VILNIUS TECH
Vilnius Gediminas Technical University

Journal of Environmental Engineering and Landscape Management
ISSN 1648–6897 / eISSN 1822-4199
2022 Volume 30 Issue 2: 276–283
https://doi.org/10.3846/jeelm.2022.16744

Special issue on *Sustainable Environmental Processes and Technologies*

SUSTAINABLE ENERGY-EFFICIENT CONVERSION OF WASTE TEA LEAVES TO REDUCING SUGAR: OPTIMIZATION AND LIFE-CYCLE ENVIRONMENTAL IMPACT ASSESSMENT

Rajat CHAKRABORTY*, Oindrila ROY

Chemical Engineering Department, Jadavpur University, 700032 Kolkata, India

Received 27 December 2020; accepted 12 July 2021

Highlights

- Effective delignification of waste tea leaves for cellulose synthesis at mild conditions.
- Energy-efficient rapid photocatalytic-hydrolysis in quartz halogen irradiation (QHI).
- Optimized process parameters for cellulose hydrolysis into reducing sugar.
- Remarkable 17.25% more TRS yield and 244.8% lower energy-input for QHI.
- LCA ascertains environmental sustainability of waste tea leaves conversion process.

Abstract. Innovative protocols involving energy-proficient pretreatment of waste tea leaves (WTL) for preparation of cellulose and its subsequent photocatalytic hydrolysis (PH) for production of total reducing sugar (TRS) have been reported. The WTL was subjected to alkali pretreatment (60 °C, 1 h) followed by bleaching (employing peracetic acid, 65 °C, 2 h) in a quartz halogen irradiated batch reactor (QHIBR) for efficient separations of lignin and hemicellulose fractions to produce WTL derived cellulose fiber (WTLDCF; 94.5% cellulose). Consequent PH of WTLDCF in QHIBR using combination of Amberlyst-15 and nano-TiO_2 catalysts was optimized (parameters: 40 min, 70 °C, 1:30 WTLDCF to water weight ratio and 5 wt. % catalyst concentration) employing Taguchi design that provided maximum 68.25% TRS yield. The QHIBR demonstrated faster hydrolysis and superior energy-efficiency over conventional reactor owing to quartz halogen irradiation. Life cycle assessment indicated an acceptable global warming potential of 2.215 kg CO_2 equivalent; thus, establishing an energy-efficient environmentally sustainable WTL valorization process.

Keywords: waste tea leaves, photocatalytic hydrolysis, quartz halogen irradiation, total reducing sugar, energy-efficiency, environmental impact assessment, environmental sustainability.

Introduction

Sustainable production of energy and platform chemicals from Lignocellulosic biomass (LB) has become immensely popular (Chheda et al., 2007; Manzanares, 2020) as biomass conversion is associated with net lower greenhouse gas emission compared to fossil fuels; as CO_2 released during energy conversion gets consumed in subsequent biomass re-growth. LBs obtained from agricultural and household wastes are competitive feedstocks for the production of several fuel additives such as bioethanol or biobutanol through an integrated biorefinery approach (Verardi et al., 2020). One of the main components of LB is cellulose which is one of the key substances for efficient synthesis of many valuable platform chemicals such as glucose, total reducing sugar (TRS), HMF, Levulinic acid etc. (Qian et al., 2013; Hassan et al., 2015). Lignocellulosic biomass (LB) is typically composed of cellulose, hemicellulose, lignin and other extractive fractions. Tea leaves are potential feedstock for synthesis of TRS, as they consist of significant amounts of cellulose (40–50%) along with hemicellulose (25–30%), and lignin (15–20%) (Fatehi, 2013; Menon & Rao, 2012). Hence, effective hydrolysis of cellulose into TRS can yield many valuable platform chemicals such as levulinic acid, 5-hydromethoxy furfural (Yan et al., 2015; Liu et al., 2013). LB was hydrolyzed using both homogenous (Harris & Kline, 1949) and heterogeneous catalysts to obtain valuable platform chemicals (Goswami et al., 2015; Guo et al., 2012a). Nonetheless, according to some previous studies, heterogeneous catalysts

---

*Corresponding author. E-mails: *zrajat_chakraborty25@yahoo.com; rajat.chakraborty@jadavpuruniversity.in*

are advantageous over homogenous catalysts as they are less corrosive, produce lesser waste stream substances and easier to recycle (Shrotri et al., 2018). Although, ionic liquids have also been promising in the hydrolytic depolymerization of cellulose into TRS (Cai et al., 2012; Guo et al., 2012b; Liu et al., 2013); nonetheless, the overall process is expensive owing to high cost of ionic liquids.

Recent studies have suggested that application of microwave radiation can accelerate the hydrolysis reaction (Zhang & Zhao, 2009; Bian et al., 2014). It was claimed that, the application of microwave radiation (MR) could accelerate the hydrolysis reaction; unfortunately, relatively high power input was required to achieve the desired TRS yield. Tsubaki et al. (2016) and Sun et al. (2015) obtained maximum 60 and 44.6 wt. % TRS yields through hydrolysis of corn starch and microcrystalline cellulose respectively applying 1 kW microwave radiation. Notably, Fan et al. (2013) could merely attain 10 wt. % TRS yield from cellulosic waste under MR at high pressure (300 psi) and temperature (220 °C); thus, making the process energy-intensive. In recent years, our research group has effectively utilized low-energy far infrared radiation for biodiesel and glyceryl laurate production, which seems attractive owing to lesser conversion time and more energy-efficiency compared to conventionally heated reactor (Pradhan et al., 2016; Chakraborty & Mandal, 2015).

