Air gasification of paper, cardboard and plastic waste

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Abstract

The research presents the results of paper, cardboard and plastics waste mixture air gasification. The experiments were carry out in a lab-scale rotary kiln reactor. The main operating process parameters were: feeding rate 1 kg/h, operating temperatures 800°C and 900 °C using air as gasifying agent, Equivalent Ratio (ER) ranging between 0.2-0.3. The gas yields increases with the increasing of temperature and gasifying agent (ER=0.3), reaching up to 1.99 m^3N/kg, at 800°C, and 2.1 m^3N/kg, at 900 °C respectively. The syngas composition was analyzed with both Testo instrument and GS-MS. Considering the operating conditions, the main composition of the combustible gas was: N_2 (48-56%), CO_2(13-16%),CO (11-14%), CH_4(5-8%), H_2 (9-10%). The solid yields, decrease with the increase of temperature and ER, varying between 14-17% from the initial feedstock. The minimum energy conversion efficiency is achieved at ER=0.25 while the maximum one is achieved at ER=0.3 for both temperatures.

Keywords: energy, gasification, paper, plastic, waste.

1. Introduction

Currently, 80 million tonnes of Packaging Waste (PW) are generated in the EU-28, where the share of paper, cardboard and plastic waste is 60% (Eurostat, 2014). From this share 19% is plastic waste where 29.7% is recycled, 39.5% is incinerated with energy recovery, and 30.8% is landfilled. However, there are still some countries (e.g. Romania) were the plastic recycling rate is below 15% (Ionescu *et al*., 2015). Furthermore, worldwide, over 60% of the total Plastic Solid Waste (PSW) produced is landfilled (Lettieri and Al-Salem, 2011). Similar situation have been reported for the paper and cardboard packaging waste valorization. Recently the strategy proposed by the EU for the Circular Economy action plan considers a common target for all member states of 75% recycling of packaging waste by 2030 (Circular Economy Action Plan,2015). Low-density polyethylene (LDPE), high-density polyethylene (HDPE), polypropylene (PP) polyethylene terephthalate (PET), polystyrene (PS), and polyvinyl chloride (PVC) are the most common packaging waste. By removing the inert fraction, the packaging waste offer a valuable alternative fuel source due to their high energetic content that varies between 42.77 – 45.78 MJ/kg for plastic packaging waste, 12.42 –15.38 MJ/kg for paper and cardboard packaging waste and 20.1-24.6 MJ/kg for packaging derived fuel (Ionescu *et al*., 2010, Di Gregorio and Zaccariello, 2012). In the last decades, a wide variety of Waste to Energy (WtE) technologies have been proposed where is considered that packaging waste is partly a renewable fuel which is regarded as not contributing to climate change, with high efficiency and high electricity-to-heat ratios where it is replacing fossil fuels (Eriksson and Finnveden, 2009, Panepinto and Genon, 2012, Rada *et al*., 2014, Nessi *et al*., 2015). In 2013, 520 plants were operational in Europe, treating around 95 million tons of Municipal Solid Waste (MSW) and commercial waste per year (Waste Management World, 2013). Incineration is a well-known and reliable type of process, with defined design and operating criteria. Due to the nature of the process, various green gas emission, such as carbon monoxide, carbon dioxide and nitrogen oxides are released into the atmosphere, which implies investments and high operating costs. Furthermore, in order to avoid corrosion caused by the waste high chlorine content the most MSW incineration plants are operated at lower steam conditions (generally at 400 °C and 40 bar), therefore the power efficiency will not succeed 22-25% (Murphy and McKeogh, 2004). As alternative to incineration, advanced thermochemical approaches, such as pyrolysis and gasification have been intensively studied in the WtE sector and have been considered effective, efficient and environmental-friendly technologies. Gasification, or ‘‘indirect combustion’’, is the thermochemical conversion of the raw material (solid or liquid) in the presence of substoichiometric air/oxygen or water vapors, which converts volatile solids (VS) into a combustible gas (syngas). The syngas is mainly composed of carbon monoxide (CO) and hydrogen (H_2). Due to the waste composition, which also contains other elements, in addition to carbon and hydrogen, such as oxygen, nitrogen, chlorine, sulfur, etc., and operational conditions, the syngas can contain carbon dioxide (CO_2), nitrogen (N_2), nitrogen oxides (NO_x), light hydrocarbons (C_nH_m). The transformation of the solid waste is divided into four stages: drying, pyrolysis, incomplete combustion, and water reduction. In comparison with traditional waste incineration, gasification has some potential advantages given by the operation conditions (mainly temperature,

