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Refereed Proceedings

2013

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STEAM GASIFICATION OF WASTE PLASTICS IN A CONICAL SPOUTED BED REACTOR

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ABSTRACT
The effect of temperature (in the 800 to 900 ºC range) and steam/plastic mass ratio (between 0 and 2) on the products of steam gasification of high density polyethylene was studied. The spouted bed reactor has an excellent performance between 850 and 900 ºC. The increase of steam/plastic ratio from 1 to 2 only improves slightly the results.

KEYWORDS: Spouted bed; steam gasification; waste plastics; hydrogen; syn-gas; waste management; plastics gasification

INTRODUCTION
The use of different kinds of plastics has greatly increased in recent years, and the disposal of plastic waste has become a major environmental issue. Consequently, the development of recycling processes at industrial scale is urgent to avoid the environmental problems related to waste plastic landfill and contribute to intensifying the upgrading of oil, of which 8% is used in the production of plastics (1).

Pyrolysis is an interesting option for the treatment of polyolefins to obtain fuels and monomers (2,3). Gasification is another alternative, given that it is a flexible process that allows operating with a wide range of plastics, mixed plastics and even combining different wastes, such as biomass and coal. Furthermore, depending on the gasification agent used, it is possible to direct the process towards the production of a gas fuel with low (using air) or high (using pure oxygen) heating value or a hydrogen rich syngas (using steam). The syngas produced is suitable for energy conversion systems such as combined cycle turbines and engines, thus improving electricity generation efficiency and avoiding the emission of harmful and toxic gases formed in the combustion of waste plastics.

Different technologies have been reported in the literature for waste plastic gasification, with the gasifying agent being air or steam and reforming catalysts been used to increase H_2 yield. Fluidized bed reactors have been widely used for the gasification of waste plastics, given that they are isothermal, versatile concerning operating parameters and can operate with continuous feed (4-6).

The conical spouted bed reactor has been successfully applied to the pyrolysis of different waste plastics and other residues whose treatment involves difficulties in fluidised bed reactors due to agglomeration and defluidization problems (7). Moreover, the steam gasification process is highly endothermic and, consequently, a considerable heat flow must be continuously supplied to the reactor in order to sustain the reforming reactions. The high heat transfer rates (8) together with the vigorous solid flow pattern characteristic of spouted beds ensure almost immediate melting, sand particle coating with fused plastic, pyrolysis and subsequent gasification of the plastic, thus minimizing the formation of undesired tar and solid residue. Furthermore, the conical spouted bed is of easy design, given that it does not require a distributor plate, has low pressure drop and its throughput by reactor volume unit is much higher than that of fluidized beds due to the lower amount of sand required for fluidization. Furthermore, the low segregation is another interesting feature of conical spouted beds for the operation with catalysts in situ for tar cracking or reforming. Moreover, the conical spouted bed reactor is suitable for continuous operation, which is especially interesting for the implementation of the large-scale process. Accordingly, improvements were carried out, such as the use of internal devices, to increase bed stability (controlled spout geometry and fountain height) and reduce gas flow rate requirements (9). This paper addresses the steam gasification of HDPE in a bench-scale plant provided with a conical spouted bed reactor. Studies was carried out on the effect of temperature and steam/plastic ratio.

EXPERIMENTAL SECTION

HDPE properties

The high density polyethylene (HDPE) was provided by Dow Chemical (Tarragona, Spain) in the form of chippings (4 mm), with the following properties: average molecular weight, 46.2 kg mol^{-1}; polydispersity, 2.89; and density, 940 kg m^{-3}. The higher heating value, 43 MJ kg^{-1}, was determined by differential scanning calorimetry (Setaram TG-DSC-111) and isoperibolic bomb calorimetry (Parr 1356).