Waste tea leaves (WTL) had been effectively used as adsorbent for dye removal (Hameed, 2009; Zuorro et al., 2013), heavy metal removal (Amarasinghe & Williams, 2007; Utomo & Hunter, 2006) and also as antioxidant (Farhoosh et al., 2007; Bharti & Singh, 2020; Zandi & Gordon, 1999). Besides, supercapacitor electrodes were derived from WTL (Peng et al., 2013; Inal et al., 2015). Noticeably, for the hydrolysis of cellulose, high crystallinity inhibits its hydrolytic degradation; thus, pre-treatment of the WTL are highly necessary in order to decrease the crystallinity of the cellulose fibre, which in turn can enhance the availability of the β-1,4-glycosidic bond for efficient degradation to valuable chemicals e.g. TRS. Ball milling was found to enhance the non-crystallinity of WTL derived cellulose fibre (WTLDCF) to a limited extent (Zhao et al., 2006).

Taguchi orthogonal design array (TODA) has been increasingly used for process optimization and also for evaluation of parametric interactions in governing the response or process output (Engin et al., 2008). The effects of individual process variables on the response variable could also be estimated by TODA (Lin et al., 2009).

Life cycle assessment (LCA) has become an imperative tool in assessing climate change potential and energy consumption (Borrion et al., 2012) of a process. LCA is a procedural tool used to quantitatively analyse the life cycle of a product or a process within a standard charter provided by ISO 14040 and 14044 (Finkbeiner et al., 2006). The major findings of LCA include the greenhouse gas (GHG) emission associated to a product development. Literature has already suggested that LB accounts for lower GHG emission compared to a fossil fuel release (Manzanares, 2020).

To the best of our knowledge, no study has yet been reported on heterogeneous catalytic hydrolysis of WTL to produce cellulose and subsequently TRS. Although several pretreatment methods for LB have been reviewed previously (Silveira et al., 2015) for deconstruction of plant cell wall; however, application of non-ionising infrared radiation (e.g. quartz–halogen radiation) was not reported. Besides, no scientific report is available on the combined application of heterogeneous acidic (Amberlyst-15) and photo (nano-TiO_2)-catalytic degradation under non-ionising, energy-efficient quartz–halogen radiation for intensification of WTL hydrolysis towards TRS synthesis.

In the present article, the optimization of WTL hydrolytic depolymerization to yield WTL derived cellulose fibre (WTLDCF) through pretreatment and consequent photocatalytic hydrolysis of WTLDCF to maximize TRS yield have been explored through one-pot conversion employing non-ionizing, energy-saving quartz halogen irradiated stirred batch reactor (QHIBR). The optimal process parameters pertaining to pretreatment and subsequent hydrolysis of WTLDCF for maximum TRS yield have been evaluated by TODA. The energy-efficient performance of the QHIBR in one-pot batch TRS synthesis has been assessed and compared with conventional thermal source assisted batch reactor (CTSABR) in terms of TRS yield and energy consumption. The LCA has been performed aiming at the optimal TRS production considering the "gate to gate" as the "scope" for evaluating the environmental impacts occurring within the process boundary only. The major "goal" of the LCA study was to evaluate the "hotspot", i.e. the particular process that had maximum contribution to global warming potential GWP. Besides, other environmental impact parameters viz. acidification potential (AP), eutrophication potential (EP), ozone layer depletion potential (ODP) and photochemical ozone creation potential (PCOP) were also evaluated to assess the environmental impact and sustainability.

1. Experimental

1.1. Materials and methods

The waste tea leaves (WTL) were collected from a local tea shop at Kolkata, India. Analytical reagent grade chemicals viz., glacial acetic acid, hydrogen peroxide (30%), DNS (di-nitro salicylic acid), aqueous NH_4OH (25%), potassium sodium tartrate etc. were procured from Merck (India); while, Amberlyst-15 (acid catalyst) and nano-titania (NT) (photocatalyst) were purchased from Sigma Aldrich.

1.2. Reactor configuration

The pretreatment and subsequent photocatalytic hydrolysis of WTL were conducted in a non-ionizing quartz–halogen irradiator (QHI; 145 W; wavelength: 360–2000 nm) equipped batch reactor (QHIBR). The QHIBR had a

250 ml three-necked round bottom flask which was fitted with a mechanical stirrer for mixing of the reaction mixture. The speed range of the stirrer was in the range of 200–1400 rpm. A thermocouple was fitted in the reactor in order to measure the temperature of the reaction vessel. A proportional-integral-derivative temperature controller was fitted to keep the temperature of the vessel at a desired magnitude. For comaparative hydrolysis performance analysis, an otherwise similar batch reactor setup was provided with a 500 W (immersed in a water-bath) electric coil as a conventional thermal source (herein after referred to as the conventional thermal source (CTS) assisted batch reactor (CTSABR)) instead of the QHI.

