Gasification Of A Plastic Waste In A Pilot Fluidized Bed Reactor

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A recycled polyethylene was fed in a pilot plant bubbling fluidized bed gasifier, having a maximum feeding capacity of 60kg/h. The experimental runs were carried out under various operating conditions: the bed temperature was kept at about 850°C, the equivalence ratio varied between 0.2 and 0.35, the fluidizing velocity between 0.5 and 0.7m/s, quartz sand and olivine were used as bed material while air and steam were used as fluidizing reactants.
The results confirm the flexibility of fluidized bed gasification process that can produce syngas of remarkable different composition by varying the operating conditions. Extremely interesting is the possibility to obtain a strong reduction of tar content by using a catalyst as active bed additive inside the gasifier itself during the process.

1. Introduction

The world needs more sustainable waste management techniques, which focus on greater value recovery from waste and easier plant acceptance by the interested people. In the last decade, a number of novel technologies utilizing gasification processes have emerged to address these issues and to improve the value of energy or materials outputs (Livingston, 2002; Malkow, 2004; Klein et al., 2004; Arena and Mastellone, 2005). Their interesting potential is mainly related to the possibility of combining the type of starting waste, the operating conditions and the features of the specific reactor in order to obtain a syngas that can be utilized in different applications. The range of products immediately obtainable from syngas extends from bulk chemicals like ammonia and methanol, through industrial gases, to utilities such as clean fuel gas and electricity (Higman and van der Burgt, 2003).

Among all waste gasification technologies, fluidization is one of the promising ones, for a series of attracting reasons. In particular, the great operating flexibility makes possible to utilize different fluidizing agents, reactor temperatures and gas residence times, to add reagents along the reactor freeboard or riser and to operate with or without a specific catalyst (Arena and Mastellone, 2005; 2006; Basu, 2006).

On the other hand, there is the main disadvantage of the risk associated with a technology less proven in operation than conventional combustion technologies, particularly taking in mind the highly heterogeneous nature of feeds like municipal solid wastes. As a consequence, the economics are far away from clear: there still is a lack of reliable assessments on the scale and operating conditions under which a full economic

convenience of the fluidized bed gasification of wastes is obtainable. In this framework, CONAI, the Italian National Consortium for Packagings, funded a three-years joint research program focused on fluidized bed gasification of several alternative fuels. The aim is to cover some of the areas of technical uncertainty by utilizing a pilot plant fluidized bed reactor, operated under various operating conditions, with various gasification agents and with wastes and alternative fuels of high interest (packaging derived fuels, pulper residues, refuse derived fuels).

The paper presents the first results obtained by feeding the pilot gasifier with a recycled polyethylene, under various conditions of equivalence ratio, fluidizing velocity and bed material type.

2. The Bubbling Fluidized Bed Gasifier and the utilized materials

The pilot scale BFBG has the design and operating features schematically listed in Table 1. It is composed of three main sections (the feeding system, the fluidized bed gasifier and the syngas treatment unit), as it is shown by the quantified process flow-sheet reported in Fig. 1. The feeding system can be divided in the blast feeding

Table 1. Main design and operating features of the bubbling fluidized bed gasifier.

Geometrical parameters	ID: 0.381m; Total height: 5.90m; Reactive zone height: 4.64m; Wall thickness:12.7mm
Feedstock capacity	30-60kg/h
Typical bed amount	140kg
Feeding equipments	In-bed (water cooled) and over-bed (air cooled) screw feeders
Gasifying agents	Air, oxygen, steam, carbon dioxide (alone or as mixture)
Range of operating temperatures	700-950°C
Range of fluidizing velocities	0.5-1m/s
Safety equipments	Water seal, safety valves, rupture disks, alarms, nitrogen line for safety inerting
Mean process variables	Reactor temperature; bed height; fluidizing velocity; blast flow rate; equivalence ratio

(measuring, mixing and injection of gasification agents) and the fuel feeding (measuring and injection of solid feedstock). The blast feeding is heated up to 200°C by an electric heater, then sent to a mixing point with a stream of steam at about 150°C and finally heated by a second electric heater up to 600°C before entering the reactor. The blast can be supplied at different levels of the gasifier: at the bottom, as fluidizing gas and primary blast, and at two freeboard heights, as secondary blast. The fuel can be fed in-bed or over-bed, by means of two separate feeding systems. The fuel and the blast flow rates are mutually adjusted so that, at the fixed fluidizing velocity, the desired equivalence ratio (ER, defined as the ratio of actual air fuel ratio to the stoichiometric air fuel ratio) is obtained. The gasification section is composed of a cylindrical BFB reactor, equipped with several control and protection devices, which is heated up to the reaction temperature thanks to the sensible heat of pre-heated blast gases and to a set of three external electrical furnaces. The gas generated in the reactor is sent to the syngas

treatment section composed of a high efficiency cyclone, a wet scrubber (for removal of tars, residual fly ashes and acid gases) and a flare. A more accurate description of the plant can be found in Arena et al. (2006).

