Steam Gasification of Refuse-Derived Fuel in a Rotary Kiln Pilot Plant: Experimental Tests

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Refuse-derived fuel (RDF) steam gasification tests were performed in a rotary kiln pilot plant. The gas features were evaluated by varying the feeding ratio (FR), in the range 0.4-2.67, at a constant temperature equal to 850 °C. Several experimental tests were carried out in order to evaluate the best values for the main operating variables: kiln temperature, gas and solid residence time, etc., before evaluating the effect of the FR increase on the gas energy content and composition.

Results show that the gas energy content decreases as the FR increases and, in the range of the FR studied, it achieves the maximum for FR=0.4, which corresponds to a volumetric gas composition of H2=59.1 %v/v, CO=16.8 %v/v, CO2=20.1 %v/v, CH4=3 %v/v (not considering N2) and the highest lower heating value (LHV) equal to 16 MJ kggas-1 is obtained.

1. Introduction

As solid waste production and landfill saturation problems increase, there is a growing number of studies on prevention, recycling and recovery of matter and energy from wastes through proper waste management. Thermal treatment, as pyrolysis and gasification of refuse-derived fuel (RDF), is an emerging waste-to-energy technique (Malkow, 2004) that overcomes some of the problems (Lancia et al., 1996) due to the need for cleaning incineration flue gas (Lancia et al. 1997; Prisciandaro et al. 2009).

RDF is a fuel produced by shredding and dehydrating municipal solid waste (MSW), after eliminating inert material (e.g. glass and metals), and largely consists of organic components of municipal waste such as plastics and biodegradable waste. The fraction of inert waste is stored in clusters of standard dimensions and weight and then destined to recycling or alternatively to landfill. Gasification permits to transform an initial matrix, such as RDF, in a gaseous energy carrier, called syngas. Gaseous energy carriers are very flexible and offer numerous advantages and possibilities when used in high efficiency generation systems, in innovative fuel cells (Molino et al., 2013; Kivisaari et al., 2004) and for the production of high valued chemicals (Hermann et al., 2001; Young, 2010; Stantec, 2010; Galvagno et al., 2007). Syngas cleaning is required for it to be used in high-efficiency power generation units. Cleaning is performed by using well-assessed processes based on condensation (Aris and Zainal, 2011), absorption (Lancia and Musmarra; 1999) or adsorption (Di Natale et al., 2011).

Different options are available for gasification in terms of operating conditions: pressure (1-30 bar), temperature (1000-1700 K), gasifying agent (air, O2, steam); and gasifier type (fixed bed, fluidized bed, rotary kiln, etc.) (Molino et al., 2012). Typical air-based gasifiers produce a syngas with a high nitrogen concentration and a lower heating value (LHV) in the range of 4-7 MJ Nm-3; O2/steam-based gasifiers, on the other hand, produce a syngas with a high concentration of H2 and CO and a LHV in the range of 10-20 MJ Nm-3 (Wang and Kinoshita, 1992). Wang and Kinoshita (1992) also performed several steam and oxygen gasification tests on an indirectly heated fluidized bed biomass gasifier to study, inter alia, the effects of gas residence time. They found that gas residence time in the reactor affects the composition, because the H2 and CO2 contents in syngas increase and decrease, respectively, with increasing gas residence times.

