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Thermo-Catalytic Reforming of municipal solid waste

Miloud Ouadi [a,c,*], Nils Jaeger [a,c], Charles Greenhalf [c], Joao Santos [c], Roberto Conti [b], Andreas Hornung [a,b,c,d]

[a] Fraunhofer UMSICHT, Germany
[b] University of Bologna, Italy
[c] Birmingham University, UK
[d] Erlangen University, Germany

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ABSTRACT

Municipal Solid Waste (MSW) refers to a heterogeneous mixture composed of plastics, paper, metal, food and other miscellaneous items. Local authorities commonly dispose of this waste by either landfill or incineration which are both unsustainable practices. Disposing of organic wastes via these routes is also becoming increasingly expensive due to rising landfill taxes and transport costs. The Thermo-Catalytic Reforming (TCR®) process, is a proposed valorisation route to transform organic wastes and residues, such as MSW, into sustainable energy vectors including (H_2 rich synthesis gas, liquid bio-oil and solid char). The aim herein, was to investigate the conversion of the organic fraction of MSW into fuels and chemicals utilising the TCR technology in a 2 kg/h continuous pilot scale reactor. Findings show that MSW was successfully processed with the TCR after carrying out a feedstock pre-treatment step. Approximately, 25 wt.% of the feedstock was converted into phase separated liquids, composed of 19 wt.% aqueous phase and 6 wt.% organic phase bio-oil. The analysis of the bio-oil fraction revealed physical and chemical fuel properties, higher heating value (HHV) of 38 MJ/kg, oxygen content <7 wt.% and water content <4 wt.%. Due to the bio-oil's chemical and physical properties, the bio-oil was found to be directly miscible with fossil diesel when blended at a volume ratio of 50:50. The mass balance closure was 44 wt.% synthesis gas, with a H_2 content of 36 vol% and HHV of 17.23 MJ/Nm^3, and 31 wt.% char with a HHV of 17 MJ/kg. The production of high quantities of H_2 gas and highly de-oxygenated organic liquids makes downstream hydrogen separation and subsequent hydro-deoxygenation of the produced bio-oil a promising upgrading step to achieve drop-in transportation fuels from MSW.

1. Introduction

Municipal Solid Waste (MSW) is a heterogeneous mixture of everyday household rubbish collected and disposed by local authorities. It is typically made up of plastic, biomass, food, metal, glass, rubber, textiles, paper, card and other inorganic materials. These materials are present in highly variable quantities and forms with mixed contents of moisture and inorganics. This mixture makes disposal expensive and limited but the materials are highly abundant with energy contents comparable to coal (on a dry basis). It is attractive for fuel synthesis if the energy within can be efficiently converted into a homogenous form, acceptable to current energy generating devices (boilers, combustors, turbines, fuel cells or engines).

Approximately 40 million tonnes of MSW is produced every year in the UK (on a wet basis) and this is forecasted to rise to 50 million tonnes per year by 2020 (ICERPA, 2005). MSW is currently disposed by incineration or landfill, both of which are unsustainable practices as they are costly, lead to greenhouse gas emissions and in the case of landfilling, use up land that could be used for other purposes, such as arable or pastoral farming (Rajendran et al., 2014). Landfilling wastes also contributes to pollution of surface water, ground water, soil and air (Environmental Agency, 2011). With the forecasted yearly rise of MSW, legislation has been enforced by the European Union (EU) under the EU landfill directive (CEC, 1999; He et al., 2010; Stringfellow et al., 2011),

Abbreviations: TCR, Thermo-Catalytic Reforming; MSW, municipal solid waste; HHV, higher heating value; TAN, total acid number; DEFRA, Department for Environment, Food and Rural Affairs; RDF, refuse derived fuel; SRF, solid recovered fuel; PAH, polycyclic aromatic hydrocarbons; MAH, mono-aromatic hydrocarbons; FAME, fatty acid methyl esters; PFD, process flow diagram.

* Corresponding author at: Fraunhofer UMSICHT, Sulzbach-Rosenberg, 92237, Germany.

E-mail addresses: miloud.ouadi@umsicht.fraunhofer.de (M. Ouadi), nils.jaeger@umsicht.fraunhofer.de (N. Jaeger), c.e.greenhalf@bham.ac.uk (C. Greenhalf), jns699@student.bham.ac.uk (J. Santos), roberto.conti5@unibo.it (R. Conti), andreas.hornung@umsicht.fraunhofer.de (A. Hornung).

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to prohibit waste sent to landfill without prior treatment. This legislation also imposes targets to reduce the amount of biodegradable MSW to landfill by 65% of 1995 levels by 2020. To achieve this target, the UK Department for Environment, Food and Rural Affairs (DEFRA) implemented a waste strategy for MSW to increase the quantity of materials recycled, composted and converted into energy (Defra, 2007). The biodegradable fraction of MSW is eligible, under the EU renewable energy directive, to contribute towards targets. This target aims to achieve 20% of all energy from renewable sources by 2020. Thermal conversion of these wastes is essential to achieving these targets.

It has been demonstrated that the biodegradable fraction of MSW can be successfully separated, recovered and converted into either a refuse derived fuel (RDF) or solid recovered fuel (SRF) (Garg et al., 2007; Gallardo et al., 2014). State of the art commercial application of this fuel, for energy generation, remains to be combustion mainly by cement industries for heating cement kilns (Yeqing et al., 2012). Combustion of these fuels could lead to a significant increase in CO_2 emissions and other pollutants such as NOx, SOx, and dioxins. These pollutants pose a hazard to both the environment and human health if they are not carefully controlled. Direct combustion of this material therefore requires sophisticated and expensive downstream flue gas conditioning equipment. The advantage of processing these waste streams via advanced thermal conversion routes such as Thermo-Catalytic Reforming is that emissions can be controlled to much lower levels without the need for expensive and sophisticated flue gas treatment. For example, it known that a lower combustion temperature of bio-oils reduces NOx emissions in downstream engines (Hossain et al., 2013).

