structures.
- Fibrillation of pretreated oil palm biomass improved over milling cycles.
- 88.5% and 100% of total sugars obtained from OPEFB and OPFF at 150 °C, 240 min.

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ABSTRACT

Combined pretreatment with hot compressed water and wet disk milling was performed with the aim to reduce the natural recalcitrance of oil palm biomass by opening its structure and provide maximal access to cellulase attack. Oil palm empty fruit bunch and oil palm frond fiber were first hydrothermally pretreated at 150–190 °C and 10–240 min. Further treatment with wet disk milling resulted in nanofibrillation of fiber which caused the loosening of the tight biomass structure, thus increasing the subsequent enzymatic conversion of cellulose to glucose. The effectiveness of the combined pretreatments was evaluated by chemical composition changes, power consumption, morphological alterations by SEM and the enzymatic digestibility of treated samples. At optimal pretreatment process, approximately 88.5% and 100.0% of total sugar yields were obtained from oil palm empty fruit bunch and oil palm frond fiber samples, which only consumed about 15.1 and 23.5 MJ/kg of biomass, respectively.

1. Introduction

Oil palm biomass is a renewable resource, cheap and available throughout the year, and not properly-utilized with lower impact value (Lee and Ofori-Boateng, 2013). Therefore, appropriate technologies for biomass upgrading is sought to increase the value and to promote sustainable growth of the oil palm industry. Lignocellulosic biomass is an importance source of natural biopolymers on the earth and their rigid and intact structures of cel lulose–hemicellulose–lignin complexes are linked with various types of chemical bonds which protect them from chemical and biological degradation (Mosier and Wyman, 2005). A number of technologies have been reported in the literature to fractionate lignocellulosic biomass and the development of chemical-free approaches with environmentally-sound technology. Furthermore low energy consumption with consideration of maximum recovery of pentose and hexose sugars are currently in progress (Hideno et al., 2009; Lee et al., 2010; Kumagai et al., 2014).

Oil palm empty fruit bunch (OPEFB) and oil palm frond fiber (OPFF) have been treated by various methods such as mechanical, chemical and thermochemical pretreatment process (Palamae et al., 2014; Cheng et al., 2014; Ofori-Boateng and Lee, 2014;

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Sabiha-Hanim et al., 2011). Previous ball milling pretreatment process demonstrated high energy usage with high recovery of both pentose and hexose sugars (Zakaria et al., 2014a). Hydrothermal pretreatment alone achieved high conversion of cellulose to glucose at higher pretreatment temperature with loss of pentose sugars by conversion to furfural and formic acids (Zakaria et al., 2015a). Hydrothermal pretreatment with subsequent wet disk milling (WDM) exhibited excellent recovery of pentose and hexose sugars with lower cellulase loadings (Lee et al., 2010; Hideno et al., 2012; Weiqi et al., 2013; Kumagai et al., 2015). Superheated steam (SHS) treatment was tested on oil palm mesocarp fiber (OPMF) and the glucose conversion yield was not pronounced in comparison to hot compressed water (HCW) due to minimal contact of water that reacts as catalyst in the hydrolysis of hemicellulose component. Combination of pretreatment using HCW (180 °C for 20 min) and WDM were the best method to recover the highest xylose (90%) and glucose (86%), and was preferred for pretreatment of OPMF (Zakaria et al., 2015b).

Therefore in this study, individual pretreatment of HCW, WDM and combination of them were conducted to understand and improve the conversion of hemicellulose and cellulose from OPEFB and OPFF samples. HCW was performed under isothermal and non-isothermal conditions to compare the behavior of the hemicellulose degradation by-products generated from pretreated liquids and properties of reactive cellulose from pretreated solids. Energy requirement by WDM from different pretreatment severities were thoroughly compared and discussed.