1.2.1. Extraction of cellulose fibers from WTL

The WTL, collected from a local shop was washed thoroughly with deionized water and dried in a hot air oven at 80 °C. Ball milling of the WTL was performed in a stainless steel coated ball mill, equipped with three ZrO_2 balls for 3 h. Mechanical sieving was done to obtain WTL particle size –240+300 BSS. For the pretreatment, ball milled WTL were taken in a three-neck round bottom vessel and 8% aqueous ammonia solution was poured through a pippet, along with DI water. The pretreatment was performed in the QHIBR for 1 h where the rotational speed of the stirrer was kept at 400 rpm. After the specified time, the reaction vessel was flushed with nitrogen gas to remove the presence of any excess ammonia. The pretreated-WTL (PWTL) were obtained as filter cake and oven dried at 80 °C. After that, for the delignification process, the PWTL were taken in the QHIBR alongside deploying peracetic acid (99% glacial acetic acid: 30% H_2O_2 = 7:3 v/v) with PWTL fibre to peracetic acid ratio of 50:1(w/v) at 65 °C for 2 h. Next, the WTL derived cellulose fibres (WTLDCF) were obtained through filtration and thoroughly cleaned with DI water to remove the unreacted acid.

1.2.2. Photocatalytic Hydrolysis (PH) of WTLDCF

For the PH reaction, measured amount of WTLDCF along with water and A15 and NT (1:1) were added in the QHIBR for specified time and temperature (Table 1). After the completion of the reaction the liquid product was separated from the catalyst and residual solid by vacuum filtration method. The filtrate was tested for the presence of TRS by standard DNS method. The PH was also carried out in the CTSAR by keeping all the parameters unaltered. This study was separately carried out to find out the influence of quartz-halogen irradiation on the photocatalytic depolymerization (hydrolysis) of cellulose and to check whether a non-conventional reactor was more energy-saving than a conventional reactor or not.

1.2.3. Preparation of DNS for TRS yield estimation

To prepare DNS solution, 125 mL of deionized (DI) water was taken and gradually 45.5 g of potassium sodium tartrate, 1.6 g 3,5-dinitrosalicylic acid and 65.5 mL 2N NaOH were added to it. The solution was then heated to 50 °C in a conventional water bath heater. Subsequently, 1.25 g phenol and 1.25 g sodium sulphite were mixed with the solution and stirred until uniform homogeneity was achieved. Next, the solution was cooled down to room temperature and 250 mL of DI water was added for dilution to obtain the final DNS solution (Miller, 1959).

1.3. Characterizations

1.3.1. Compositions of WTL and WTLDCF

The chemical compositions of WTL and WTLDCF were studied. For the compositional analysis of holocellulose i.e, cellulose and hemicellulose, WTL were treated with acidified aqueous sodium chlorite ($NaClO_2$) to degrade the lignin fraction. The $NaClO_2$ solution was kept at a pH of 4 by mixing it with sulphuric acid (H_2SO_4). The WTL fibers were mixed in a solution of 5 wt. % $NaClO_2$ at 60 °C for 1 h with a weight ratio of 1:15 WTL-to-$NaClO_2$ solution. The residue was then filtered from solution using vacuum filtration, thoroughly washed with lukewarm distilled water, and dried in a hot air oven at 60 °C. Next, the composition of cellulose was determined by soaking the holocellulose fraction for 24 h with 6 wt. % of potassium hydroxide solution at room temperature. After that, the sample was filtered, washed with distilled water, and oven-dried at 60 °C for 5 h. The hemicellulose content of the fibers was determined by subtracting the values of cellulose from that of holocellulose. The lignin content was determined by soaking the WTL for 1 h in 72 wt. % H_2SO_4 solution at 30 °C. This method was followed according to Technical Association of the Pulp and Paper Industry (TAPPI) standard method T222. The mixture was diluted to 3% H_2SO_4 and then kept for 2 h in reflux. The solid residue was filtered, washed using lukewarm distilled water, and oven-dried at 60 °C for 7 h. The processes were repeated for the WTLDCF to determine its cellulose, hemicellulose and lignin contents.

1.3.2. Estimation of TRS yield

In order to determine the TRS yield obtained from the PH reaction, a mixture of 0.5 ml of DNS solution, 0.5 ml of the hydrolysate and 0.5 ml of DI water were mixed in a test tube and heated at 100 °C in a conventional water bath heater. After 5 min of heating time, the solution was cooled down to room temperature. Next it was diluted using 4 ml DI water at 1 l dilution factor. This solution was inspected in UV spectrophotometer to obtain an absorbance vs concentration of TRS standard curve of glucose at 540 nm wavelength (Figure 1).

The yield of TRS has been calculated using Equation (1):

$$\text{TRS Yield} = \text{Mtrs} \times \frac{\text{MG} - \text{MW}}{\text{MW} \times 100} \times 100\ \%, \tag{1}$$

where: Mtrs (mg) is the mass and is calculated by Equation (2):

$$\text{Mtrs} = \text{Ctrs} \times \text{V} \times \text{DF}, \tag{2}$$

where: Ctrs = Concentration of the product obtained from the calibration curve (Figure 1) (mg/ml); V = Reaction solution volume (ml); DF = Dilution factor = 11; MG = Molecular weight of glucose monomer = 180; MW = Molecular weight of water = 18.

