Process simulation and life cycle assessment of converting autoclaved municipal solid waste into butanol and ethanol as transport fuels

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15 **Abstract**

16 In 2015/2016, the total municipal solid waste (MSW) collected by local authority in the U.K.
17 was 26 million tonnes and over 57% is still put into landfill or incinerated. MSW is a promising
18 feedstock for bio-butanol production as it has a high lignocellulosic fibre content such as paper,
19 wood, and food waste, about 50 wt% of a typical MSW stream. The study evaluates acetone,
20 butanol, ethanol and hydrogen production from autoclaved municipal solid waste feedstock.
21 Life cycle assessment is undertaken to evaluate the acetone, butanol, ethanol and hydrogen
22 production process, considering cogeneration of heat and power from residual biogenic waste
23 based on experimental data and process modelling. Acetone, butanol, and ethanol product yield
24 can be achieved at 12.2 kg butanol, 1.5 kg ethanol, 5.7 kg acetone, and 0.9 kg hydrogen per
25 tonne MSW. The product yield is relatively low compared to other lignocellulosic feedstocks
26 primarily because of the lower hydrolysis yield (38% for glucose) achieved in this study;
27 however, hydrolysis yields could be improved in future optimisation work. The conversion
28 shows a net primary energy demand of -1.11 MJ/MJ liquid biofuels (butanol and ethanol) and
29 net greenhouse gas emission of -12.57 g CO₂eq/MJ liquid biofuels, achieving a greenhouse gas
30 reduction of 115% compared to gasoline comparator.

31 **Keywords**

32 Municipal solid waste, Waste autoclaving, Enzymatic hydrolysis, ABE fermentation, Life
33 cycle assessment

1 Introduction

The EU transport sector accounted for 25.8% of the total greenhouse gas (GHG) emissions in the EU in 2015 at 1.05 Gt/yr including international aviation and maritime emissions (European Environment Agency, 2017). The EU's climate change targets have already stated transport emissions must be cut by 60% by 2050 compared with 1990 levels (The European Union, 2014). Renewable energy sources can contribute to climate change mitigation through the reduction of GHG emissions and achieve sustainable development, driving the increased demand for renewable fuels. The EU issued the Renewable Energy Directive (RED), for instance, requires that renewable energy content should account for at least 10% of the energy used in transportation by 2020 (European Commission, 2018).

Butanol (C_4H_9OH) is an attractive renewable liquid transport fuel. Its superior properties have been well documented: butanol fits the existing fuel infrastructure as it can be stored in a mixture with traditional gasoline and diesel at a varied ratio; it has a better energy density (30% greater than ethanol) and combustion performance due to similar air: fuel ratio to that of gasoline than ethanol and can be used in higher quantities in a standard petrol engine (up to 20% without any engine modifications) but ethanol is limited to 15% (Wu et al., 2007); and exhibits low solubility in water which reduces the risk of groundwater contamination from unintended release. Butanol thus has the potential to substitute both ethanol and biodiesel in the biofuel market to reach \$185.3 billion in 2021 as estimated by Pike Research (Microbiology Society, 2013). In addition to being a potential biofuel, butanol is also a valuable C4 feedstock for chemical synthesis (e.g., methacrylate esters, butyl glycol ethers, butyl acetates, and plasticizers) and an industrial solvent or co-solvent for surface coatings (Bankar et al., 2013). However, challenges still need to be addressed, such as high feedstock costs and associated high operational cost requiring cheaper and sustainable feedstocks.

Municipal solid waste (MSW) is a promising feedstock for butanol production as it has a high lignocellulosic fibre content such as paper, wood, and food waste, about 50 wt% of a typical MSW stream. Unlike other cellulosic feedstocks, MSW has low/negative feedstock prices, immediate availability, and high potential in reducing GHG emissions of biofuel production compared to first and second generation biofuels from food crop and agricultural or forest residue wastes. Further, diverting the organic content of MSW for biofuel production is also beneficial in addressing MSW waste management issues (Gharfalkar et al., 2015; Jeswani and Azapagic, 2016). In 2015/2016, the total MSW collected by local authority in the U.K. was over 26 million tonnes and over 57% is still put into landfill or incinerated (Department for Environmental Food and Rural Affairs (Defra), 2017b; Science and Technology Select Committee, 2014). The gate fee charged by landfilling in the UK reveal a cost of £19/tonne excluding landfill tax and £102/tonne including landfill tax, while tipping fees for incineration are £86/tonne in 2015/2016 (WRAP (Waste & Resources Action Programme), 2016). Therefore, the production of biofuel from MSW is extremely attractive in terms of environmental and economical perspective.

Life cycle assessment (LCA) provides a transparent methodology that can be used to examine lignocellulosic biofuel production as it examines the environmental burdens over the entire life, from production, through use and on to disposal or recycling (McKechnie et al., 2011). Existing LCAs of biofuel production (e.g., ethanol) from various feedstocks including corn stover, wheat straw, poplar, eucalyptus and waste papers amongst others have been widely reported in the literatures (Borrion et al., 2012; Michael et al., 2012; Mohammad et al., 2013; Zhang et al., 2010b). Few studies by Chester & Martin (Chester and Martin, 2009), Schmitt et al. (Schmitt et al., 2012) and Kalogo et al. (Kalogo et al., 2007) evaluated ethanol production from MSW via dilute acid pretreatment and hydrolysis and presented the life cycle

environmental impacts of the conversion. It showed MSW derived ethanol can reduce energy use and GHG emissions compared to gasoline and other cellulosic ethanol production. But environmental impacts are highly dependent on conversion technology, process conditions used, and waste classification and its overall impact of MSW to ethanol is limited due to availability of MSW.