In the last 10 years, the conversion of MSW for renewable energy recovery and chemicals has gained significant importance as a research topic. Conversion of MSW via incineration has been state of the art for decades, whereas gasification and pyrolysis are not well-established technologies, with only a few applications worldwide (Rada et al., 2009). Zaman, 2009, performed a comparative life cycle assessment comparing fast pyrolysis of MSW with anaerobic digestion, incineration and landfill. The authors found that pyrolysis, for bio-oil production, was one of the most environmentally friendly routes for MSW valorisation. One key advantage of using pyrolysis to process MSW wastes is that a significant proportion of the waste is converted into a homogenous solid char which does not produce tars when combusted or gasified downstream. Most industrial pyrolysis facilities can use MSW as a feedstock by integrating gasification or combustion stages (Chen et al., 2014). Of particular interest is the use of plasma gasifiers which implement extremely high temperatures >1200 °C for the destruction of tars and dioxins. Research efforts are exploring alternative thermal conversion routes such as gasification (Choy et al., 2004; Luo et al., 2012) and pyrolysis (Fichtner, 2004) of MSW. However, advances within this area are difficult due to the degree of separation and pre-processing required to condition MSW into a suitable homogenous form for subsequent thermal conversion. Gasification and pyrolysis are promising conversion routes, when compared to combustion, because of lower emissions, higher recycling rates, smaller carbon footprints and lower capital costs.

Luo et al., 2012 investigated catalytic steam gasification of MSW to produce hydrogen rich gas, using $NiO/\gamma\text{-}Al_2O_3$ as catalyst. Total gas yields (wt.%) and hydrogen gas composition (mol.%) were found to increase with increasing steam to carbon ratios. Similarly, catalyst addition further improved hydrogen composition and total gas yields. In comparison to gasification without steam or catalysts addition, the additional upgrading step is crucial to the optimisation of gas composition and yields. It is important to consider the additional expenses associated with upgrading and its impact on overall process economics. Ates et al. (2013) investigated pyrolysis of MSW using a small laboratory scale batch reactor at temperature of 500 °C with catalysts (Y-zeolite, β-zeolite, FCC, MoO3, Ni–Mo, HZSM-5 and $Al(OH)_3$). Catalyst tests were carried out at a temperature of 500 °C with 10 wt.% of catalyst. Each catalyst tested produced higher bio-oil yields (between 11.2 and 20.0 wt.%) when compared with test without catalyst (9.75 wt.%).

This paper investigates an alternative thermal conversion route for processing pre-conditioned (organic fraction) of MSW into value added products for power and heat generation. The route explored is a novel Thermo-Catalytic Reforming process (TCR®) which essentially combines intermediate pyrolysis (heating in the complete absence of oxygen at moderate temperatures and solid residence times of a couple of minutes) with post catalytic reforming (further heating at elevated temperatures under catalytic conditions). The advantages of this process compared to incineration are that it produces low oxygenated high-energy dense liquids that are easily transportable and storable, energy dense chars and H_2 rich synthesis gas. The compositions of these products make them extremely valuable for energy storage or further downstream synthesis into liquid fuels for transport (Neumann et al., 2016). The energy efficiency of the TCR process coupled to an engine generator is also much higher at <5 MWth scale when compared with direct fluidised bed incineration and grate incineration which require downstream steam turbines for electricity production, these suffer from inefficiencies and are uneconomical to implement at small scales. This makes it more favourable to implement at smaller decentralised scales. The trials described in this paper were performed using a continuous pilot scale 2 kg/h TCR reactor.

2. Materials and methods

2.1. Raw materials

MSW acquired for this work was sourced from a household kerbside collection authority in Merseyside UK. The waste is also known in the UK as residual MSW (black bag waste). The general composition of the collected waste is representative of household MSW produced across the UK in 2014, as depicted in Fig. 1 (DEFRA, 2014). As received, the feedstock had an average moisture content of 35 wt.% and was composed of a heterogeneous mixture of materials, consisting of plastics, biomass, metals, glass, papers, textiles, rubbers, building rubble, concrete, soil, fines, electrical pieces and food wastes. The particular composition of MSW used in TCR trials is shown in Table 1.

2.2. Feedstock pre-treatment

Before commencing TCR trials the feedstock required an initial pre-treatment step (drying, sorting, shredding and pelletising). This was carried out at a demonstration mechanical heat treatment facility located at Huyton UK. The plant was built as part of DEFRA's new technologies demonstrator programme. The demonstration plant treated 20,500 tonnes of household residual waste, via a combination of heat treatment and mechanical separation technologies, to produce a solid recovered fuel (SRF) pellets and recyclables (glass, plastics, ferrous and non-ferrous metals) (Stringfellow et al., 2011).

The pre-treatment steps are shown in Fig. 2 (Stages 1–4) and consisted of the following procedure. First the incoming waste was separated by a trommel into two size fractions, the oversized material was directed to an inspection conveyor belt for manual removal of large items (such as gas bottles, and concrete blocks). The remaining oversized material was shredded and mixed with the undersized material. The material was then dried to a moisture

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