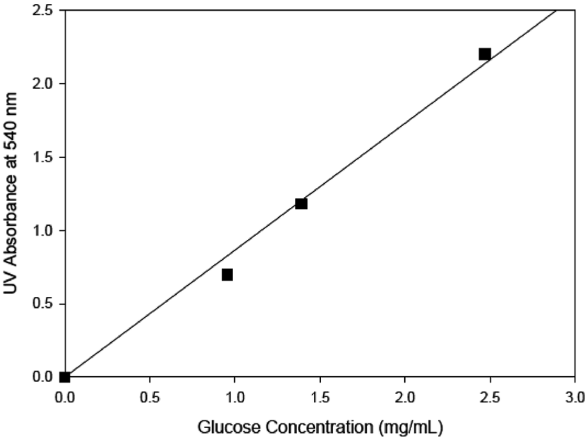

Figure 1. Standard calibration curve for determination of glucose concentration in hydrolysate

1.3.3. Experimental design and optimization

To optimize the hydrolytic degradation reaction parameters, Taguchi orthogonal design array (TODA) has been employed (Radhakumari et al., 2014). The TODA was created in MINITAB software; wherein 4 self-governing parameters (Table 1) were considered that affected the process response (TRS yield). A 3 level 4 parameter experimental design was created which required to take only 9 experimental runs involving specific combinations of the important process parameters to predict the optimal parametric combinations corresponding to maximum TRS yield. Signal to noise ratio was calculated using TRS yield data for each batch experiment (Table 2).

1.4. Life cycle assessment

Life cycle assessment (LCA) study has been carried out according to the ISO 14,040:2006 standards. The study consisted of 4 stages; viz. goal and scope of the process, life cycle inventory (LCI) analysis, impact assessment evaluation and environmental impact assessment interpretation. The environmental impact of any process can be assessed by LCA method. In the present work, the environmental indicators assessed have been: global warming potential (GWP), acidification potential (AP), eutrophication potential (EP), ozone layer depletion potential (ODP) and photochemical ozone creation potential (PCOP). The GWP is the indication of the equivalent amount of CO_2 liberated from the process. It is measured in kg of CO_2 equivalent. Acidification potential is the measure of the acidic effluent emitted from a process. It is measured in kg SO_2 equivalent. EP is the measure of aquatic pollution level by compounds of elements like nitrogen and phosphorus. It is measured using kg phosphate equivalent. ODP measures the amount of harmful pollutant emitted in a process that can cause the depletion of ozone layer. It is measured using kg of CFC11 equivalent. POCP measures amount of emission that can cause the formation of ground level ozone gas. It is measured in kg ethane equivalent. The method adopted in LCA is a "gate to gate" approach. The environmental impact occurring within the boundary of the process has been measured for the PH process.

1.4.1. Goal and scope of the study

Boundary of a process is usually defined by the scope; between the two major types of boundaries, "gate to gate" approach is responsible for counting the impacts occurring within the boundary only. Goal of the study evaluates hotspot, which determines the particular process that is responsible for the highest amount of GWP emissions. Along with it, other environmental impact parameters such as AP, EP, ODP, and POCP have also been evaluated. Present study focused on the three major steps encompassing TRS synthesis; viz. pre-treatment, delignification and PH reaction (Figure 2). The sustainability analysis included environmental impact assessment only. Moreover, the treatment and emission of waste effluent streams were kept out of the scope for this work.

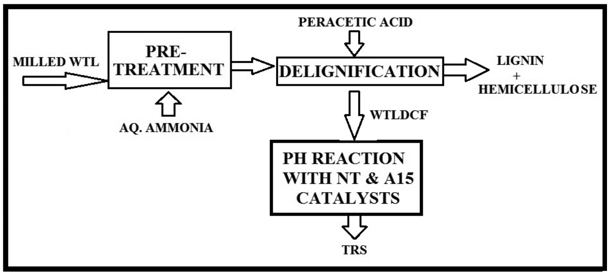

Figure 2. Block diagram of the overall process for environmental impact assessment using "gate to gate" approach

1.4.2. Life cycle inventory (LCI)

LCI is the study associated with the several input and output data analyses of a particular process in terms of mass and energy balances. Primarily Indian standard datasheets have been employed in modeling the present PH process. Furthermore, since the chemicals used in this process have been manufactured by Merck (a German company), remaining inventory datasheets are Germany standard.

1.4.3. Environmental impact assessment

OpenLCA professional software (Ecoinvent CML 2001 impact assessment method) has been employed for LCA. The following five impacts were assessed:

(1) Acidification potential (AP); (2) Eutrophication potential (EP); (3) Global warming potential (GWP); (4) Ozone depletion potential (ODP); (5) Photochemical ozone creation potential (POCP).

2. Results and discussions

2.1. Chemical composition analysis of the WTL and WTLDCF

The chemical composition of WTL and WTLDCF were measured to analyze the holocellulose and lignin contents of the samples. Initial analysis evaluated that the WTL contained 18.7% cellulose, 62.4% hemicellulose and 19.2% lignin. The alkaline treatment of WTL with aqueous NH_4OH (25%) eradicated its surface impurities and majority of the hemicellulose fraction. Furthermore, treatment of PWTL with peracetic acid, enhanced the delignification process. The 94.5% cellulose content in WTLDCF implied that the pre-treatment-delignification process was remarkably effective in increasing the cellulose fraction from 18.7 to 94.5%; while the hemicellulose and lignin contents were only 5.1% and 2.7% respectively. The significant removal of lignin and hemicellulose parts from WTL to produce WTLDCF could be ascribed to the severe deconstruction of WTL cell structure owing to severe stretching-bending vibrations of the WTL/PWTL underlying molecular bonds by virtue of strong penetration of the electromagnetic QHI through the pretreatment-delignification reaction mix.

2.2. TRS yield analysis

From the graph depicted in Figure 1, the relation between UV absorbance at 540 nm and glucose concentration in mg/ml has been evaluated. For an absorbance of 11.6775 nm, the glucose concentration was found to be 13.5 mg/ml furthermore, using Equation (1), the yield percentage of glucose was found to be 66.825%.

