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Fluidized Bed Combustion of Wet Biomass Fuel (Olive Husks)

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The present paper reports on experiments of fluidized bed combustion of dry and wet olive husks. The olive husks are a biogenic residue of the olive oil industry, accounting for around 80 % of olive mass on wet basis. They have residual water content up to 70 % and rather high heating value (i.e. 22.3 MJ/kg on dry basis). Huge production of olive husks occurs seasonally in the Mediterranean area, posing problems for proper disposal and valorisation because of the difficulty to store this material for long times. The research demonstrated that the olive husks can be smoothly and effectively burnt in fluidized bed with high combustion efficiency and very low emissions of pollutants and solid particulate in a bed temperature range between 800 and 850 °C. The energy balance on the combustor shows that a fraction of 10-15 % of the heat input can be directly extracted from the fluidized bed, the remaining being available in the hot flue gases. The focus was also on the emissions of nitrogen oxide. The research can have practical application for small scale and local installation, e.g. at olive oil factory, as demonstrated by the experimental campaign at pilot scale.

1. Introduction

Residues and wastes from agriculture and related industrial process are a valuable source of energy and materials for civil and industrial applications (McKendry, 2002). In most cases, they have a substantially high humidity and may contain elements different from C, O, and H, which poses problems in utilization with traditional plants because of pollution or deposition. The olive husks are a biogenic residue of the olive oil industry, accounting for around 80 % of olive mass on wet basis. Olives are hugely produced in regions facing to the Mediterranean Sea, particularly in Spain, Italy and Greece. Olive husks can be attractive as energy source because of their appreciable heating value and the easiness to be burned. In contrast, olive husks have a high water content that is between 40 and 70 %, depending on the typology of the production cycle. In order to avoid the cost and impact for delivery to large power plants, burning the husks on site in small-scale combustors is appealing. However, the high water content hinders the direct combustion by means of conventional equipment, unless adopting pre-treatments such as evaporation, centrifugation or pressing (Miccio and Poletto, 2009), that could also give rise to release of odours or polluted waters with impact on the environment.

Fluidized bed (FB) technology is suitable for burning a variety of biomass fuels, thanks to its intrinsic flexibility towards particle size, ash content as well as moisture content (Werther et al., 2000). The biomass volatiles are prevalently burned in the freeboard, causing overheating and high CO levels (Miccio et al., 2005), also in the case of dry olive husks (Akpulat et al., 2010). Furthermore, agglomeration phenomena and formation of nitrogen oxides are likely to occur. In fact the presence of alkalis in the ashes of the fuel leads to the chemical formation of amorphous phases with the silica that is the predominant constituent in common bed materials (Grimm et al., 2011). Since olive husks have large potassium content, their employment in FB combustion could be associated to the onset of agglomeration that can be extensive, leading to bed defluidization (Grimm et al., 2011). The emissions of nitrogen oxides are typically low in course of fluidised bed combustion of coal or biomass fuels, the generation of NO_x via thermal or prompt

mechanisms (Miller and Bowman, 1989) being normally negligible. Olive husks have substantially high N content, so nitrogen oxides (i.e. NO, NO₂ and N₂O) can be formed and released from fuel nitrogen, by a complex pathway of chemical reactions (Wartha et al., 2000).

The present work reports on an experimental research about the valorisation of olive husks via fluidized bed combustion, also carried out at pilot scale with wet fuel. Main results of the experiments include the combustion efficiency and the emissions of solid and gaseous pollutants for the combustion tests as well as the nitrogen conversion to NO_x at changing the fluidized bed material. The research can have practical application for small scale and local installation, particularly in olive oil factories and rural areas.

2. Experimental

2.1 Pilot scale plant

The pilot scale plant is a stainless steel fluidized bed (Figure 1) with square section (750 mm x 750 mm) and 4,000 mm high. It is insulated with ceramic fiber board, able to stand temperatures up to 1,000 °C. A lobe compressor supplies the air flow rate for combustion through a distribution perforated plate, having 1,296 holes 1 mm ID. The maximum power is 600 kW_{th}.