Equipment

Steam gasification runs were carried out in a bench scale plant whose scheme is shown in Figure 1. The main element of the plant is the conical spouted bed reactor whose design is based on previous hydrodynamic studies (10) and on the application of this technology to the pyrolysis of different solid wastes, such as biomass (11), plastics (7) and scrap tires (12).

The reactor is located within an oven which is in turn placed in a forced convection oven maintained at 270 ºC to avoid the condensation of steam and tars before both the analysis and condensation systems. This forced convention oven contains a high-efficiency cyclone and a sintered steel filter (5 μm) for retaining the fine sand particles entrained from the bed and the soot or char particles formed in the gasification process.

The plant is provided with a system for feeding plastic, which allows operating under continuous regime. The system for solid feeding consists of a vessel equipped with a vertical shaft connected to a piston placed below the material bed. The plastic is fed into the reactor by raising the piston at the same time as the whole system is vibrated by an electric engine. A very small nitrogen flow rate introduced into the vessel stops the volatile stream entering the feeding vessel. The plastic feed rate can be varied from 0.2 to 5 g min^{-1}. The pipe that connects the feeding system with the reactor is cooled with tap water to avoid the plastic melting and blocking the system.

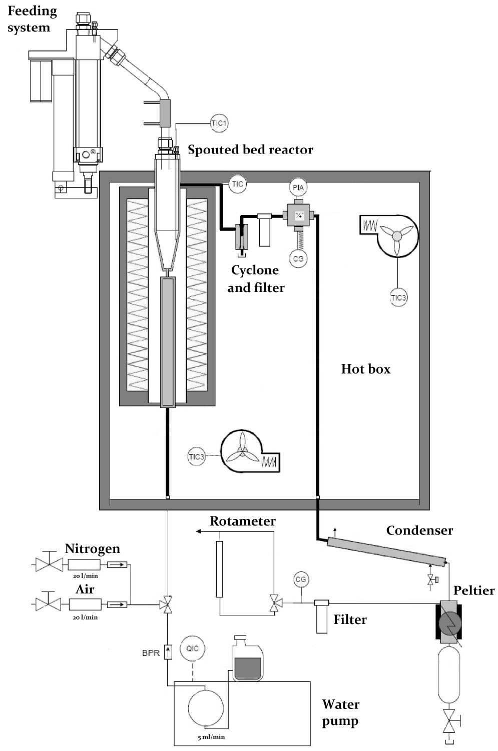

Figure 1. Scheme of the laboratory scale plant.

Water was fed by means of a Gibson 307 pump that allows a precise measuring of the flow rate. The water stream was vaporized by means of an electric cartridge (not plotted in Figure 1) placed inside the forced convection oven and prior to the entrance of the reactor. Moreover, N_2 is used as fluidizing agent during the heating process and its flow rate is controlled by a mass flow controller that allows feeding up to 20 L min^{-1}. The plant is also equipped with an additional mass flow meter (20 L min^{-1}) for measuring air flow rate.

The gases leaving the forced convection oven circulate through a volatile condensation system consisting of a condenser, a Peltier cooler and a coalescence filter. The condenser is a double-shell tube cooled by tap water. The coalescence filter and the Peltier cooler ensure the total condensation and retention of the tars.

Experimental conditions

The conical spouted bed reactor contains 70 g of sand (particle diameter in the 0.35-0.4 mm range). Water flow rate is 1.5 mL min^{-1} in all the studied conditions, corresponding to a steam flow rate of 1.86 L min^{-1}, which is approximately 1.5 times that corresponding to the minimum spouting velocity. The runs were conducted in continuous regime by feeding 1.5 g min^{-1} of HDPE and using a steam/plastic ratio equal to 1. In the runs to study a steam/plastic ratio of 2, the plastic feed rate was reduced to 0.75 g min^{-1} in order to maintain the same steam flow rate (1.5 mL min^{-1}). Consequently, the residence time of the products in the reactor and the hydrodynamic performance are similar, which allows comparing the results under different steam/plastic ratios. The performance of the operation without steam was also studied by replacing the steam by a nitrogen flow rate of 2 L min^{-1}. The effect of temperature on gasification was studied in the 800-900 ºC range and the effect of steam/plastic ratio at 900ºC.