2.3. Experimental design and optimum conditions

Photocatalytic hydrolysis (i.e. depolymerization) reaction parameters (Table 1) were optimised in a 4 factor, 3 level TaODA to achieve the maximum yield of TRS. The 4 factors were: $\varnothing_T$ = PH reaction temperature (°C), $\varnothing_t$ = PH reaction time (min), $\varnothing_w$ = WTLDCF to water ratio, $\varnothing_{cc}$ = catalyst concentration (wt. %). Three levels are designated as L_1, L_2, L_3 representing lower, middle and higher levels respectively.

Table 1. Self-governing process parameters for one-pot hydrolysis of WTLDCF

Process variable	$\varnothing_T$ (°C)	$\varnothing_t$ (min)	$\varnothing_w$	$\varnothing_{cc}$ (wt. %)
L_1	60	20	1:20	5
L_2	70	30	1:30	7.5
L_3	80	40	1:40	10

TODA was employed to optimize the process parameters pertaining to the hydrolysis of WTLDCF using A15-NT catalysts. The optimization was performed in MINITAB (v.16) software for windows 10, 64 bits. The optimization was performed in terms of yield of TRS % (Ω_{RS}) obtained from the PH reaction. Using the software, "signal to noise ratio" for each experimental run was evaluated corresponding to the Ω_{RS} (Table 2).

Table 2. Taguchi orthogonal design layout for photocatalytic hydrolysis (PH) reaction

Trial No.	$\varnothing_T$ (°C)	$\varnothing_t$ (min)	$\varnothing_w$	$\varnothing_{cc}$ (wt. %)	Ω_{RS} (%)
1	L1	L1	L1	L1	47.25
2	L1	L2	L2	L2	56.14
3	L1	L3	L2	L3	64.59
4	L2	L1	L3	L2	49.25
5	L2	L2	L1	L3	58.23
6	L2	L3	L2	L1	68.25
7	L3	L1	L2	L3	45.33
8	L3	L2	L3	L1	53.87
9	L3	L3	L1	L2	63.84

2.4. Effects of time and reactor type on TRS yield

To study the effect of batch time on TRS yield, the PH reaction was carried out over a range of time (10–70 min) keeping other parameters fixed at the TODA predicted optimized conditions; i.e. 70 °C, WTLDCF to water ratio of 1:30 (w/w) and 5 wt. % A15-NT catalyst concentration. The TRS yield vs time plot has been exhibited in Figure 3; which evinced that after 40 min of PH reaction time, max 68.25% TRS yield could be achieved in QHIBR. This could be attributed to the fact that beyond 40 min, consecutive reactions led to TRS degradation that eventually reduced net TRS yield beyond the threshold PH time. Notably, the CTSABR provided comparatively much lower (~51.50%) TRS yield within the same 40 min duration; this clearly manifested the supremacy of QHR over conventional heating method (employed in CTSABR) in terms of faster hydrolysis of cellulose into TRS. The intensified hydrolysis achieved in QHIBR could be principally attributed to the intense stretching and bending molecular shakings created by QHI that resulted in augmented molecular colissions.

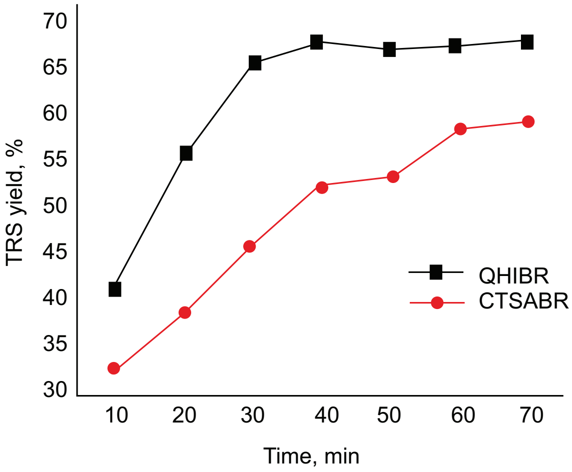

Figure 3. Effect of hydrolysis time on TRS yield in CTSABR and QHIBR

2.5. Energy-efficiency and catalytic-efficacy

In order to compare the energy-efficiency of the QHIBR with CTSAR, the hydrolysis experiments were carried out at the TODA predicted optimal condition. It was found that, in order to provide 68.25% maximum yield of TRS in 40 min hydrolysis time (Figure 3), QHIBR consumed 348 kJ of energy; whereas, the CTSABR could render only 51% TRS yield within the same time at the expense of much higher (1200 kJ) energy. From Figure 3, it could be further confirmed that the QHIBR would always render lower TRS yield over the entire range of time considered in the present study. Notably, QHI provided faster and more uniform heating and created intense molecular stretching and bending vibrations; thereby promoting rapid hydrolysis of WTLDCF towards augmented TRS yield. Besides, the use of NT catalyst could trigger photocatalytic actions under the electromagnetic radiation (i.e. QHI); however, the photocatalytic properties of NT couldn't be activated while using the CTS. Evidently, the combined effects of NT and QHI resulted in much superior TRS yield in QHIBR. Furthermore, it could be inferred that within the required 40 min hydrolysis time (corresponding to maximum TRS yield in QHIBR), the CTSABR consumed 244.8% more energy compared to the highly-efficient QHIBR.