Autoclaving is a new pretreatment technology replacing conventional dilute acid pretreatment to process unsorted MSW. Prior LCA study considered autoclaving unsorted MSW with subsequent composting in tunnels/in confined windrow/ in turning windrow, or anaerobic digestion of biogenic fibres (Quirós et al., 2015). The results showed that autoclaving with sorting, digesting anaerobically and composting had the lowest environmental impact values for eutrophication and global warming potential. However, there is no consideration of higher value uses for this biogenic fibre materials, such as liquid biofuels (e.g., butanol).

To address the gaps in technical and environmental aspects of acetone, butanol, ethanol (ABE) production from pretreated MSW, in this study, we develop process models for ABE production from autoclaved MSW feedstock. The overall technical performance and environmental impacts (i.e., primary energy demand and global warming potential) are evaluated across the integrated processes, including autoclave, hydrolysis, fermentation, and distillation.

2 Method

The study models in detail the production of ABE and hydrogen from autoclaved municipal solid waste feedstock. The overall process design converts MSW into ABE and hydrogen by autoclave, hydrolysis, fermentation and distillation. Product recovery, energy recovery, wastewater treatment and utilities are also included in the design. The recovery of non-biogenic content of MSW after autoclave, including plastics, metal, and glass materials, are excluded in

this study. The input and output mass and energy flow of the system are extracted from experimental data and process modelling as discussed below. Inventory data is also supplemented by Ecoinvent database (Wernet et al., 2016) and literature data where available, e.g., enzyme production for hydrolysis (Nielsen et al., 2006) (see Table 1). The LCA is undertaken in GaBi 8.2 (2017) using Ecoinvent 3.3 inventory databases. Two environmental impacts are quantified: primary energy demand (PED) in terms of MJ and global warming potential (GWP), based on the most recent IPCC 100-year GWP factors to quantify GWP in terms of CO₂ equivalents (CO₂ eq.) (Stocker et al., 2013). Direct CO₂ emissions from the fermentation and combustion of butanol and ethanol fuel are excluded as the emitted carbon from renewable biomass resources can be counted as “carbon neutral”. Avoided emissions from diverting waste to butanol and ethanol production from other conventional waste treatment routes (e.g., landfill, incineration) are also excluded.

2.1 Scope and functional unit

We develop an LCA model of MSW-ABE following the ISO Standards 14040 and 14044 (International Organization for Standardization, 2006a, b). The functional unit is defined as one MJ of liquid biofuels (butanol and ethanol)) used as a reference to quantify all inputs and outputs of the process steps. A schematic process flow diagram defining the system boundaries is shown in Fig. 1. The system boundary begins with the sorting and separation of MSW (energy use and environmental burdens of the processes and products generating MSW are excluded from the study) and ends with the combustion of the fuel in a light duty vehicle. System expansion method is used to consider the co-products’ benefits where liquid biofuels (butanol and ethanol) are the main product and acetone, hydrogen, and excess electricity are considered as co-products thereby allocating their impacts to main product liquid biofuels.

2.2 Waste composition

The waste composition used is representative of the UK MSW with the following wet composition by mass: paper and cardboard (22%), food waste (17%), wood (8.7%), plastic (22%), glass (1%), garden waste (3%), metals (4%), textiles (6.6%) and others (15.7%) (Department for Environmental Food and Rural Affairs (Defra), 2017a).

2.3 Life Cycle inventory

2.3.1 Autoclave Pretreatment

The autoclave system is a mechanical heat treatment process developed by Wilson Bio-Chemical (Wilson Bio-Chemical, 2017), based on existing autoclaving and steam boiler technologies. The autoclave is a front-loading and rotating pressure vessel in which the MSW is treated with high pressure saturated steam in a batch process. The organic fraction is broken down to a cellulose-rich fibre, which has a high sugar composition (40–50%) and thus could be suitable for biofuel production via fermentation. Untreated wastes such as plastic, glass, textiles and metals are sterilised and able to be recovered post-autoclave.

Based on the best performance running of the autoclave process, processing parameters of 160°C for two hours for each batch have been determined. Thus the energy requirement of 43 MJ electricity and 274 MJ natural gas and 245 L water is determined from plant operation to pretreat one tonne of MSW (Wilson Bio-Chemical, 2017).

2.3.2 Hydrolysis

The waste stream has been subjected to high temperature processing and this is similar to the pretreatment normally required to overcome the inherent recalcitrance of biomass feedstocks to subsequent enzymatic saccharification to sugars. Preliminary studies have previously demonstrated that between 30-40% of the fibre sugar can be released by subsequent enzyme hydrolysis (Ibbett, 2018). A commercially available enzyme cocktail - Novozymes Cellic

CTec2 (Novozymes, 2018) - is used in this study. Samples of the MSW fibre are milled to a consistent particle size (0.5mm) and then loaded into the hydrolysis vessel where it is diluted with water from the mains water tank into a dilute slurry (20% solids assumed in this study but a range of 20–30% solids are evaluated in the sensitivity analysis). At this point, adjustments may also be made to process conditions such as pH. The slurry is then dosed with an enzyme solution (5% wt/wt) (15–60 filter paper units (FPU) per gram cellulase) which is stored in its own separate storage tank, causing hydrolysis of the biogenic fibre to a solution of fermentable sugars. There will also be solids remaining in suspension which are either un-hydrolysable or have not been fully hydrolysed. All of this mixture is transferred downstream to the filtration unit. The efficiency of hydrolysis is assessed by monitoring glucose release into the media, over a period of 48h at the temperature of 50°C, by high pressure ion chromatography.