The BFBG was fed with a recycled polyethylene, coming from separate collection of municipal solid wastes. Two types of bed materials were used during the experimental runs: a quartz sand, having a size range of 0.2-0.4mm and a particle density of 2600kg/m^3, and an olivine, having the same size range and a particle density of

Table 2. Chemical composition of the olivine used as bed material.

MgO	48.0-50.0
SiO$_2$	39.0-42.0
Fe$_2$O$_3$	8.0-10.5
CaO	<0.4
Al$_2$O$_3$+ Cr$_2$O$_3$+ Mg$_3$O$_4$	0.8

3100kg/m^3. The olivine chemical composition is reported in Table 2 (Magnolithe Ltd, 2006).
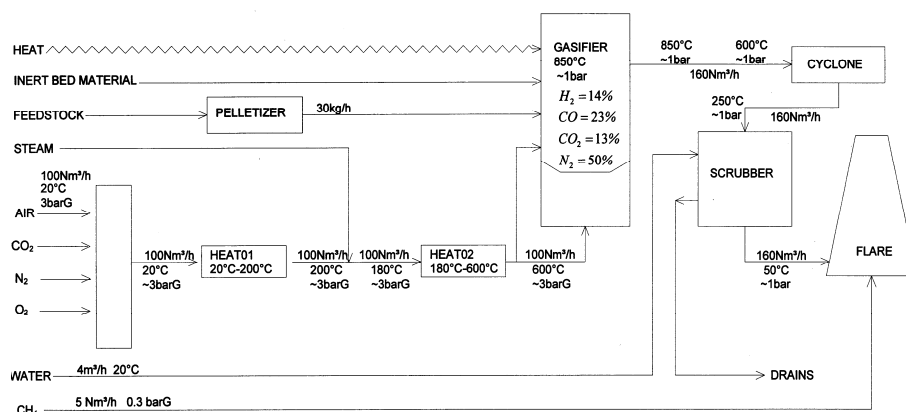

Figure 1 Process flow sheet of the gasification pilot plant.

3. Experimental results and discussion

The pilot scale BFBG was operated under various operating conditions: the equivalence ratio varied between 0.2 and 0.35, the fluidizing velocities between 0.5 and 0.7m/s while the bed temperature was kept at about 850°C and air or an air/steam mixture were used as fluidizing reactants. The utilization of olivine as active bed additive inside the gasifier was suggested by the relevance of tar formation problem. Tar is a complex mixture of a large spectrum of condensable hydrocarbons, which includes single ring to multiple ring aromatic compounds along with other oxygen containing hydrocarbons. Tar is undesirable because of various problems associated with condensation, formation of tar aerosols and polymerization of complex structures, which cause problems in the process equipments as well as the devices for end-use application (Dayton, 2002; Devi et al., 2003). It is evident from results reported in Table 3 that in all the experiments carried out using quartz sand as bed inert material, the average tar concentration in the exit gas is very high, i.e. of the order of 100g/m^3_N while the required tar and dust loads in gases must be lower than 10mg/m^3_N for gas engines and turbines and one or two orders of magnitude lower for catalytic conversion processes (Devi et al., 2005).

Tar removal technologies can substantially be divided in treatments inside the gasifier (primary methods) and hot gas cleaning after the gasifier (secondary methods).

Secondary methods (thermal or catalytic tar cracking downstream the gasifier and mechanical methods such as use of cyclone, ceramic filter, fabric or electrostatic filter, and scrubber) are greatly promising and in several cases proven to be effective. On the other hand, treatments inside the gasifier (adequate selection of operating parameters, use of a proper bed additive or catalyst, gasifier design modifications) are gaining much attention for solid waste gasification since they may eliminate the need for downstream cleanup. It is likely that an adequate combination of different primary treatments may optimize the gasifier performance and allow to produce a syngas with minimum tar concentration.