The TRS yield could not reach 68.25% even after 70 min in CTSABR unlike the QHIBR (Figure 3). Notably, in both the reactors, A15 catalyst could provide the necessary Brønsted acidity for the hydrolysis of WTLDCF. However, in case of CTSABR no photon energy could be supplied to the hydrolysis mix, hence, the photocatalytic efficacy of the NT catalyst couldn't be triggered; therefore, rendering slower hydrolysis rate in comparison with QHIBR (which is provided with photon source QIR). Evidently, in QHIBR, both Brønsted acidity of A15 catalyst and photocatalytic activities of NT catalyst could be exploited, while in case of CTSABR only the Brønsted acidity could be availed.

2.6. LCA interpretation

Table 3 presents the LCA outcomes deploying the "gate to gate" approach. From the data it could be concluded that the delignification step was responsible for the highest global warming potential of 1.425 kg CO_2 equivalent. The overall process contributed a total of 2.215 kg of CO_2 equivalent which was permissible. In terms of acidification potential, pretreatment and PH reaction steps contributed the major values owing to the presence of nitrogen and sulphonate groups.

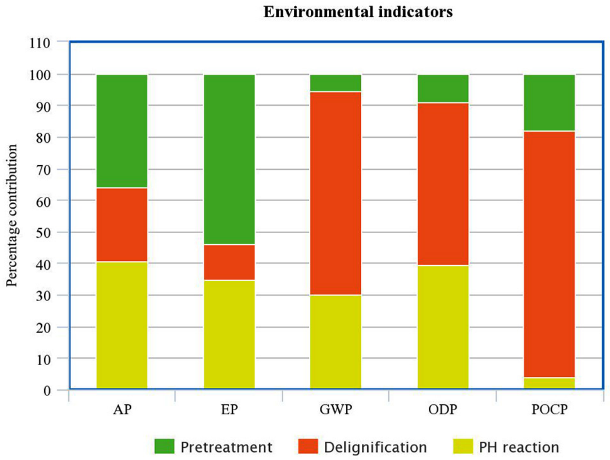

Figure 4. Percentage contributions of pretreatment, delignification and photocatalytic hydrolysis towards AP, EP, GWP, ODP and POCP

Figure 4 depicts a graphical representation of percentage contribution of each process, viz. pretreatment, delignification and photocatalytic hydrolysis towards AP, EP, GWP, ODP and POCP. The results indicate that all the five environmental impacts were within acceptable limit (Arctic Monitoring and Assessment Programme, 2004) to ascertain an ecofriendly process.

Conclusions

The extraction of cellulose fibers from WTL and its subsequent hydrolysis into TRS using energy-efficient QHIBR has been found promising. The delignification process ensured the appreciable removal of lignin fraction. The combined acidic-photocatalytic hydrolysis process was found more energy-efficient (244.8% lower energy-consumption) in presence of non-ionizing quartz-halogen radiation in comparison with conventional thermal energy. The four independent process parameters viz., reaction time, reaction temperature, biomass to water ratio and catalyst concentration could be optimized by Taguchi orthogonal design array to render maximum TRS yield. Significantly, only 40 min was required to accomplish maximum TRS yield (68.25%) at optimized conditions in the QHIBR which was much higher compared to that (51%) obtained through CTSABR; exhibiting a faster and more proficient

Table 3. LCA outcomes for the overall process

	AP (kg SO_2 equi.)	EP (kg phos-phate equi.)	GWP (kg CO_2 equi.)	ODP (kg R11 equi.)	POCP (kg ethane equi.)
Pre-treatment	4.09	0.532	0.123	5.76E-14	0.0303
Delignification	2.651	0.113	1.425	3.23E-13	0.129
PH reaction	4.597	0.343	0.667	2.48E-13	0.0068
Total	11.338	0.988	2.215	6.286E-13	0.1661

process by QHIBR. The developed energy-efficient process for depolymerization of WTL towards synthesis of valuable platform chemicals, viz. WTLDCF and TRS is expected to be attractive for many potential biorefinery applications. The LCA of the production process has suggested that the overall process emission is at permissible measure based on product mass and quality. Thus, the developed overall waste tea leaves conversion process is energy-efficient and environmentally sustainable.

Acknowledgements

This work was supported by DHESTBT (ST/P/S&T/4G-2/2018); Government of West Bengal, India) and RUSA 2.0 (Ref. No. R-11/316/19) Jadavpur University, India.

Disclosure statement

The authors declare that they have no competing interests.

Authors' contributions

Oindrila Roy conducted experiment, analyzed and interpreted the data and prepared the draft manuscript under the guidance of Rajat Chakraborty. Besides, Rajat Chakraborty designed and developed the reactor, conceptualised the entire research and provided overall guidance in writing and editing the manuscript. Rajat Chakraborty solely revised the final manuscript.

References

Amarasinghe, B., & Williams, R. (2007). Tea waste as a low cost adsorbent for the removal of Cu and Pb from wastewater. *Chemical Engineering Journal*, *132*, 299–309. https://doi.org/10.1016/j.cej.2007.01.016

Arctic Monitoring and Assessment Programme. (2004). *Persistent toxic substances, food security and indigenous peoples of the Russian North* (Final Report). Oslo.