equivalent or steam ratio) and reactors (fixed bed, fluidized bed, entrained bed, vertical shaft, moving grate furnace, rotary kiln, plasma reactor) making the syngas suitable for different application (combustion in traditional burners, connected to a boiler and a steam turbine, gas reciprocating engines or gas turbines or its conversion in valuable products as chemicals and fuels due to CO and H_2 content) (Arena, 2012). Over more, the syngas can be converted into liquid fuels or used for electricity and/or heat production, reaching up to a performance production of 34%, 40% respectively (Wilk and Hofbauer, 2013, Liu et. al, 2016). The WtE tendencies towards thermal treatment with partial oxidation, over traditional combustion is sustained by the absence of dioxins and furans and significant energy loss with nitrogen sensitive heat in the flue-gas (Marculescu, 2011, Mărculescu *et al*., 2016).

2. Material and methods

The origin of the packaging waste samples came from the household source separate collection of Bucharest Municipality, Romania. Parts of the current experiments were carry out during the development of a PhD thesis in co-tutela (Ionescu,2012). In the first stage the materials were hand sorted by fraction in order to delimit their qualitative and quantitative characteristics. During this operation, other liquids (e.g. traces of fluids from the plastic containers) or solids (e.g. organic matter such as food) residues were removed from the process. At the end the feedstock analysed in this study consisted on a 1:1 mixed ratio of HDPE, PET, PP, paper and cardboard packaging waste. In order to improve the handling characteristics, homogeneity of the material and the gasification power plant industrial condition the feedstock was reduced in size by using a Cutting mill Fritsch (Pulverisette 15). The methods used for the determination of the proximate and ultimate analysis and heating value of the materials are described and presented in a previews research conducted by the authors Ionescu *et al*., 2010. The experiments were performed in a gasifier lab-scale pilot plant located The Faculty of Power Engineering, University Politehnica of Bucharest. The lab-scale plant is aimed for the study of different solid and liquid fuels, especially waste as an energy source, for their thermochemical conversion in a free, partial or complete oxidant atmosphere, considering the mass, energetic and environmental outlooks. The gasifier lab-scale pilot plant is a rotary kiln reactor with external heating that has a processing capacity of 30 kg/h, an overall volume of about 0.008 m^3 and an energy consumption of 15kW/h. The simplified scheme of the lab-scale rotary kiln reactor is presented in Figure 1.