The composition of biogenic fibre transported from the Wilson System as stated above can be evaluated by analysis of the monosaccharides liberated by total acid hydrolysis of the polysaccharides present in the fibre cell-wall matrix (Ibbett et al., 2011). This method gave a monosaccharide content as follows: glucose (40-45%), xylose (4-5%), galactose (0.7%) and arabinose (2.9%), as shown in Fig. 2, where glucose is mainly derived from cellulose and the other three monosaccharides are derived from hemicellulose.

The net enthalpy change for each reaction (see the reactions outlined below) is calculated using the heat of formation (ΔH_f) (Humbird et al., 2011), see eq (1) as below.

$$Q_{reaction} = \sum \frac{\Delta H_f \times (m_b cy)}{M_w} \quad (1)$$

where ΔH_f is the heat of formation (kJ/mol), m_b is the number of moles of sugar formed from 1 kg MSW fibre (mol), c is the sugar composition (%), y is sugar hydrolysis conversion yield (%), M_w is the molecular weight of the sugar (kg/mol).

Mixing is achieved by an agitator and heat control is achieved by a centrifugal pump which pumps the hydrolysate around a loop through a heat exchanger cooled with cooling water, as is assumed in previous biorefinery models (Humbird et al., 2011).

2.3.3 Fermentation

After hydrolysis, the sugary solution is adjusted (as required) with antifoam, pH altering agents and other required additions (such as nutrients) in preparation for fermentation. The microorganism is *Clostridium acetobutylicum* ATCC 824 and the medium of all fermentations contain (per L) KH_2PO_4 1 g, K_2HPO_4 0.76 g, $\text{CH}_3\text{COONH}_4$ 2.9 g (of which CH_3COO^- is 2.2 g), yeast extract (Duchefa) 2.5 g, $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ 6.6 mg, $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$ 1 g, and *p*-aminobenzoic acid (*p*-ABA) 0.1 g. Following this, the vessel is inoculated with a previously prepared *Clostridium* culture from the seed culture vessel. The fermentation is conducted at 37°C over a duration of 48 hours per batch. We assume that the inventory data of producing *Clostridium acetobutylicum* would be similar to that of producing *Z. mobilis* as in the design (Dunn et al., 2012; Humbird et al., 2011). Inventory data of nutrients are obtained from publicly available data (Adom and Dunn, 2015; Edwards, 2016).

Fermentation then proceeds, producing ABE as well as hydrogen and CO_2 . As the butanol concentration builds, it inhibits the growth of the *Clostridium*. Therefore, nitrogen gas stripping is used to selectively remove the ABE which escapes as a vapour to the condenser along with CO_2 , hydrogen, nitrogen and some water vapour. The hydrogen and CO_2 are collected during the fermentation and purified by Pressure swing adsorption in the subsequent distillation stage. Totally, ABE fermentation yield can be achieved at 33% of which has been validated

experimentally by a 30% yield and hydrogen yield is 1.6% based on molar ratio shown in Table

2. The relative ratio of produced solvents acetone, butanol and ethanol is 28%, 62% and 10%.

The reactions represent a qualitative measure rather than stoichiometric quantitative relationships for the ABE conversion process. Based on eq (1) above, reaction energy shows the conversion is mildly exothermic; as such, we assume fermentation unit does not require thermal management (i.e., heat loss to surroundings is balanced by heat release from fermentation).

2.3.4 Distillation

The acetone, butanol, ethanol and water vapours are condensed in the beer column and are subsequently transferred to rectification column for separation. The un-condensable hydrogen, nitrogen and CO₂ continue through the condenser to pressure swing adsorption. Pressure swing adsorption technology (HyGear, 2017) is used to purify hydrogen from the gaseous mixture leaving the condenser. This hydrogen can be pressurised and stored in a tank, whilst the remaining N₂ and CO₂ are discarded. The acetone, butanol, ethanol and water are distilled based upon their relative volatilities into pure streams of each component by a gas stripping procedure. This concentrated solution of solvent will then be distilled to obtain the three distinct compounds: butanol, acetone and ethanol to meet the requested biofuel specifications. Recirculation of the stripping gas will reduce the costs. Firstly, acetone is assumed to be extracted in the first distillation column with a purity of 99 wt%. After separation of acetone, the remaining liquid goes through a decanter before going to the second distillation column to separate ethanol which is then dehydrated through the molecular sieve adsorption to a purity of 99.5 wt%. The bottom stream which is mainly butanol and water after separation of ethanol goes to the third distillation for butanol separation (99.7 wt%) (Baral and Shah, 2016; Grisales Díaz and Olivar Tost, 2017). These pure products are finally stored in tanks, ready for

distribution or analysis. The waste biofuels and water mixture are assumed to be discharged to the lignin separator and wastewater treatment system mixed with stillage from hydrolysis and fermentation units.

2.3.5 Wastewater Treatment

Wastewater treatment process in previous model (Humbird et al., 2011) is adapted in this study. After hydrolysis, fermentation and distillation, the stillage contains unfermented sugars, organic acids and solid residuals, e.g., lignin. They are sent to a press filter to separate solids (mainly lignin) to the combustor while the liquid fraction is sent to wastewater treatment . Wastewater treatment consists of anaerobic and aerobic digestion treating and recycling the wastewater to minimise the amount of water discharged and fresh water requirement. The total chemical oxygen demand is assumed to be proportional to the solid content concentration in the wastewater and is calculated to be 245 g/L. In anaerobic digestion, 91% of organic content is converted into biogas (86%) and microorganism cell mass (5%). The biogas from the digester has a composition of 51% CH₄/49% CO₂ on a dry molar basis. Methane is produced on the basis of the organic content at a yield of 228 g biogas/kg chemical oxygen demand while the cell mass is produced at 45 g cell mass/kg chemical oxygen demand (Humbird et al., 2011).