Bharti, R., & Singh, B. (2020). Green tea (*Camellia assamica*) extract as an antioxidant additive to enhance the oxidation stability of biodiesel synthesized from waste cooking oil. *Fuel*, *262*, 116658. https://doi.org/10.1016/j.fuel.2019.116658

Bian, J., Peng, P., Peng, F., Xiao, X., Xu, F., & Sun, R.-C. (2014). Microwave-assisted acid hydrolysis to produce xylooligosaccharides from sugarcane bagasse hemicelluloses. *Food Chemistry*, *156*, 7–13. https://doi.org/10.1016/j.foodchem.2014.01.112

Borrion, A., McManus, M., & Hammond, G. (2012). Environmental life cycle assessment of lignocellulosic conversion to ethanol: A review. *Renewable and Sustainable Energy Reviews*, *16*, 4638–4650. https://doi.org/10.1016/j.rser.2012.04.016

Cai, H., Li, C., Wang, A., Xu, G., & Zhang, T. (2012). Zeolite-promoted hydrolysis of cellulose in ionic liquid, insight into the mutual behavior of zeolite, cellulose and ionic liquid. *Applied Catalysis B: Environmental*, *123–124*, 333–338. https://doi.org/10.1016/j.apcatb.2012.04.041

Chakraborty, R., & Mandal, E. (2015). Fast and energy efficient glycerol esterification with lauric acid by near and far-infrared irradiation: Taguchi optimization and kinetics evaluation. *Journal of the Taiwan Institute of Chemical Engineers*, *50*, 93–99. https://doi.org/10.1016/j.jtice.2014.12.024

Chheda, J. N., Huber, G. W., & Dumesic, J. A. (2007). Liquid-phase catalytic processing of biomass-derived oxygenated hydrocarbons to fuels and chemicals. *Angewandte Chemie International Edition*, *46*(38), 7164–7183. https://doi.org/10.1002/anie.200604274

Engin, A., Özdemir, Ö., Turan, M., & Turan, A. (2008). Color removal from textile dyebath effluents in a zeolite fixed bed reactor: Determination of optimum process conditions using Taguchi method. *Journal of Hazardous Materials*, *159*, 348–353. https://doi.org/10.1016/j.jhazmat.2008.02.065

Fan, J., De bruyn, M., Zhu, Z., Budarin, V., Gronnow, M., Gomez, L., Macquarrie, D., & Clark, J. (2013). Microwave-enhanced formation of glucose from cellulosic waste. *Chemical Engineering and Processing: Process Intensification*, *71*, 37–42. https://doi.org/10.1016/j.cep.2013.01.004

Farhoosh, R., Golmovahhed, G., & Khodaparast, M. (2007). Antioxidant activity of various extracts of old tea leaves and black tea wastes (Camellia sinensis L.). *Food Chemistry*, *100*, 231–236. https://doi.org/10.1016/j.foodchem.2005.09.046

Fatehi, P. (2013). Recent advancements in various steps of ethanol, butanol, and isobutanol productions from woody materials. *Biotechnology Progress*, *29*, 297–310. https://doi.org/10.1002/btpr.1688

Finkbeiner, M., Inaba, A., Tan, R., Christiansen, K., & Klüppel, H. (2006). The new international standards for life cycle assessment: ISO 14040 and ISO 14044. *The International Journal of Life Cycle Assessment*, *11*(2), 80–85. https://doi.org/10.1065/lca2006.02.002

Goswami, M., Meena, S., Navatha, S., Prasanna Rani, K., Pandey, A., Sukumaran, R., Prasad, R., & Prabhavathi Devi, B. (2015). Hydrolysis of biomass using a reusable solid carbon acid catalyst and fermentation of the catalytic hydrolysate to ethanol. *Bioresource Technology*, *188*, 99–102. https://doi.org/10.1016/j.biortech.2015.03.012

Guo, F., Fang, Z., Xu, C., & Smith, R. (2012b). Solid acid mediated hydrolysis of biomass for producing biofuels. *Progress in Energy and Combustion Science*, *38*, 672–690. https://doi.org/10.1016/j.pecs.2012.04.001

Guo, H., Qi, X., Li, L., & Smith, R. (2012a). Hydrolysis of cellulose over functionalized glucose-derived carbon catalyst in ionic liquid. *Bioresource Technology*, *116*, 355–359. https://doi.org/10.1016/j.biortech.2012.03.098

Hameed, B. (2009). Spent tea leaves: A new non-conventional and low-cost adsorbent for removal of basic dye from aqueous solutions. *Journal of Hazardous Materials*, *161*, 753–759. https://doi.org/10.1016/j.jhazmat.2008.04.019

Harris, E., & Kline, A. (1949). Hydrolysis of wood cellulose with hydrochloric acid and sulfur dioxide and the decomposition of its hydrolytic products. *The Journal of Physical and Colloid Chemistry*, *53*, 344–351. https://doi.org/10.1021/j150468a003

Hassan, E., Mutelet, F., Moïse, J., & Brosse, N. (2015). Pretreatment of miscanthus using 1,3-dimethyl-imidazolium methyl phosphonate (DMIMMPh) ionic liquid for glucose recovery and ethanol production. *RSC Advances*, *5*, 61455–61464. https://doi.org/10.1039/C5RA08946H

Inal, I., Holmes, S., Banford, A., & Aktas, Z. (2015). The performance of supercapacitor electrodes developed from chemically activated carbon produced from waste tea. *Applied Surface Science*, *357*, 696–703. https://doi.org/10.1016/j.apsusc.2015.09.067

Lin, Y., Wang, A., Wang, D., & Chen, C. (2009). Machining performance and optimizing machining parameters of Al_2O_3–TiC ceramics using EDM based on the Taguchi method. *Materials and Manufacturing Processes*, *24*, 667–674. https://doi.org/10.1080/10426910902769285

Liu, B., Zhang, Z., & Zhao, Z. (2013). Microwave-assisted catalytic conversion of cellulose into 5-hydroxymethylfurfural in ionic liquids. *Chemical Engineering Journal*, *215–216*, 517–521. https://doi.org/10.1016/j.cej.2012.11.019