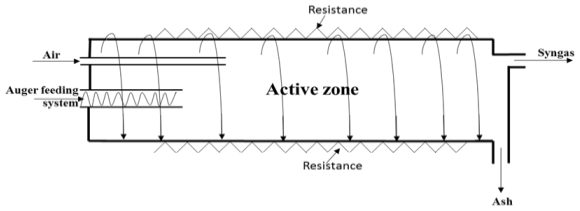

Figure 1. The simplified scheme of the lab-scale rotary kiln reactor

The reactor allows the variation of its speed ratio, along with its angle inclination that can vary between 0-20º. The kiln is provided with a feed hopper in the form of a screw conveyor charging. Therefore the speed of the feed rate can be controlled by a frequency electronic controller. In the current experiments the feeding rate was determinate by achieving several try-outs on the feed hopper only, considering the proposed conditions operations and feedstock characteristics. The operating temperature in continuous mode can reach up to 900ºC, going up to 1200 ºC for a short operating time. Several temperature sensors are installed in the central heating area in order to monitor and control the gasification process. The reactor furnace has three nozzles: two for the gaseous species (inlets and outlets) and one at the lower end of the tube where due the rotation and gravity the solid sub-product is discharge. A rotameter is used for the gas flow input and control of the process. In order to verify the gasification process conditions and process stability, in the first phase of the trials the gaseous species were analysed with a TESTO 350 M / XL exhaust gas analyser. This advanced equipment allows the determination of SO_2, CO, CO_2, C_nH_m/H_2, O_2, NO and NO_2 concentrations, gas flow velocity and mass flow rate performed at short intervals of time. Consecutively the Gas Chromatography – Mass Spectrometer (GC-MS SCHIMADZU QP 210 Plus) unit was used for the gas composition analysis. It was estimated that the syngas is analysed by the GS-MS unit, after 50 up to 60 seconds, from the moment of gas extraction until the being of its analysis. The latter observation had led to the integration of the TESTO 350 M / XL gas analyser in the determination syngas composition. In the current study, packaging waste gasification was performed by using air as gasifying agent. In order to define the necessary air-fuel ratio of the feedstock partial oxidation the Equivalent Ratio (ER) was obtained. In the present study, firstly the determination of the minimum amount of necessary air for paper, cardboard and plastics waste mixture complete combustion was determinate considering: the material in its dry basis, the CO and NO_x are not formed, sulphur is oxidized until the formation of SO_2, the excess of air is considered 1.3 and the relative humidity of wet air. Over more the calculations were made based on the elemental composition of the materials where dedicated formulas were used for the determination of the stoichiometric air-fuel ratio for complete combustion. Considering a comprehensive literature review and the syngas composition at the chemical equilibrium as a function of equivalence ratio a 0.2-0.3 ER was chosen (Kim *et al*., 2011, Arena, 2012). The main operating gasification test conditions of the paper, cardboard and plastic waste mixture (Table 1). In the experiments the composition of the syngas was determined considering CO, H_2, CH_4, C_nH_m, N_2 in volume fraction. The syngas lower heating value (LHV) is calculated with equation (1) (Zhao *et al*., 2010) :

$$LHV = (CO \times 30 + H_2 \times 25.7 + CH_4 \times 385.4 + C_nH_m \times 151.3) + 4.2 [kJ/Nm^3] \quad (1)$$

The Energy Conversion Efficiency (ECE), can be defined as the net energy of the syngas to net energy content of the input fuel (equation 2):

$$ECE = \frac{Q_{syngas} \times LHV_{syngas}}{Q_{fuel} \times LHV_{fuel}} \qquad (2)$$

where: Q_{syngas} , Q_{fuel} are the syngas flow rate, and feed rate respectively and LHV_{syngas} , LHV_{fuel} are the lower heating values of the syngas and packaging waste mixture.

Table 1. Operating parameters

Feed size (mm)	0.5-5
Process temperature (°C)	800-900
Air inlet temperature (°C)	400-500
The fuel feed rate (kg/h)	1-1.2
Air-fuel ratio (Nm^3 /min)	0.07-0.10
Equivalence ratio	0.2–0.3

3. Results and discussion

In accordance with the experimental methodology, the influence of the temperature at 800°C and 900°C, by varying the ER=0.2-0.3 over the syngas composition is presented in Figures 2-4. The syngas composition was analyzed with both Testo instrument and GS-MS.