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Yang et al. (2001) showed that RDF pyrolysis in the range of 500-900 °C produces a gas whose main components are H2, CO, CH4, C2H6, C2H4, C3H8 and C3H6. Furthermore, H2 and CH4 content increases as temperature increases. Steam gasification of RDF in the range of 850-1050 °C was studied by Galvagno et al. (2006), they observed that higher temperatures result in higher syngas production and H2 content in syngas. Dalai et al. (2009) studied the gasification of two different RDFs in a fixed bed reactor using steam and oxygen like gasifying agents. They showed that H2 and CO contents in syngas increase at increasing process temperatures up to 725 °C. At 725 °C, over 70% of volume of syngas is made of H2 and CO. Moreover, the heating value of gas decreases as temperature increases. However, few studies were conducted to investigate the RDF gasification in pilot-scale with different conditions. Arena et al. (2010) carried out experimental air gasification of RDF in a bubbling fluidized bed reactor utilizing beds of natural olivine under various values of equivalence ratio (ER). The gasification agent is fuelled bottom-up from the bed through a distribution grid, with a velocity between 1 and 3 m/s. In these conditions, the bed of olivine is fluidised and is continuously stirred by gas bubbles whose mobility ensures uniform conditions and enhances both matter and heat exchange between solid and gas. The size of the air feeding grid significantly affects mass and heat transfer between the gasification agent and the bed solids (Vaccaro et al., 1997; Musmarra, 2000). The results confirmed that a high heating value, a high hydrogen concentration and a drastic reduction in tar content are obtained. This paper reports the results of experimental tests performed on a continuous pilot-scale rotary kiln plant, using RDF as feedstock. The influence of steam/RDF ratio on yield and composition of the syngas was investigated, keeping the temperature of reactor and the solid residence time at a constant value.

2. Gasification Process

Partial oxidation in the gasification process transforms feedstock into a syngas composed primarily of H2, CO, CO2, H2O, CH4, N2 and other hydrocarbons (C2+). Schematically it comprises three main steps:

a. Drying (for moisture loss in the matrix)

$$\text{RDF}_{\text{wet}} \rightarrow \text{RDF}_{\text{dry}} + \text{H}_2\text{O}$$

b. Pyrolysis (thermal degradation of matrix in absence of an oxidant agent with formation of char, tar, and gaseous compounds)

$$\text{RDF}_{\text{dry}} \rightarrow \text{char} + \text{tar} + \text{wH}_2\text{O} + \text{cCO} + \text{bCO}_2 + \text{hH}_2 + \text{aCH}_4$$

Product ratio and gas composition depend on the type of matrix, heating velocity and temperature.

c. Gasification (products of pyrolysis react with the gasifying agent in order to obtain different combustible product: CO, H2, CO2, CH4, CxHy)

Gasification is a complex chemical process and the main reactions involved are summarized below (Littlewood, 1997):

Oxidation reactions:

$$\text{C} + \frac{1}{2}\text{O}_2 = \text{CO} \quad \Delta H_r = -110.6 \text{ kJ·mole}^{-1} \quad (1)$$

$$\text{C} + \text{O}_2 = \text{CO}_2 \quad \Delta H_r = -393.8 \text{ kJ·mole}^{-1} \quad (2)$$

$$\text{nH}_\text{m} \text{ + } \frac{n}{2}\text{O}_2 = n\text{CO} + \frac{n}{2}\text{H}_2 \quad \text{Exothermic} \quad (3)$$

Boudouard reaction:

$$\text{C} + \text{CO}_2 = 2\text{CO} \quad \Delta H_r = 172.6 \text{ kJ·mole}^{-1} \quad (4)$$

Steam gasification:

$$\text{C} + \text{H}_2\text{O} = \text{CO} + \text{H}_2 \quad \Delta H_r = 131.4 \text{ kJ·mole}^{-1} \quad (5)$$

$$\text{C} + 2\text{H}_2\text{O} = \text{CO}_2 + 2\text{H}_2 \quad \Delta H_r = 87.2 \text{ kJ·mole}^{-1} \quad (6)$$

$$\text{nH}_\text{m} \text{ + } n\text{H}_2\text{O} = n\text{CO} + \left( n + \frac{n}{2} \right)\text{H}_2 \quad \text{Endothermic} \quad (7)$$