Liu, M., Jia, S., Gong, Y., Song, C., & Guo, X. (2013). Effective hydrolysis of cellulose into glucose over sulfonated sugar-derived carbon in an ionic liquid. *Industrial & Engineering Chemistry Research*, *52*, 8167–8173. https://doi.org/10.1021/ie400571e

Manzanares, P. (2020). The role of biorefinering research in the development of a modern bioeconomy. *Acta Innovations*, *37*(47), 47–56. https://doi.org/10.32933/ActaInnovations.37.4

Menon, V., & Rao, M. (2012). Trends in bioconversion of lignocellulose: Biofuels, platform chemicals & biorefinery concept. *Progress in Energy and Combustion Science*, *38*(4), 522–550. https://doi.org/10.1016/j.pecs.2012.02.002

Miller, G. (1959). Use of dinitrosalicylic acid reagent for determination of reducing sugar. *Analytical Chemistry*, *31*, 426–428. https://doi.org/10.1021/ac60147a030

Peng, C., Yan, X., Wang, R., Lang, J., Ou, Y., & Xue, Q. (2013). Promising activated carbons derived from waste tea-leaves and their application in high performance supercapacitors electrodes. *Electrochimica Acta*, *87*, 401–408. https://doi.org/10.1016/j.electacta.2012.09.082

Pradhan, P., Chakraborty, S., & Chakraborty, R. (2016). Optimization of infrared radiated fast and energy-efficient biodiesel production from waste mustard oil catalyzed by Amberlyst 15: Engine performance and emission quality assessments. *Fuel*, *173*, 60–68. https://doi.org/10.1016/j.fuel.2016.01.038

Qian, X., Lei, J., & Wickramasinghe, S. (2013). Novel polymeric solid acid catalysts for cellulose hydrolysis. *RSC Advances*, *3*, 24280. https://doi.org/10.1039/c3ra43987a

Radhakumari, M., Ball, A., Bhargava, S., & Satyavathi, B. (2014). Optimization of glucose formation in karanja biomass hydrolysis using Taguchi robust method. *Bioresource Technology*, *166*, 534–540. https://doi.org/10.1016/j.biortech.2014.05.065

Shrotri, A., Kobayashi, H., & Fukuoka, A. (2018). Cellulose depolymerization over heterogeneous catalysts. *Accounts of Chemical Research*, *51*, 761–768. https://doi.org/10.1021/acs.accounts.7b00614

Silveira, M. H. L., Morais, A. R. C., Lopes, A. M. D. C., Olekszyszen, D. N., Bogel-Łukasik, R., Andreaus, J., & Ramos, L. P. (2015). Current pretreatment technologies for the development of cellulosic ethanol and biorefineries. *ChemSusChem*, *8*, 3366–3390. https://doi.org/10.1002/cssc.201500282

Sun, B., Duan, L., Peng, G., Li, X., & Xu, A. (2015). Efficient production of glucose by microwave-assisted acid hydrolysis of cellulose hydrogel. *Bioresource Technology*, *192*, 253–256. https://doi.org/10.1016/j.biortech.2015.05.045

Tsubaki, S., Oono, K., Onda, A., Yanagisawa, K., Mitani, T., & Azuma, J. (2016). Effects of ionic conduction on hydrothermal hydrolysis of corn starch and crystalline cellulose induced by microwave irradiation. *Carbohydrate Polymers*, *137*, 594–599. https://doi.org/10.1016/j.carbpol.2015.11.022

Utomo, H., & Hunter, K. (2006). Adsorption of divalent copper, zinc, cadmium and lead ions from aqueous solution by waste tea and coffee adsorbents. *Environmental Technology*, *27*, 25–32. https://doi.org/10.1080/09593332708618619

Verardi, A., Lopresto, C., Blasi, A., Chakraborty, S., & Calabrò, V. (2020). Bioconversion of lignocellulosic biomass to bioethanol and biobutanol. In *Lignocellulosic biomass to liquid biofuels* (pp. 67–125). Elsevier. https://doi.org/10.1016/B978-0-12-815936-1.00003-4

Yan, K., Jarvis, C., Gu, J., & Yan, Y. (2015). Production and catalytic transformation of levulinic acid: A platform for speciality chemicals and fuels. *Renewable and Sustainable Energy Reviews*, *51*, 986–997. https://doi.org/10.1016/j.rser.2015.07.021

Zandi, P., & Gordon, M. (1999). Antioxidant activity of extracts from old tea leaves. *Food Chemistry*, *64*, 285–288. https://doi.org/10.1016/S0308-8146(98)00047-8

Zhang, Z., & Zhao, Z. (2009). Solid acid and microwave-assisted hydrolysis of cellulose in ionic liquid. *Carbohydrate Research*, *344*, 2069–2072. https://doi.org/10.1016/j.carres.2009.07.011

Zhao, H., Kwak, J., Wang, Y., Franz, J., White, J., & Holladay, J. (2006). Effects of crystallinity on dilute acid hydrolysis of cellulose by cellulose ball-milling study. *Energy & Fuels*, *20*, 807–811. https://doi.org/10.1021/ef050319a

Zuorro, A., Santarelli, M., & Lavecchia, R. (2013). Tea waste: A new adsorbent for the removal of reactive dyes from textile wastewater. *Advanced Materials Research*, *803*, 26–29. https://doi.org/10.4028/www.scientific.net/AMR.803.26