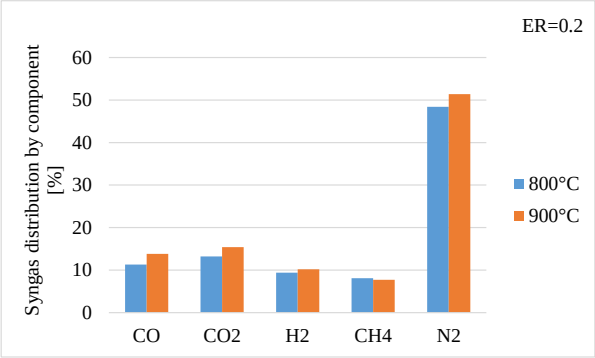

Figure 2. The influence of temperature at 800°C and 900°C, and constant ER=0.2 over the syngas composition

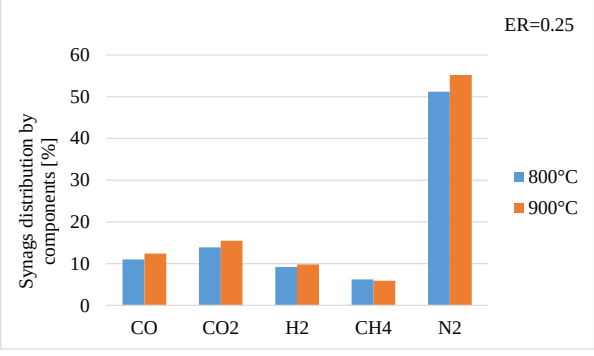

Figure 3. The influence of temperature at 800°C and 900°C, and constant ER=0.25 over the syngas composition

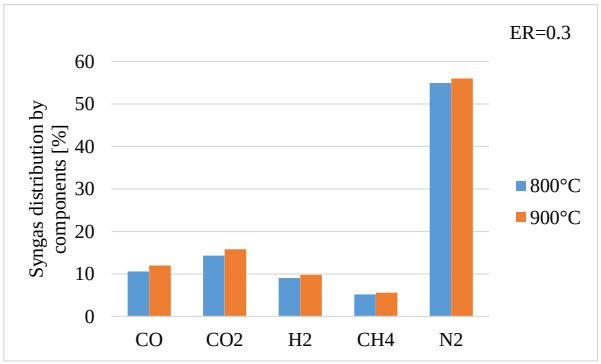

Figure 4. The influence of temperature at 800°C and 900°C, and constant ER=0.3 over the syngas composition

The combustible gas contains CO_2, CO, H_2, CH_4, N_2, and trace amounts of higher hydrocarbons, various contaminants such as small char particles, ash and tars. Considering the operating conditions and the results presented in Figures 2-4, the main composition of the combustible gas was: N_2 (48-56%), CO_2 (13-16%), CO (11-14%); CH_4 (5-8%), H_2 (9-10%). It is notice that with the increase of the temperature the CO_2 breaks down to form CO. This can be explained by the O_2 reaction with carbon to form CO and CO_2 which is more powerful in comparison with hydrogen for water formation. As it was expected and reported in previews studies on gasification of plastic or biomass Cho *et al.*, 2013, Fu *et al.*, 2014, the gas yield expands with the increasing of temperature and gasifying agent, while the LHVs reduces as ER increases, as presented in Figure 5. The latter can be explained by the notable reduction of the energetic hydrocarbons such as methane CH_4, ethylene C_2H_4, acetylene C_2H_2 with the complementary increase of H_2 and CO, therefore conducting to the partial oxidation of the syngas, consecutively the decrease of syngas energetic content. Over more due the ER diminution leads to the incomplete conversion of char and possibly to the increase of tar yield. During the experiments at maximum ER=0.3 the gas yield reaches out near to 2 m^3N/kg and 2.1 m^3N/kg, at 800°C and 900°C respectively. The syngas LHV was determinate based on the gas produced composition (Figure 5) and calculated with Equation 1.

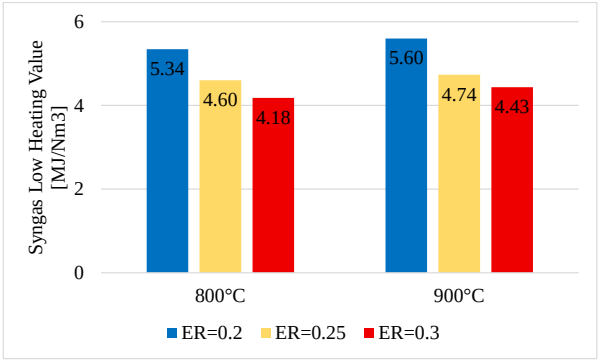

Figure 5. Syngas Low Heating Value influence by temperature and ER.