In this part of research project, preliminary indications about the role of some operating parameters (reactor temperature, gasifying agent, equivalence ratio, residence time) in the formation and decomposition of tar were obtained. It is confirmed, for instance, that longer is the residence time less is the residual tar amount (runs 5, 4 and 3 in Table 3) and that higher is the temperature lower is the tar content in the producer gas (runs 7 and 5). Moreover, tar yield decreases (from 12.1 to 7kg/h) as the equivalence ratio ER increases (from 0.22 to 0.31), as a consequence of the larger oxygen amount that can react with volatiles in the pyrolysis zone (runs 5 and 6). It is also interesting that CH_4 concentration results an indicator of the amount of heavier hydrocarbons in the producer gas: higher is its value larger is the C_nH_m and tar yield. The addition of steam, with a flow rate of $0.5kg/kg_{fuel}$, leads to a reduction of tar yield (runs C and D compared with runs 7 and 5, respectively) as the consequence of steam reforming reaction of heavier hydrocarbons: $C_nH_m + n\ H_2O \leftrightarrow nCO + (n + {}^m/_2)\ H_2$.

Very interesting appears the potential of using an active bed additive such as calcined olivine inside the gasifier. This additive was chosen on the basis of its excellent performances demonstrated in the operation of fluidized bed biomass gasifier (Pfeifer et al., 2004; Devi et al., 2005). Experimental runs from 8 to 13 in Table 3 utilized a bed completely made of olivine. The first run (#8) already gave a remarkable decrease (85%, i.e. from $100g/m^3_N$ to $14.5g/m^3_N$) in tar yield, compared with operation under the same operating conditions but with a bed of quartz sand (#4). The performance greatly

Table 3. Operating conditions and main results of experiments carried out with PE in the pilot plant bubbling fluidized bed gasifier.

Run #	Bed material	T_{bed} °C	U m/s	ER	S/F	Q_{syngas} m^3_N/h	PCI_{syngas} kg/h	W_{tar} kg/h	CGE	CCE
3	sand	850	0.50	0.26	0	63	6100	6.2	0.50	0.64
4	sand	869	0.61	0.24	0	78	7000	7.8	0.55	0.66
5	sand	867	0.71	0.22	0	92	7300	12.1	0.52	0.62
6	sand	898	0.73	0.31	0	86	5500	7.0	0.51	0.68
7	sand	845	0.70	0.20	0	93	7900	12.6	0.51	0.59
C	sand	831	0.68	0.19	0.47	76	9200	8.3	0.59	0.67
D	sand	853	0.70	0.22	0.56	71	7200	7.7	0.52	0.64
8	olivine	813	0.68	0.24	0	131	7100	1.9	0.77	0.72
9	olivine	807	0.67	0.29	0	122	6500	0	0.82	0.79
10	olivine	819	0.68	0.21	0	132	7600	0	0.76	0.60
11	olivine	794	0.58	0.25	0	112	6700	0	0.76	0.77
12	olivine	816	0.68	0.28	0	123	6500	0	0.79	0.76
13	olivine	825	0.69	0.35	0	119	6300	0	0.93	0.63

improved in the successive runs, according to the evidence that an increased calcination time in air at temperatures between 850 and 900°C strongly increases the catalytic activity of olivine (Devi et al., 2005). The measurements made in these runs indicated an almost completely absence of tar and an increased production of syngas from about 80 to about $120 m^3_N/h$ It must also be noted that the reactor temperature in the experiments with olivine stabilizes at a lower value (about 815°C), probably as a consequence of the great extension of the endothermic tar decomposition reactions activated by the iron contained in the olivine (Dayton, 2002; Devi et al., 2005): that of thermal cracking ($pC_xH_y \rightarrow qC_nH_m + rH_2$) and that of carbon formation ($C_nH_m \rightarrow nC + {}^m/_2H_2$), where C_xH_y represents tar and C_nH_m represents hydrocarbons with a smaller carbon number than C_xH_y. Accordingly, it is particularly impressive the increase of H_2 concentrations (from 8-9% to 24-32%) in the exit gas (Fig. 2). There is also a remarkable increase of the amount of C fines found in the bed or collected in the cyclone as elutriated fines. This high production of solid carbon affects the equilibrium of Bouduard reaction and increases CO concentrations from about 2.5% to 18-20%.