The liquid after anaerobic digestion is further treated in aerobic digestion where 96% of the remaining soluble organic content is digested, with 76% converting into water and carbon dioxide and 22% producing cell mass. Chemicals such as caustic soda must be added to adjust the pH for digestion. After this step, the liquid is sent to a membrane bioreactor clarification system where the aerobic biomass sludge is separated to sludge centrifuge mixed with anaerobic sludge for dewatering. The centrifuge solid is dewatered and sent to the combustor for energy recovery and the remaining water is recycled to aerobic digester for additional treatment. The clarified water is pumped to the reverse osmosis for salt removal. About 79%

water is assumed to pure and recycled to the process and 21% of the water rejected from reverse osmosis is further concentrated in an evaporator system. In the evaporator, about 7% dry brine content (mainly sodium nitrate) is sent for disposal while the concentrated water is assumed to be clean and recycled to the process.

2.3.6 Energy recovery

The lignin from hydrolysis and fermentation residue and biogas and sludge produced from wastewater treatment are fed to the combustor for heat and power generation. A grate incinerator was modelled with electricity and heat production efficiency of 18.7% and 21.6% of the waste's lower heating value, respectively (Veolia, 2012). The cogenerated heat is assumed to be used to meet process heat requirements; the excess could be used for sterilization or for cooling generation via absorption refrigeration plant on site, or exported via a heat network, but such uses are not considered in the present study and excess heat is assumed to not have a practical use. The electricity is used to supply the process and any surplus electricity is assumed to be exported to the grid. The electricity system is assumed to be representative of the UK average mix in 2015, composed of nuclear (20.9%), coal (22.5%), natural gas (29.7%), hydro (1.9%), heavy fuel oil (0.6%), wind (14.2%), other renewables (8.7%), and other (1.4%) (Digest of UK Energy Statistics (DUKES), 2017).

2.4 Sensitivity analysis

2.4.1 Sensitivity analysis of background systems

Relative environmental performance of MSW derived liquid biofuels depends on reference fuels system, enzyme production and which electricity types (i.e., renewable content of the electricity) the exported electricity is displacing. It arises from regional variability of electricity generation sources and associated impacts. Thus we conducted sensitivity analyses to study the influence of background systems on life cycle emissions of MSW derived liquid biofuels.

2.4.2 Sensitivity analysis of process variables

The overall yield of ABE from MSW depends on the biogenic fibre content of MSW, the conversion efficiencies from cellulose to glucose and xylose during hydrolysis, glucose and xylose yield and glucose and xylose conversion during fermentation. The ultimate ABE concentration is also dependant on the solid concentration during hydrolysis. Distributions are fitted where sufficient data are available (e.g., hydrolysis yields) or assigned based on minimum/maximum values to model parameters. Monte Carlo simulations (Ruth and Jechura, 2003; Tu and McDonnell, 2016) enable an investigation into how input uncertainty propagates through the mass and energy balance model and LCA model. These distributions and underlying data sources are summarised in Table 4 for processing parameters including hydrolysis yield and solid content. Triangular distributions are used for these parameters where the peak values – indicating highest probability - are those from experiments, with the probability decreasing linearly until reaching zero at the upper and lower bounds considered. The number of iterations for the Monte Carlo analysis to estimate the probability of different results was set to 10,000 as a general rule of thumb (Barreto and Howland, 2005) using Crystal Ball (Oracle, 2016). Results are shown in Section 3.3.

3 Results and Discussion

3.1 Material and Energy Balance

Fig. 3 indicates the flows of MSW into the main processing system including mass and energy balance. The total input is one tonne MSW, of which 53% of wet mass is lignocellulosic content. It has a moisture content of 40%, with remaining components broken down by sugar dry content as 45% glucose and 5% xylose as measured experimentally (totally 31.8% dry convertible lignocellulosic content). Sugar levels are key as they are the molecules that react within the hydrolysis and fermentation stages to produce ABE. Hydrolysis yield was previously

found to be 38% for glucose from experimental measurement and 70% for xylose. Thus the lignocellulosic feedstock can produce 54.4 kg glucose and 11.1 kg xylose per tonne MSW after hydrolysis reaction. The outputs from the conversion are 12.2 kg butanol (7.6 kg gasoline equivalent on energy basis), 1.5 kg ethanol (0.9 kg gasoline equivalent), 5.7 kg acetone, and 0.9 kg hydrogen as well as 0.3 kg acetic acid and 1.0 kg butyric acid. ABE product yield achieved in this study is relatively low compared to other lignocellulosic feedstocks which have been subjected to higher severity hydrothermal deconstruction pretreatments (Ibbett et al., 2011) (3.2 wt% in this study versus 11.8–14.9 wt%, on a dry substrate basis) (Baral and Shah, 2016; Montano, 2009), primarily because of the lower hydrolysis yield (38% for glucose) achieved. However, it should be noted that hydrolysis yields could be improved up to 85% and more details are discussed in Section 3.3.2. These mass transfers from MSW to biorefinery products (ABE and hydrogen) are used as the bases of LCA analysis.

The energy recovery and process energy (i.e., electricity and heat) required for each step of ABE production system are also shown in Fig. 3, which are obtained from plant operation data and process modelling. Enthalpy of materials in hydrolysis and fermentation depends on the mass and temperature of the materials. The autoclave process is the most energy intensive step accounting for about 42% of the total heat requirement and 41% total electricity requirement. The hydrolysis requires about 32% of the heat requirement and only 0.4% electricity use for mixing compared to 0.7% electricity use for fermentation. About 26% of the total heat and 4% of the total electricity use are required for distillation process for ABE product recovery. Wastewater treatment process also consumes about 54% of the total electricity to recover process water, separate lignin solid, produce biogas via equipment such as water pump, digester blower, anaerobic basin, sludge centrifuge and evaporator.