The application of the formula and accuracy of the results is influenced by the lack of experimental validated results and the absence of data on hydrocarbons amounts like

C_2H_2, C_2H_4, C_2H_6. However, similar results were registered in other studies conducted on biomass and plastic gasification by fraction, at temperatures of 850°C -890°C and an equivalent ratio of 0.21-0.24, where the biomass LHV will reach up 8.84 MJ/$kg_{biomass}$, while plastics 7.4-11.4 MJ/Nm^3 (Xiao *et al*., 2007 Alauddin *et al*., 2010).

The minimum ECE is achieved at ER=0.25 while the maximum one is achieved at ER=0.3 for both temperatures as presented in Figure 6.

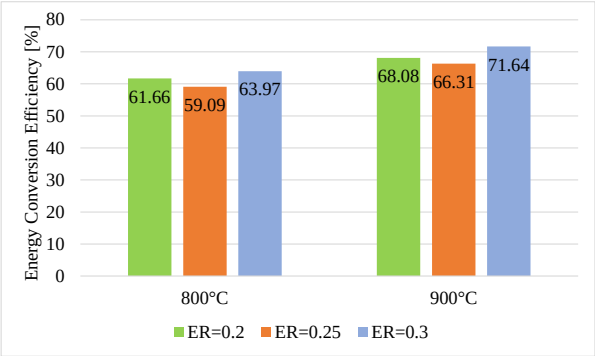

Figure 6. Energy conversion efficiency [%]

Even thou one of the main technological impediment that needs to overcome with gasification is tar, no significant amounts were registered during the experiments. Generally the amount of tar produced during the plastic waste gasification is higher in comparison with biomass due to the higher amount of the plastics volatile matter. The solid residue amount is strongly influenced by temperature and ER, varying between 16%-17% at 800°C and 14%-15% at 900°C, as presented in Figure 7. The rest of yields distribution is represented by the amount of the syngas, since no tar yield were determined or registered during the process.

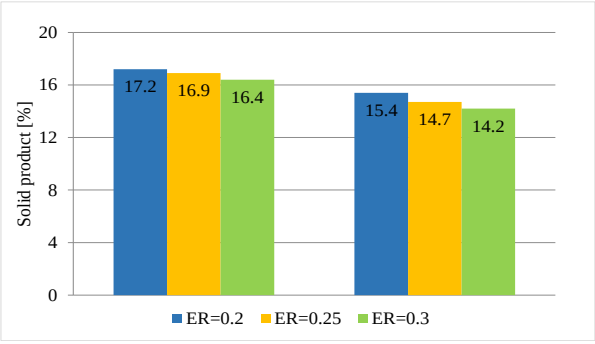

Figure 7. Solid product mass distribution

Correlating the data from Figures 2-4 and 7 the char conversion is lower at 800°C.

4. Conclusions

The gasification of paper, cardboard and plastic packaging waste mixture was investigated in a rotary kiln reactor. Correlating the results obtained by the authors in a preview study Ionescu *et al*., 2012 on the pyrolysis on the same waste, we can conclude that the gaseous and solid products distribution are influenced by the plastic and paper behavior regarding the thermal cracking of each waste fraction. The gas yield increase with the increasing of temperature and gasifying agent. Considering the operating conditions, the main composition of the combustible gas presented in Figures 2-4 was: N_2 (48-56%), CO_2(13-16%),CO (11-14%); CH_4(5-8%), H_2 (9-10%).Without considering the C_nH_m hydrocarbons except CH_4, in the present experiments the gas LHV will reach to its maximum at 5.6 MJ/ Nm^3 at 900°C at the minimum ER used in the experiments of 0.2. The solid yields, decrease with the increase of temperature and ER, varying between 14-17% from the initial feedstock.

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