Methanation and reforming:

$$\text{C} + 2\text{H}_2 = \text{CH}_4 \quad \Delta H_r = -74.9 \text{ kJ·mole}^{-1} \quad (8)$$

$$\text{CH}_4 + \text{H}_2\text{O} = \text{CO} + 3\text{H}_2 \quad \Delta H_r = 201.9 \text{ kJ·mole}^{-1} \quad (9)$$
\[
\begin{align*}
2C + 2H_2O & = CH_4 + CO_2 & \Delta H_r & = 15.3 \text{ kJ} \cdot \text{mole}^{-1} \quad (10) \\
CO + H_2O & = CO_2 + H_2 & \Delta H_r & = -41.2 \text{ kJ} \cdot \text{mole}^{-1} \quad (11) \\
\text{Thermal cracking reactions:} \\
pC_{x}H_{y} & = qC_{m}H_{m} + rH_{2} & \text{Endothermic} \\
C_{n}H_{m} & = nC + \frac{m}{2}H_{2} & \text{Endothermic} \\
\end{align*}
\]

3. Material and apparatus

3.1 Material
The RDF under investigation was a simulated material, produced by a local company and characterized by the composition indicated below:

*Plastic:* plastic containers for packing of food and objects in plastic (30.5 %w/w);
*Paper:* miscellaneous paper used to food, office, packing, etc. (16 %w/w);
*Cardboard:* containers for miscellaneous packing (22.5 %w/w);
*Textiles:* rags, woven and wood (31 %w/w).

3.2 Apparatus
The experimental tests were performed in a continuous pilot-scale rotary kiln plant, with indirect electric heating system to control the temperature of reaction (Donatelli et al. 2010). On attainment of process temperature (after electrical heating of reactor) the RDF and the gasifying agent (steam or air) were fed to the rotary kiln. If the gasifying agent used was steam, a secondary N\textsubscript{2} flow-rate was sent to reactor:
- to create an inert atmosphere in the starting phase of reactor heating (washing);
- to transport gas in the plant (carrier);
- to avoid spillages of syngas outside the system.

Steam was produced by a heat exchanger powered with flue gas resulting from the combustion of liquefied petroleum gas (LPG) and, also, of syngas. Then the steam was fed to the reactor by means of a valve and a volumetric counter placed directly on the circuit of water adduction. The steam was heated into the rotary kiln up to 700 °C before the contact with the material to gasify.

Feeding of RDF and discharge of solid residue (char) from the reactor were continuous, and a stationary condition was reached after 0.5 h. The char was collected in a tank at the end of the rotary kiln and unloaded after the test.

The syngas produced in the process passed through a quencher for cooling and condensation of steam in excess. Subsequently, the syngas passed through a filter to remove any particle, and through a hydraulic guard, that assured an upstream pressure of 50 millimetres of water column. Then it was quantified through a volumetric counter and analysed by a gas chromatograph. Finally, the syngas was sent to combustion for steam production; the syngas steam in excess was fed to a torch for complete oxidation.

A hydraulic emergency guard, placed between the quencher and the reactor, assured the drainage of the gas produced in the rotary kiln in case of undesired overpressure.

A schematic diagram of the experimental pilot plant is shown in Figure 1.

At the end of each test, the residual RDF in the feed hopper and the water in the quencher tank, were unloaded and weighed in order to evaluate the total mass balance.

After each test the char produced from gasification was unloaded, weighed and characterized. The quencher tank was emptied to quantify the condensed steam. The feed hopper was emptied to evaluate the amount of unprocessed RDF and, therefore, the actual amount of RDF used in the test.
4. Results and discussions

4.1 Experimental results

The experimental RDF was characterized in agreement with UNI TS standards, and the results of the characterization are collected in Table 1. The RDF used in the experiments had a HHV of about 20 MJ kg\textsubscript{sol}^{-1}, mainly due to the high carbon content.

In order to investigate the effects of the steam/RDF feeding ratio on the yield and composition of the syngas, a number of experiments (precisely 3) were performed at constant temperature (850 °C) and solid residence time in the rotary kiln (7 min). These values were determined based on experimental evidence and equations reported in a previous work (Liu and Specht, 2006). The details of the experimental tests performed are given in Table 2.

Figure 2 shows that when the feeding ratio increases, the char yield decreases and the gas yield increases: this is due to the steam gasification reactions between C and H\textsubscript{2}O, as shown in equations (5) and (6) which contribute to the consumption of solid material.