Wastewater produced during the MSW-to-ABE process can be treated into 94% clean water through anaerobic and aerobic digestion. The wastewater treatment unit can produce 389 kg lignin (35% moisture content) and 21 kg biogas and 8 kg sludge (79% moisture content) to the energy recovery unit. Considering the calorific values of lignin, biogas (51% CH₄/49% CO₂), and sludge (16 MJ/kg (Demirbas, 2017), 14 MJ/kg, and 4 MJ/kg (Humbird et al., 2011), respectively), energy recovery unit totally produces 266 kWh electricity and 1108 MJ heat (Veolia, 2012). These energy sources are used to provide energy use for autoclave and ABE biorefinery onsite. Excess electricity can be sold to the grid which can contribute to the grid to reach the target of 15% electricity using renewable sources by 2020 as regulated by the Renewable Obligation produced from (Department for Environmental Food and Rural Affairs (Defra), 2013). Due to lack of integrated district heating system in UK, excess heat is assumed to be discarded to the environment.

As shown in Fig. 3, the biogenic portion of MSW has a calorific value of 13018 MJ. For a plant capacity of 150000 t MSW/yr with operating hours of 8000, the total energy content is 28.4 MW/yr. The total products have energy values of 3.6 MW/yr from main products of ABE (37.3 MJ/kg butanol, 31.8 MJ/kg acetone and 29.7 MJ/kg ethanol) and hydrogen (141.7 MJ/kg) (The Engineering Toolbox, 2019) and 4.5 MW/yr from excess electricity. Therefore, the total energy efficiency of 28% can be obtained through the MSW derived ABE conversion system (see Table 3). In this study, electricity co-product is higher compared to other feedstocks, as a greater share of biomass is not converted to fuels and thus is available for energy recovery. Excess heat is available and, if it could be utilised (e.g., by co-located industrial process and/or district heating, for sterilization, or for cooling generation) then total energy yield could improve to 37% compared to 47% presented for ethanol production from corn stover (Humbird et al., 2011).

3.2 Life Cycle Assessment Results

LCA evaluation based on the mass and energy balance data is performed to quantify the life cycle PED and GWP associated with converting one tonne of MSW feedstock into ABE (see Fig. 4). Excluding the impact of further treating the non-biogenic content of MSW, the ABE production from MSW shows a net PED of -1.11 MJ/MJ liquid biofuels (-559.69 MJ/t MSW) and net GHG emission of -12.57 g CO₂eq/MJ liquid biofuels (-6.32 kg CO₂eq/t MSW), including co-products credits of acetone, excess electricity, and hydrogen. MSW to BE can thus achieve better PED as bioethanol produced from agricultural residues (-0.1 – 0.6 MJ/MJ fuel) (Michael et al., 2012), achieving reductions in fossil energy use by 2.3 MJ/MJ fuel compared to conventional gasoline (1.2 MJ/MJ) (Michael et al., 2012). If excess heat could be utilised, additional credits would result in net PED of -2.13 MJ/MJ liquid biofuels and net GWP of -17.56g CO₂eq/MJ liquid biofuels (40% further emission reduction).

As seen in Fig. 4, the largest energy and GHG emissions source arising from ABE production is the manufacture of enzymes, contributing approximately 2.31 MJ PED/MJ liquid biofuels and 186.7 g CO₂eq./MJ liquid biofuels. Other process inputs (pH control; *C. acetobutylicum* bacterium) and fermentation nutrients have substantially smaller impacts, totalling approximately 10% of autoclave/biorefinery GHG emissions. Process energy (electricity and heat) related emissions have been avoided through the energy recovery from the unconverted biomass (primarily lignin solids), biogas generated by the wastewater treatment plant and waste sludge. Transport of ABE to depot and to filling station account for less than 1% of the total emission.

It is noted that a low glucose hydrolysis yield of 38% has been considered, based on the experimental evidence from this study. However, a lower sugar hydrolysis and ABE yield results in more residual biomass available for co-product electricity production, thereby

providing a larger co-product GHG credit when displacing grid electricity. The impacts of these process variables on product yield and environmental impacts are discussed further in Section 3.3.

3.3 Sensitivity Analysis

3.3.1 Sensitivity analysis of background systems

The life cycle GHG emissions per MJ MSW derived liquid biofuels is compared to results of gasoline (the EU fossil fuel comparator, US Environmental Protection Agency (EPA) and US oil shale based on Energy Research Architecture (ERA)), and bioethanol (i.e., ethanol produced from ethanol, sugarcane, corn stover, switchgrass, and miscanthus). Various life cycle GHG emissions are also present for various electricity replacement of electricity mix, electricity from hard coal, natural gas, hydro power, wind power in UK and electricity mix in US and China (see Fig. 5).

Overall GHG emission of MSW derived liquid biofuels provides 115%, 114% and 109% reduction compared to the transportation gasoline based on EU RED (84 g CO₂eq/MJ) for the base case, US EPA (93 g CO₂eq/MJ) and US ERA (139 g CO₂eq/MJ) (Pieprzyk et al., 2009), respectively. MSW to bioethanol can give 14.5–56.4 g CO₂eq/MJ ethanol (Chester and Martin, 2009; Kalogo et al., 2007; Schmitt et al., 2012; Zhang et al., 2010a); therefore MSW to BE can achieve over 100% relative GHG reduction than MSW to bioethanol only. The difference is primarily because of the different system boundaries (e.g., waste collection and hauling, classification, coproducts considerations, and landfill treatment of residual waste) and assumption (e.g., enzyme production and fermentation microorganism) made in the studies. MSW derived liquid biofuels has smaller GHG emissions than other lignocellulosic ethanol pathways (corn stover, switchgrass, miscanthus) and reduction of over 100% relative to corn and sugarcane (Michael et al., 2012).