All the runs were performed in continuous mode for at least 20 min in order to ensure the steady state of the process. Moreover, the runs were repeated several times (at least 3) under the same conditions in order to guarantee reproducibility of the results. The amount of char formed was determined by weighting the mass retained in the cyclone and sintered steel filter, and its yield by also monitoring the amount of plastic fed into the reactor in each run.

The volatile stream leaving the gasification reactor were analysed on-line by means of a GC Agilent 6890 provided with and HP-Pona column and a flame ionization detector (FID). The sample was injected into the GC by means of a line thermostated at 280 ºC, once the reactor outlet stream was diluted with an inert gas. The purpose of this system is to avoid the condensation of tars in the transfer line.

The non-condensable gases were injected into a micro GC (Varian 4900). The identification of the tars collected in the condensation system was carried out in a gas chromatograph/mass spectrometer (GC/MS, Shimadzu UP-2010S provided with a HP-Pona column).

RESULTS

Effect of gasification temperature

The effect of temperature on steam gasification was studied in the 800 to 900 ºC range using a steam/plastic ratio of 1. Figure 2a shows the effect of temperature on the yields (in terms of weight percentage of the plastic in the feed) for the different fractions (gas, tar and char). The weight percentage of the water reacted was also plotted, which was calculated from the oxygen content on the product stream. The gas fraction is made up of H_2, CO and CO_2, together with C_5- (mainly C_3-) hydrocarbons. The tar (C_6+) is made of aromatic hydrocarbons, with benzene being the lighter one. The char (a marginal product) is a carbonaceous material and is retained in the sintered steel filter and in the cyclone.

As observed in Figure 2a, an increase in temperature improves the efficiency of the process, i.e., increases the gas yield and reduces the yields of both tar and solid residue. The yield of the gas fraction increases from 148.1 g/100 g of plastic operating at 800 ºC to 178.7 g/100 g of plastic at 900 ºC.

The main byproduct formed is the tar, whose yield decreases with temperature from 8.9 g/100 g plastic at 800 ºC to 6.0 g/100 g plastic at 900 º due to the enhancement of thermal cracking. These tar yields correspond to a concentration in the gas stream of 29.5 and 16.7 g $(Nm^3)^{-1}$, respectively.

In order to compare the results in this paper with those reported in the literature, differences in reactors, conditions and materials should be taken into account. Furthermore, some authors do not include benzene in the tar fraction and, in our results, benzene accounts for 65 to 75 % of the tar yield. Arena et al. obtained very high tar yields, up to 40 % wt, in the 800 to 900 ºC temperature range, using air and air/steam mixtures as gasifying agent and operating with sand fluidized beds (5). Kim et al. also operated with sand fluidized beds and they report tar yields in air gasification of up to 17 % wt at 806 ºC (13). The yields obtained by Xiao et al. in the air gasification of polypropylene are of the same order as those in this study and, moreover, they conclude that tar yield is highly dependent on the equivalence ratio (14). Similarly, an increase in gasification temperature reduces the yield of char, which is recovered as a fine powder in the cyclone and the sintered steel filter placed downstream the reactor (see Figure 1), from 1.41 % at 800 ºC to 0.45 % at 900 ºC.