The feeding system of the fuel is mechanical-pneumatic, consisting of a hopper kept under stirring via rotating propellers and connected to a peristaltic pump driven by an inverter. The pressure in the air chamber of the distributor is monitored with precision through a water column manometer.

The primary abatement of dust is obtained by means of a cyclone (ID = 320 mm) that allows the separation of entrained particles with a diameter up to 10 μ m.



Figure 1: Schematic of the pilot scale fluidised bed plant

The combustor is equipped with a quick and efficient pre-heating system consisting of a gas oil burner with electronic regulation and flame control. The monitoring system consists of type K thermocouples and a multiple continuous gas analyzers for the measurement of the concentrations of O₂, CO, CO₂ and NO. The control system can be implemented via a programmable logic controller (PLC) and feedback loops arising from the signals of the gas concentration and temperature of the bed. The final control elements are given by two inverters connected to the air compressor and the pump of the fuel supply. A precision pump is used for sampling the dusts downstream the cyclone, collected in a cartridge filter for further analysis.

2.2 Laboratory scale facility

The laboratory equipment is a 110 mm ID and 1.4 m height stainless steel fluidized bed combustor. The reactor is electrically heated. The fluidizing air is pre-heated at variable temperature. The fuel is pneumatically fed at the bed bottom, by means of a rotating screw. The composition of the exhaust gases is measured by means of a set of continuous gas analyzers for measuring O_2 , CO, CO_2 , and NO. K-type thermocouples and high-precision piezo-resistive gas pressure transducers are installed in various points of the facility for monitoring the bed fluid-dynamics and the properties uniformity. The elutriated solid fines are separated by a cyclone and collected for determining the elutriation rate and the residual carbon content.

2.3 Materials

The bed material used for the combustion tests in the pilot plant was chromite with a bed inventory of 800 kg. Wet olive husks from a local oil factory, having water content between 60 and 70 % by mass, was used as fuel.

Four different bed materials were used during the laboratory tests: quartzite, two natural catalysts (olivine and chromite), and synthetic Cu oxide catalyst, already used for low temperature combustion of methane (lamarino et al., 2006).

Olive husks from south of Italy (particle size 2.0-4.0mm) was used as fuel, whose properties are reported in Table 1. The heating value of olive husks is substantially larger than that of wood. The content of potassium in the fuel is high (5026 ppm by mass), as determined by ICP analysis. Aluminium (1139 ppm) and calcium (704 ppm) are the most abundant elements in addition to the K (Si not determined). The characteristic temperatures of the ash (sintering, softening and flowing) were determined by means of a heated-microscope following standard procedure. The rather high values (1,260-1,300 °C) suggest that the ash cannot determine fouling phenomena, unless interactions with other elements, leading to low temperature eutectics, occur.

A thermogravimetric analysis carried out at 3 % of O_2 in Ar and 10 °C/min heating rate has shown that the devolatilization ends at around 350 °C. Afterwards, weight loss by char-carbon conversion takes place until complete burn-off at 830 °C, confirming the good reactivity of olive husks with respect to oxidation, as already reported by Senneca (2007).

Proximate analysis		Elements ir		ash (ppm)	
Moisture	9.3	Size, mm	2.0 - 4.0	K / 39	5026
Volatiles	72.2	Low heating value, MJ/kg	22.3	AI / 27	1139
Fixed carbon	14.3	Bulk density, kg/m ³	610	Ca / 40	704
Ash	4.2			P / 31	394
Ultimate analysis				Fe / 56	389
Carbon	52.6	Sintering temperature, °C	1,260	Mg / 24	326
Hydrogen	6.4	Softening temperature, °C	1,280	Na / 23	210
Nitrogen	0.8	Flowing temperature, °C	1,300	Ti / 47	37
Sulphur	0.1			Mn / 55	16
Oxygen (by diff.)	40.1				

Table 1: Physical and chemical properties of dry olive husks

3. Results

3.1 Catalytic effect of bed materials

Since no data are reported in literature about the use of chromite for thermo-chemical process, the chemical and thermal stability was evaluated by performing TPR/TPO (Temperature Programmed Reduction/ Oxidation) cycles. No relevant chemical changes of the chromite, even if heated up to 900 °C, have been observed during the TPR/TPO cycles denoting good thermal stability.