The GHG emissions assigned to enzyme production in the current study are 5.9 g CO₂eq/g of produced enzyme (commercially Novozymes Cellic CTec2) (Kløverpris, 2018) while some onsite enzyme production process using glucose as a feedstock have been reported to emit 4.1–11.5 g CO₂eq/g cellulose (Hsu et al., 2010; McKechnie et al., 2015). However, the newly produced Cellic 1.0 series (0.64 g CO₂eq/g enzyme) have achieved significant reduction in GHG emissions. We also considered different Novozymes Cellic CTec series enzyme products: Cellic CTec in 2009 (7.25 g CO₂eq/g), Cellic CTec3 in 2012 and 2014 (5.1 g CO₂eq/g), and assumed Cellic X in 2022 (0.32 g CO₂eq/g) but their respective dosage is not described here due to commercial confidentiality. Their relative impacts have been shown in Fig. 5 (MSW- liquid biofuels 2), indicating a range of –135 to 22.13 g CO₂eq/MJ liquid biofuels. Furthermore, improved potency of enzyme will lead lower dosages of enzyme in hydrolysis in the future, achieving further GHG emission reductions but beyond the scope of this study.

Due to the low GHG intensity of 1.7 g CO₂eq per MJ electricity produced from wind power, 1 MJ MSW derived liquid biofuels emits 201 g CO₂eq compared to –248.88 g CO₂eq using coal electricity source of which the GHG intensity is 266.3 g CO₂eq per MJ electricity (see Fig. 5). This is due to different credits of fixed excess electricity can achieve via electricity displacement. GHG intensity of China electricity mix (240.8 g CO₂eq/MJ electricity) lead the largest GHG reduction of –205.54 g CO₂eq/MJ liquid biofuels compared to –96.74 g CO₂eq/MJ liquid biofuels using US electricity mix (176.9 g CO₂eq/MJ electricity) and -12.57 g CO₂eq/MJ liquid biofuels using UK electricity mix (112.1 g CO₂eq/MJ electricity).

3.3.2 Sensitivity analysis of process variables

Across various parameters as listed in Table 4, ABE product yield can be achieved at a range of 10.73–82.14 kg/t MSW(see Fig. 6a) while a certainty of 90% in the range of 18.3–48.8 kg

/t MSW. The composition of incoming MSW feedstock varies but the biogenic content including paper, card, food waste, garden waste, wood, and other organic lies in the range of 30-85%. The maximum 85% of biogenic content is assumed for residue MSW exiting material recycling facility where non-biogenic content such as plastics, ferrous and non-ferrous metals are removed. By applying Defra average data, lignocellulosic content of MSW is about 82% (paper 21.6%, card 17.3%, food waste 31%, garden waste 5.3%, wood 3.2%, and other organic 3.5%) after going through material recycling facility. A hypothetical lower bound of 30% is considered to evaluate the impact of low biogenic content on the product yield. We have conservatively assumed a glucose hydrolysis yield of 38% based on the current experimental evidence, but we also analyse scenarios where the efficiencies of theoretical glucose ranges of 30–85% and xylose ranges of 60–90% (Kalogo et al., 2007). Solid concentration rate ranges from 20% to 30%; the base case is 20%. It should be noted that the variability with product yield would have a large impact on associated financial aspects, where the trade-offs need to be addressed in future optimisation. But before considering the optimisation, how process parameters affect the product yield needs to be understood as follows in the next section.

The impacts of process parameter variability on the product yield are evaluated as in Fig. 7a. As biogenic fibre content and glucose hydrolysis yield determines the quantity of total inputs of the conversion, it has the largest impact on the product yield. It indicates the importance of selection of higher biogenic content of MSW from collection facilities or materials recycling facilities. Also optimisation of hydrolysis yield is also the key in future research focus as this is the main barrier to maximising product output. Solid concentration in hydrolysis does not have any direct impact on the absolute product yield but affects the final product concentration as discussed below.

Under these scenarios, the ABE product concentration lies in the range of 15.1–79.6 g ABE/L (9.4–47.5 g butanol/L) while the base case is 17.8 g ABE/L (11.2 g butanol/L) (see Fig. 6b) compared to 6.2–64.2 g ABE/L (~10 g/L) as reported (Bankar et al., 2013; Grisales Díaz and Olivar Tost, 2017). However, the high concentration of substrate and ABE products can be toxic to microorganisms that the fermentation can be inhibited completely at butanol concentration of approximately 15 g/L (Grisales Díaz and Olivar Tost, 2017). Glucose hydrolysis yield and solid concentration contribute to the largest variance of the results (88%) (see Fig. 7b). Therefore, future optimisation also needs to consider trade-offs amongst increasing product yield and product concentration level and reducing growth inhibition effect, for instance, by continuous removal of solvents (Brito and Martins, 2017).

Under the parameters considered as in Table 4, the net GWP ranges from –26.68 to 6.61 g CO₂eq/MJ liquid biofuels while the base case is -12.57 g CO₂eq/MJ liquid biofuels (see Fig. 6c). With 90% certainty the GWP of conversion from MSW to liquid biofuels lies in the range of -16.82–2.60 g CO₂eq/MJ liquid biofuels. Compared to the gasoline, MSW derived liquid biofuels can reduce GHG emissions by 92%% to over 100%, which can significantly contribute to GHG emission reductions of transport fuels by a minimum of 6% by 2020 compared to the 2010 level as regulated by EU Fuel Quality Directive (European Union Fuel Quality Directive, 2009), and meet the sustainability criteria for biofuels by EU Renewable Energy Directive (GHG savings of at least 60% in comparison to fossil fuels in 2018) (European Union Renewable Energy Directive, 2009). Glucose hydrolysis yield and biogenic fibre content has the largest impact on the quantity of total GWP as in Fig. 7c.