Figure 2b shows the effect of temperature on the composition of the gases. An increase in temperature gives way to an increase in the concentrations of H_2, CO and CH_4 in the gaseous stream, which are 60.3, 28.2 and 7.2 % vol., respectively, at 900 ºC. Temperature has an opposite effect on the remaining gaseous products, i.e., CO_2 and C_2-C_5 hydrocarbons (made up mainly of olefins, with ethylene being the prevailing one). The high concentration of H_2 (slightly higher than 60 %) obtained at both 850 and 900 ºC corresponds to the yields of 17.3 and 18.4 g of H_2 for 100 g of HDPE in the feed. The aforementioned results are excellent compared to those in the literature. The H_2 concentrations obtained in the steam gasification with sand in a fluidized bed by Wu and Williams are in the 20-50 % range, and only when using reforming catalysts they obtained H_2 concentration between 60 and 70% (15). Likewise, the H_2 concentration obtained by He et al. in the steam gasification of polyethylene is below that in this study, even when they used a NiO/Al_2O_3 catalyst (16). Namioka et al. obtained concentrations in the 65-70 % range, but they only studied the steam gasification of polystyrene with a ruthenium based catalyst (17).

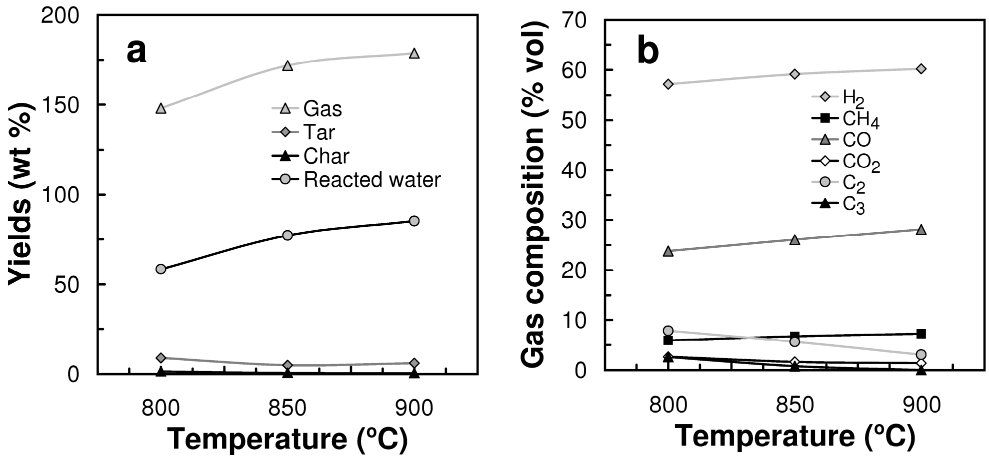

Figure 2 Effect of gasification temperature on product yields for 100 g of plastic fed into the reactor (Figure 2a) and on gas fraction composition (Figure 2b).

Effect of steam/plastic ratio

The effect of steam/plastic mass ratio (S/P) was studied at 900 ºC by varying this parameter between 0 (using N2 as fluidizing agent) and 2. Figure 3a shows the effect of S/P ratio on the yields of gas, tar and char fractions, and on the percentage of water reacted. An increase in S/P ratio from 0 to 2 increases carbon conversion from 91.0 % to 93.6 %. It is noteworthy that performance is poor when operating with a S/P = 0 (pyrolysis), given that the high yields of both tar and char. The lack of steam in the reactor at high temperatures favours the formation of aromatic compounds, thus increasing the tar yield to values as high as 19 %. Accordingly, an increase in S/P ratio favours the cracking of tar compounds.

The presence of steam in the reaction environment also attenuates the char formation, with char yield being 10 times lower when operating with a S/P = 1 than operating without steam.

The gas yield by operating with S/P = 2 (188 g per 100 g of plastic) is only slightly higher than that obtained with S/P = 1 (179 g per 100 g of plastic), and is due to the following aspects: (i) enhancement of hydrocarbon reforming reactions due to the higher steam concentration in the reaction environment, and; (ii) lower tar and char formation rate, although this effect is of lower significance.