To assess a possible catalytic effect of olivine and chromite, CO to CO_2 oxidation catalytic activity tests (0.1 % vol. CO and 6 % vol.O₂ in a balance of N₂) were carried out in a fixed bed reactor (12.7 mm size and 600 mm length) operating under atmospheric pressure in the temperature range 600–850 °C with an equivalent contact time between 0.03 and 0.1 g/sNcm³. For comparison the tests have been also

performed on the copper catalyst that in a previous work (lamarino et al., 2006) exhibited a good activity during methane combustion in fluidized bed.

An appreciable homogenous contribution to the oxidation of CO to CO_2 , under the experimental conditions chosen that are representative the combustion tests, has been observed during fixed bed reactor tests. As expected the copper catalyst shows a very high activity. It is able to give complete conversion of CO to CO_2 at the lower temperature (600 °C) whatever is the contact time used. The chromite performance is not different from that observed under homogeneous oxidation suggesting that this material does not act as catalyst in the oxidation of CO. A marked increase in CO conversion, even if lower than that observed for the Cu catalyst, was observed when olivine has been used.

On the basis of the results obtained in the fixed tests the activity towards CO oxidation is the following: Cucatalyst >> olivine > chromite > quartzite.

3.2 Results of laboratory tests

The main results of the experimental tests carried out under steady state conditions are reported in Table 2. The analysis of the results indicates that an increase of temperature results in better combustion performance and lower CO concentration, with the exception of an anomalous variation for quartzite. Cu oxide is the most effective material in lowering carbon monoxide in agreement with the results obtained in fixed bed tests. The combustion efficiency $(1-CO/(CO+CO_2))$ was always higher than of 99.5 %.

The nitrogen oxides are generated from the fuel N, the thermal mechanism of NO_x generation being appreciable only at temperature well in excess of 1,500 °C (Miller and Bowman, 1989). The NO_x concentration only in some cases (in gray in Tab. 2) is above the threshold prescribed by the Italian normative that is 500 mg/Nm³ @ 11% O₂ vol., corresponding to 392 ppm @ 6%. The fuel nitrogen conversion to NO_x is between 10 % and 40 % and increases with T, CO having a reducing effect on NO_x concentration. This aspect is evident when the bed temperature changes: the former decreases whilst the latter increases, as clearly shown in Figure 2 reporting data obtained at different operating conditions.

The already noted anomalous variation of CO for quartzite had a beneficial effect on the NO_x concentration. In fact, the catalytic activity exerted at particle surface promotes the NO_x chemical reduction that is operated via CO interaction over silica (Vix-Guterl et al., 1996).

The ranking of materials with respect to NO reduction is: olivine > quartzite > Cu oxide >> chromite.



Figure 2: Nitrogen oxide concentration against carbon monoxide for different bed materials. Bed temperature 760-879°C; fluidization velocity 0.500-0.717 m/s; excess air ratio 1.27-1.52.

The limited effectiveness in reducing nitrogen oxide can be firstly ascribed to the segregation of fuel particles that is likely to occur with a fuel having high content of volatile matter. The fast release of the volatiles into the freeboard makes ineffective the presence of the catalyst due to the very low contact time between the N rich species and the active surface. For small fuel particles, as in the case of this research, the devolatilization time lasts a dozen of seconds, whilst the segregation of the particle at the bed surface is very fast. Following Fiorentino and Miccio (2000), only 1/3 of the released volatiles can efficiently contact the catalytic or sorbent fuel particles. Therefore, the NO_x reduction by CO is shifted into the freeboard and can result not completely effective.