2. Methods

2.1. Hot compressed water (HCW) pretreatment

The OPEFB was collected from Seri Ulu Langat Palm Oil Mill, Dengkil, Selangor, Malaysia, and the OPFF was obtained from the oil palm plantation at Universiti Putra Malaysia, Serdang, Selangor, Malaysia. The samples were prepared as discussed earlier (Zakaria et al., 2014a) prior to component analysis. Hot compressed water (HCW) pretreatment was performed using a 1 L stainless steel autoclave (Nitto Koatsu Co., Tsukuba, Japan) (Zakaria et al., 2015b). The oil palm biomass was pretreated at temperature ranges from 150 to 190 °C for 10 and 240 min, respectively. The pretreated solids were separated from the slurry by using Advantec No. 2 membrane filter and rinsed with distilled water until neutral pH. The soluble sugars in pretreated liquids were identified by HPLC as describing below. The pretreated solid portion recovered after filtration was subjected to subsequent treatment using wet disk milling. The intensity of the hydrothermal treatment was expressed as severity factor ($\log R_o$). The severity parameters corresponding to different hydrothermal pretreatment conditions are calculated as in Eq. (1), in which t is the reaction time (min), and T is the hydrolysis temperature (°C) (Overend and Chornet, 1989).

$$R_o = t \exp[(T - 100)/14.75] \quad (1)$$

2.2. Wet disk milling (WDM) pretreatment

Wet disk milling was performed using Supermass colloider MKZA10 (Masuko Sangyo Co., Ltd., Saitama, Japan) following the method reported earlier (Zakaria et al., 2015b). The pretreated samples were centrifuged at 10,000g for 15 min and the solids recovered were subjected to freeze-drying prior to enzymatic hydrolysis and other analysis.

2.3. Enzymatic hydrolysis

Enzymatic hydrolysis was performed using an enzyme cocktail constituting 40 FPU/mL *Acremonium* cellulase (Meiji Seika Co, Japan), and 10% Optimash BG (Genencor International, California, USA) as reported previously (Zakaria et al., 2014b). The enzymatic hydrolysis was performed at 50 °C for 72 h with stirring/shaking. The experiment was performed in triplicates and the results are presented as the average values. The enzymatic digestibility was evaluated by the obtained sugars (mg sugars/g materials) or sugar yield as calculated using Eq. (2):

$$\text{Sugar yield}(\%) = [\text{weight of monomeric sugars after enzymatic hydrolysis/weight of total monomeric sugars of biomass after hydrolysis using } H_2SO_4] \times 100 \quad (2)$$

2.4. Energy consumption in WDM pretreatment

The actual measurement values of power consumption during WDM were used for energy calculation as reported earlier (Hideno et al., 2009).

2.5. Analytical methods

Detection of monomeric and oligomeric sugars, acetic acid, furfural, 5-HMF, and formic acid from untreated materials, HCW-treated and enzymatic hydrolysis were performed using high-performance liquid chromatography (HPLC) according to the analytical method (NREL/TP-510-42623) (Sluiter et al., 2008). Morphological analysis was conducted by using scanning electron microscope (SEM; S-3400 and S-4800, Hitachi, Japan) at 1.0 kV. The preparation of fibrillated pretreated samples was performed as reported earlier (Zakaria et al., 2015b).

3. Results and discussion

3.1. Effect of pretreatment severities on pretreated liquids

Previous works reported hydrothermal pretreatment of oil palm biomass under non-isothermal conditions with short reaction time (Zakaria et al., 2015a). In contrast, in the present study oil palm biomass is hydrothermally pretreated under both isothermal and non-isothermal conditions with mild reaction temperature and longer reaction time to understand the degradation mechanism of hemicellulose and degradation by-products formation as well as reactive cellulose recovered. Formation of hydrolyzed sugars from OPEFB and OPFF samples are presented in Table S1. In general, both monomeric and oligomeric sugars were increased with higher pretreatment severities (either higher temperature or longer reaction time). The highest monomeric and oligomeric sugars detected were xylose, followed by glucose, arabinose, galactose and mannose. The highest xylo-oligomers (XOS) detected was 4.8 g/100 g of OPEFB samples at 190 °C, 20 min ($\log R_o = 4.0$) (Table S1). This value is equivalent to 30.8% of XOS from OPEFB samples. Meanwhile, XOS generated from OPFF was 4.5 g/100 g of OPFF at 190 °C, 10 min ($\log R_o = 3.6$) which is about 26.6% from OPFF sample. It was obvious that the production of XOS was greater at the higher temperature than reaction time. This is because hemicellulose degradation takes place at or greater than 180 °C (Bobleter, 1994). The monomeric and oligomeric glucose, arabinose, galactose and mannose levels were higher from OPFF samples compared to OPEFB probably due to the differences in hemicellulose components, amounts and molecular structures of the fibers. A lower XOS production from OPEFB recorded in this