As mentioned previously for the yields of the different fractions, an increase in S/P ratio from 1 to 2 does not give way to a significant change in gas composition (Figure 3b). Nevertheless, there is a great difference between the gas fraction composition obtained operating with steam or nitrogen. An increase in S/P ratio favours H_2 and CO_2 formation but reduces that of CO and CH_4 because the higher concentration of steam in the reactor enhances both water gas shift and methane reforming reactions.

In the runs (pyrolysis) performed without steam (S/P = 0), the gas is made up of H_2 (28.7 % vol.), CH_4 (28.6%), ethylene (35.4 %) and other light olefins (propylene and butenes).

An aspect of great significance observed at 900 ºC and S/P = 2 is the high H_2/CO ratio (2.2) in the syngas together with a low concentration of CO_2, hydrocarbons and polyaromatic tar. Accordingly, this syngas is especially suitable for use as raw material in catalytic processes for the synthesis of hydrocarbons (Fischer-Tropsch) or DME.

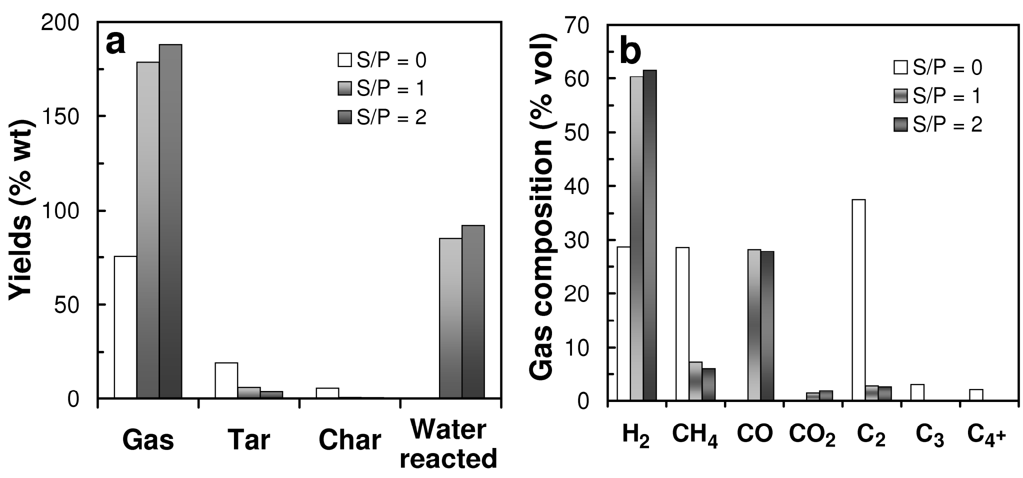

Figure 3 Effect of S/P ratio on product yields for 100 g of plastic fed into the reactor (Figure 3a) and on gas fraction composition (Figure 3b).

CONCLUSIONS

The conical spouted bed reactor is an interesting technology for the continuous steam gasification of waste plastics due to the high heat transfer rates provided by these beds for a highly endothermic process as gasification. An increase in the gasification temperature from 800 to 850 ºC has a significant positive effect on gas yield, H_2 content and tar formation, but a subsequent increase to 900 ºC only improves the process performance slightly. Between 850 and 900 ºC the hydrogen concentration in the syngas produced is above 60 %.

An increase in steam/plastic ratio produces a positive effect, and operating at 900 ºC with a steam/plastic ratio of 2, the H_2 concentration is of 61.6 %, with minimum tar formation. The syngas obtained has a H_2/CO ratio of 2.2, which is suitable for catalytic processes aimed at the synthesis of hydrocarbons or DME. A low content of hydrocarbons (mainly CH_4 and ethylene) is an inconvenience for these processes but it can be solved by using a reforming catalyst. Furthermore, the results obtained without steam (steam/plastic ratio of 0) are not satisfactory due to the high tar and char yields.

A low tar yield (6 %) is obtained in the steam gasification of HDPE, with its composition being made up of mainly monoaromatics (65-75 % benzene) and its concentration being minimum at 900 ºC with a steam/plastic ratio of 2.

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