Unlike the impact on product yield, uncertainty of process parameters shows different impacts on GHG emissions. Although the increase of sugar yield/sugar content can be beneficial to the increase of product yield, it is detrimental to the GWP reduction. High

biogenic content with relatively low ABE product yield (e.g., low sugar content/yield) can minimise overall GWP per MJ liquid biofuels. This is primarily because more unconverted biogenic fibre residue goes to energy recovery unit and generates more co-product electricity as discussed in Section 3.2.

4 Conclusions

The study presents the conversion of ABE from municipal solid waste feedstocks via autoclave, enzymatic hydrolysis, and ABE fermentation based on experimental data and process modelling, demonstrating the conversion to be a promising way to valorise organic wastes in the production of liquid biofuels. The mass and energy balance results show the overall conversion process is energy self-sufficient as process energy demand (steam and electricity) is fully covered by the energy recovery from lignin from hydrolysis and fermentation residue and biogas and sludge produced from wastewater treatment while surplus electricity be exported to the grid as co-product benefits. In total, the integrated process can produce 12.2 kg butanol (7.6 kg gasoline equivalent), 1.5 kg ethanol (0.9 kg gasoline equivalent), 5.7 kg acetone, and 0.9 kg hydrogen as well as 0.3 kg acetic acid and 1.0 kg butyric acid from each tonne raw MSW. It should be noted that hydrolysis yield can be potentially improved up to 85% for glucose conversion as discussed which would increase ABE solvent output to about 39 kg.

MSW-derived liquid biofuels can contribute to reduce GHG emissions by over 100% % compared to gasoline base on EU RED standard and conventional bioethanol. Monte Carlo simulations indicate the ABE product yield with a certainty of 90% in the range of 18.3-48.8 kg /t MSW. Correspondingly, the net GWP ranges from -16.82 to 2.60 g CO₂eq/MJ liquid biofuels with a 90% certainty while the base case is -12.57 g CO₂eq/MJ liquid biofuels.

Sensitivity analysis results indicate high biogenic content with relatively low ABE product yield (e.g., low sugar content/yield) can minimise overall GWP per MJ liquid biofuels.

However, key technology challenges still need to be addressed in improving hydrolysis yield which has already showed the potential to be up to 90% as this is the main barrier to maximising product output for future commercialisation. Ongoing investigation is studying the non-biogenic content of MSW recycling such as metal and plastic recycling benefits in order to comprehensively quantify the waste management of MSW. Future work also goes for waste supply chain investigation and facility design optimisation (e.g., capacity; colocation).

Nomenclature

GHG	Greenhouse gas
MSW	Municipal solid waste
LCA	Life cycle assessment
ABE	Acetone, butanol, ethanol
PED	Primary energy demand
GWP	Global warming potential
RED	Renewable Energy Directive
US EPA	US Environmental Protection Agency
US ERA	US Energy Research Architecture

Notes

The authors declare no competing financial interest.

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Figure Captions

Fig. 1 Overall diagram for MSW to ABE conversion (dashed line means outside the system boundary)

Fig. 2 Sugar composition in MSW fibre by total acid hydrolysis as measured by ion chromatography (weights determined in the monomer form, against total dry weight of sample). Residual is difference from 100, which accounts for all other organic and non-organic material present in the biofiber, which is not suitable for fermentation.

Fig. 3 Diagram showing the inputs into and outputs from the process of ABE production from MSW. Dashed line shows the excluded flow in this analysis

Fig. 4 a) Primary energy demand and b) Global warming potential of MSW derived liquid biofuels

Fig. 5 Comparison of life cycle global warming potential per MJ fuels amongst gasoline, MSW derived liquid biofuels, and bioethanol.

Fig. 6 Monte Carlo analysis of the values of a) ABE product yield; b) ABE product concentration; c) life cycle GWP of MSW derived liquid biofuels; at various process parameters. Dashed lines show the base case.

Fig. 7 Relative contribution of processing parameters on the sensitivity of a) ABE product yield; b) ABE product concentration; c) life cycle GWP of MSW derived liquid biofuels.

Table 1 Process data of MSW to ABE conversion.

Table 2 Parameters of ABE fermentation of MSW hydrolysates.

Table 3 Overall mass and energy balance of ABE production from MSW.

Table 4 Parameters for Monte Carlo simulation.

648 **Tables and Figures**649 **Table 1**

650 Process data of MSW to ABE conversion.

	Input		Quantity of product	
Feedstock	MSW	1 t MSW		
	Moisture content of MSW	40%		
Autoclave	Fibre yield ex autoclave+ sieve	53%	Autoclaved fibre dry	318 kg
	Energy consumption			
	Electricity	43 MJ/t MSW		
	Heat	274 MJ/t MSW		
	Process water	245 kg/t MSW		
Hydrolysis	Glucose content	45%	Sugar	65.51 kg
	Xylose content	5%		
	Glucose hydrolysis yield	38%		
	Xylose hydrolysis yield	70%		
	Solid content in hydrolysis slurry	20%		
	Energy consumption			
	Electricity	0.41 MJ/t MSW		
	Heat	214.63 MJ/t MSW		
	Enzyme loading, w enzyme/w sugar	15.90 kg/t MSW		
	volume 6M H2SO4 (for pH control)	8.59 kg/t MSW		
	Weight of water added for slurry	1035.51 kg		
	Clean water usage	5366.25 kg		
Fermentation	Sugar use efficiency	90%		
	ABE yield	33%		
	Energy consumption			
	Electricity	0.68 MJ/t MSW		
	<i>Clostridium acetobutylicum</i>	0.16 kg		
	Monopotassium phosphate	1.09 kg		
	Dipotassium phosphate	0.83 kg		
	Ammonium acetate	3.17 kg		
	Yeast extract (Duchefa)	2.73 kg		
	FeSO4.7H2O	0.01 kg		
	MgSO4.7H2O	1.09 kg		
	Process water	4509.87 kg		