The change in gas composition, clearly highlighted by Fig. 2 for runs carried out at a same fluidizing velocity, implies a remarkable improvement in the value of the two main parameters of gasifier performance: the Cold Gas Efficiency (CGE), which is calculated by dividing the chemical energy of the product gas by the chemical energy of the fuel at the feeding point, and the Carbon Conversion Efficiency (CCE), which is calculated by dividing the carbon in the product gas with that in the fuel (Basu, 2006). Data in Table 3 show that CGE lies in the range 50-59% in the runs with quartz sand and increases to 76-93% in those with olivine. There is a large increase also for CCE (from 59-68% to 60-79%), as a direct consequence of the larger cracking of heavier hydrocarbons, even though part of the carbon produced by the cracking covers the bed particles and remains in the reactor or is elutriated by attrition and collected in the
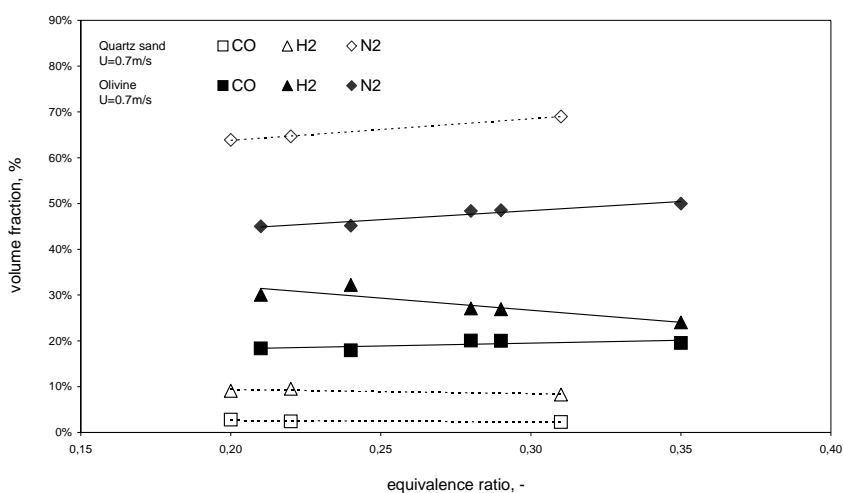

Figure 2 Volume fractions of CO, H_2 and N_2 in the syngas obtained at different equivalence ratios for runs with quartz sand and olivine as bed material.

cyclone.

In conclusion, the described results suggest that the utilization of natural olivine as bed material in plastic waste gasification could remarkably improve the quality of the product gas, in terms of both low tar content and high hydrogen volume fractions. The low cost of the mineral and its characteristic to be highly attrition resistant (Pfeifer et al., 2004; Devi et al., 2005) further support the opportunity of more investigations to test its performance in gasification experiments with other kind of wastes.

4. References

Arena U. & M.L. Mastellone, 2005, Fluidized Pyrolysis and Gasification of Solid Wastes, in Proc. of Industrial Fluidization South Africa 2005, The South African Institute of Mining and Metallurgy (ISBN 1-919782-83-0), pp. 53-68.

Arena U. & M.L. Mastellone, 2006, Fluidized Bed Pyrolysis of Plastic Wastes, chapter 16 in Scheirs, J. & Kaminsky, W. (Eds.), Feedstock Recycling and Pyrolysis of Waste Plastics, J. Wiley&Sons Ltd, pp. 435-474.

Arena U., E. Romeo, M.L. Mastellone, 2006, Fluidized Bed Gasification of Solid Wastes: Analysis of Possible Deviations of the Main Process Variables, in F. Winter (Ed.), Proc. of The 19th Int. Conf. on Fluidized Bed Combustion, Vienna, Austria, May 21-24.

Basu, P., 2006, Combustion and Gasification in Fluidized Beds, CRC Press.

Dayton, D., A review of the literature on catalytic biomass tar destruction, Milestone completion report, NREL/TP-510-32815 (project No. DE-AC36-99-GO10337), National Energy Laboratory, Golden, Colorado, USA, 2002.

Devi, L., K.J. Ptasinski, F.J.J.G. Janssen, 2003, A review of the primary measures for tar elimination in biomass gasification processes, Biomass and Bioenergy, 24:125-140.

Devi, L., M. Craje, P. Thune, K.J. Ptasinski, F.J.J.G. Janssen, 2005, Olivine as tar removal catalyst for biomass gasifier: catalyst characterization, Applied Catalysis, 294:68-79.

Higman, C. & M. van der Burgt, 2003, Gasification, Gulf Professional Publishing.

Klein, A., K. Whiting, E. Archer E., J. Schwager, September 2004, Gasification and pyrolysis: what is the current situation for waste management?, Waste Management World, 71-75.

Livingston W.R., 2002, Technical and Economic Assessment of Energy Conversion Technologies for MSW, Report No. B/WM/00553/REP for DTI Sust. Energy Progr. PUB URN NO: 02/1347.

Malkow, T., 2004, Novel and innovative pyrolysis and gasification technologies for energy efficient and environmentally sound MSW disposal, Waste Management, 24:53-79.

Magnolithe Ltd., 2006, Technical Datasheet of Olivine, St. Lorenzen 36, Austria

Pfeifer, C., R. Rauch, H. Hofbauer, 2004, In-bed catalytic tar reduction in a dual fluidized bed biomass steam gasifier, Ind. Eng. Chem. Res., 43:1634-1640.