An analysis of the influence of the feeding ratio on the gas composition highlights that when the steam on RDF ratio increases it results in a H\textsubscript{2}, CO\textsubscript{2} and CH\textsubscript{4} content increase and in a decrease in CO, C\textsubscript{2}H\textsubscript{4} and C\textsubscript{2}H\textsubscript{6} content, due to the steam gasification of carbon content species. This behaviour is shown in Table 3, which also reports the high heating value of gas that remains constant when the feeding ratio increases from 0.4 to 2.67.

Table 1: characteristics of RDF

<table>
<thead>
<tr>
<th>Material</th>
<th>Ultimate analysis [% weight]</th>
<th>Proximate analysis [% weight]</th>
<th>HHV [MJ kg\textsubscript{sol}^{-1}]</th>
</tr>
</thead>
<tbody>
<tr>
<td>RDF</td>
<td>C 48.78 H 0.34 N 0.05 S 0.00 O 29.31</td>
<td>Moisture 78.66</td>
<td>Volatile 6.20</td>
</tr>
</tbody>
</table>

Table 2: process parameters in the experimental tests

<table>
<thead>
<tr>
<th>Test #</th>
<th>Gasifying agent</th>
<th>Temperature [°C]</th>
<th>Feeding ratio</th>
<th>Nitrogen flow-rate [Nm\textsuperscript{3} h\textsuperscript{-1}]</th>
<th>Solid residence time [min]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>steam</td>
<td>850</td>
<td>0.4</td>
<td>1</td>
<td>7</td>
</tr>
<tr>
<td>2</td>
<td>steam</td>
<td>850</td>
<td>1</td>
<td>1</td>
<td>7</td>
</tr>
<tr>
<td>3</td>
<td>steam</td>
<td>850</td>
<td>2.67</td>
<td>1</td>
<td>7</td>
</tr>
</tbody>
</table>

Table 3: influence of feeding ratio on gas composition

<table>
<thead>
<tr>
<th>Test #</th>
<th>Temperature [°C]</th>
<th>Feeding ratio</th>
<th>H\textsubscript{2}</th>
<th>CO</th>
<th>CO\textsubscript{2} [% volume]</th>
<th>CH\textsubscript{4}</th>
<th>C\textsubscript{2}H\textsubscript{4}</th>
<th>C\textsubscript{2}H\textsubscript{6}</th>
<th>LHV [MJ kg\textsubscript{gas}^{-1}]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>850</td>
<td>0.4</td>
<td>59.1</td>
<td>16.8</td>
<td>20.1</td>
<td>3</td>
<td>0.8</td>
<td>0.2</td>
<td>16</td>
</tr>
<tr>
<td>2</td>
<td>850</td>
<td>1</td>
<td>61.9</td>
<td>9.9</td>
<td>23.5</td>
<td>4.4</td>
<td>0.3</td>
<td>0</td>
<td>15</td>
</tr>
<tr>
<td>3</td>
<td>850</td>
<td>2.67</td>
<td>66.9</td>
<td>2.2</td>
<td>25.3</td>
<td>5.6</td>
<td>0</td>
<td>0</td>
<td>15.3</td>
</tr>
</tbody>
</table>
5. Conclusion

The results of RDF experimental steam gasification tests in a rotary kiln pilot plant are reported in this paper. The process performance and the gas features were evaluated by varying the feeding ratio (FR) at constant temperature (850 °C) and solid residence time in the rotary kiln (7 min). Three experimental tests were carried out, showing that:

- when the feeding ratio grows, the char yield decreases and the gas yield increases: this trend is due to the steam gasification reactions between C and H₂O which contribute to the consumption of solid material;
- when the feeding ratio increases, the gas energy content has a minimum for FR = 1.0 in conjunction with a lower heating value of 15 MJ kg⁻¹gas⁻¹.
- when the steam on RDF ratio increases, the H₂, CO₂ and CH₄ syngas concentration increases while the CO concentration decreases: this trend is due to steam gasification reactions involving of C and CH₄.

References