Distillation	Energy consumption			Acetone	5.716	kg
	Electricity	3.31	MJ/t MSW	Butanol	12.24	kg
	Heat	125.30	MJ/t MSW	Ethanol	1.52	kg
				Acetic acid	0.28	kg
				Butyric acid	1.02	kg
Gas stripping	Energy consumption			H ₂	0.93	kg
	Electricity	0.56	MJ/t MSW	CO ₂	30.66	kg
	Heat	46.15	MJ/t MSW			
	Process water	10.75	kg			
Wastewater Treatment	Energy consumption			Clean water recovered	10871.18	kg
	Electricity	56.55	MJ/t MSW	Net water required	-296.20	kg
	Heat	0.02	MJ/t MSW			
Energy recovery	Electricity	959.03	MJ/t MSW			
	Heat	1107.76	MJ/t MSW			
	Excess Electricity	854.51	MJ/t MSW			
	Excess Heat	447.66	MJ/t MSW			
Transport of ABE to depot	Transport per					
	Truck for liquids (diesel)	150	km	2.92	ton·km / t MSW	
	Fuel	Diesel				
	Energy consumption depot					
	Electricity UK mix	0.00084	MJ/MJ liquid biofuels			
Transport to filling station	Transport per					
	Truck for liquids (diesel)	150	km	2.92	ton·km / t MSW	
	Fuel	Diesel				
	Energy consumption depot					
	Electricity UK mix	0.0034	MJ/MJ liquid biofuels			

Table 2

Parameters of ABE fermentation of MSW hydrolysates.

Type	Formula	Molar ratio	Weight percentage
Glucose	C ₆ H ₁₂ O ₆	1	-
Acetic acid	CH ₃ COOH	0.014	0.5%
Butyric acid	C ₄ H ₈ O ₂	0.035	1.7%
Acetone	C ₃ H ₆ O	0.3	9.8%
Butanol	C ₄ H ₉ OH	0.5	20.8%
Ethanol	C ₂ H ₅ OH	0.1	2.6%
Carbon dioxide	CO ₂	2.1	52.0%
Hydrogen	H ₂	1.4	1.6%
Microbial cells	CH _{1.8} O _{0.5} N _{0.2}	-	11%

Table 3

Overall mass and energy balance of ABE production from MSW.

Inputs	Tonne/year	MW
MSW (40% moisture)*	150000	28.4/yr
Total input		28.4/yr
Outputs		
Acetone	947	0.9/yr
Butanol	2028	0.2/yr
Ethanol	252	2.4/yr
Hydrogen	139	0.001/yr
Heat generation		5.8/yr
Electricity generation		5.0/yr
Total output		14.4/yr
Process heat demand		3.2/yr
Process electricity demand		0.5/yr
Net heat surplus		2.4/yr
Net electricity surplus		4.5/yr
Energy efficiency (main product + net electricity surplus)		28%
Energy efficiency(main product + net electricity surplus)- ethanol from corn stover (Humbird et al., 2011)		47%

* (Excluding non-biogenic content)

Table 4

Parameters for Monte Carlo simulation.

Parameter	Distribution
Biogenic fibre content	Triangular (Min: 30%, Max: 85%, Mode: 53%)
Glucose content	Triangular (Min: 40%, Max: 65%, Mode: 45%)
Xylose content	Triangular (Min: 5%, Max: 15%, Mode: 5%)
Glucose hydrolysis yield	Triangular (Min: 30%, Max: 85%, Mode: 38%)
Xylose hydrolysis yield	Triangular (Min: 60%, Max: 90%, Mode: 70%)
Solid concentration in hydrolysis	Triangular (Min: 20%, Max: 30%, Mode: 20%)

Fig. 1 Overall diagram for MSW to ABE conversion (dashed line means outside the system boundary)

Fig. 2 Sugar composition in MSW fibre by total acid hydrolysis as measured by ion chromatography (weights determined in the monomer form, against total dry weight of sample). Residual is difference from 100, which accounts for all other organic and non-organic material present in the biofiber, which is not suitable for fermentation.

Fig. 3 Diagram showing the inputs into and outputs from the process of ABE production from MSW. Dashed line shows the excluded flow in this analysis.

Fig. 4 a) Primary energy demand and b) Global warming potential of MSW derived liquid biofuels.

Fig. 5 Comparison of life cycle global warming potential per MJ fuels amongst gasoline, MSW derived liquid biofuels, and bioethanol¹.

¹ Notes: MSW-BE Electricity source (sensitive coproduct credits from electricity replacement of electricity mix, electricity from hard coal, natural gas (NG), wind power in UK and electricity mix in US and China); MSW-BE Enzyme (various series of enzyme production); gasoline (the EU fossil fuel comparator, US EPA and US oil shale based on ERA); bioethanol (cellulosic ethanol produced from ethanol, sugarcane, corn stover, switchgrass, and miscanthus)

688 Fig. 6 Monte Carlo analysis² of the values of a) ABE product yield; b) ABE product
689 concentration; c) life cycle GWP of MSW derived liquid biofuels; at various process
690 parameters. Dashed lines show the base case.

691

² Note: For the box plot, the ranges are calculated as follows: The box, from bottom to top, indicates the 25th (Q1), 50th and 75th (Q3) percentile values from the data plotted. The interquartile range (IQT) is $Q3 - Q1$. The whisker below is defined as minimum and the whisker above is defined as maximum.

Fig. 7 Relative contribution of processing parameters on the sensitivity of a) ABE product yield; b) ABE product concentration; c) life cycle GWP of MSW derived liquid biofuels.