		Quartz	ite		Chro	omite	Cu	oxide	Oli	vine
Bed inventory, kg	5	7.5	5	5	5	5	5	5	5	5
Density, kg/m ³		2,600)		4,	170	1,	800	3,2	200
Size, mm		0.1-0.	4		0.20	-0.30		1	0.20	-0.30
Bed temperature, °C	806	838	760	880	782	878	782	847	786	846
Fluidization velocity, m/s	0.510	0.530	0.500	0.500	0.526	0.524	0.717	0.683	0.566	0.540
Umf, m/s	0.058	0.058	0.062	0.058	0.073	0.069	0.216	0.203	0.066	0.062
Excess air ratio	1.27	1.29	1.47	1.46	1.52	1.52	1.42	1.47	1.46	1.46
O2, %	5.27	5.49	5.6	5.4	5.7	6.8	6.7	5.9	6.3	7.2
CO2, %	14.56	14.35	13.9	14.3	14.0	12.9	13.6	14.4	13.8	13.1
CO, ppm	439	452	79	172	439	87	127	16	1319	309
NOx, ppm	247	331	305	308	485	693	392	477	235	410
Elutriation rate, g/h	10.04	0.45	12.40	11.20	10.20	10.12	15.30	38.13	28.53	8.31
C elutriation rate, g/h	0.10	0.00	0.87	0.34	0.71	0.30	0.18	0.34	0.27	0.13

Table 2: Experimental conditions and results of combustion tests in laboratory scale facility

3.3 Results of pilot scale tests

In general, combustion tests of wet olive husks were carried out in the pilot scale plant under steady state conditions. The operation was smooth and reliable, in spite of the high water content in the fuel. Even upon prolonged operation (> 100 hours), no trace of sand agglomeration was found, confirming the suitability in this concern of the chromite as bed material. The main results of the tests are reported in Table 3. Again, there is an inverse relation between CO and NO emission levels, as already evidenced in lab scale tests. The combustion efficiency was very high and the carbon content in elutriated fines was less than 1% by mass. So far, the pilot scale tests fully confirmed the preliminary results at laboratory-scale. The energy balance on the combustor showed that a fraction of 10-15 % of the heat input can be directly extracted from the fluidized bed, the remaining being available in the hot flue gases.

Test	#1	#2	#3	#4	#5
Bed temperature, °C	820	820	840	825	825
Freeboard temperature, °C	840	840	850	840	840
Water content in the fuel, % mass	65.0	65.0	63.3	62.5	62.5
Excess air ratio	1.66	1.78	1.26	2.14	1.76
Fluidization velocity, m s ⁻¹	0.92	0.92	0.92	0.92	0.76
O ₂ , %	4.8	5.9	2.6	10.0	7.5
CO ₂ , %	12.5	11.8	16.8	9.5	12.3
CO, ppm	45	15	299	5	50
NO, ppm	450	500	339	565	400
Efficiency	99.6	99.6	99.4	99.9	99.8
Power, kW _{th}	341	319	447	264	264

Table 3: Experimental conditions and results during combustion tests in the pilot scale plant

It must be remarked that only a slight increase of temperature (≤ 20 °C) in the freeboard occurred, the most of the combustion being completed within the bed. This can be explained by considering the interaction of the wet fuel jet with the hot bed particles (Miccio et al., 1997), leading to formation of aggregates. It is likely that such fuel-sand aggregates have a more favourable combustion mechanism in the bed thanks to higher fuel residence time and lower bed bypass of combustible matters.

Particulate concentration measured at the exit was in the order of 1 g/m³, accounting for the fines that are generated from the comminution of the ash skeleton. The use of a downstream bag filter would easily reduce the particulate emissions in the flue-gas, allowing to comply with normative prescriptions (100 mg/m³ @11%O₂).

The most of ashes were elutriated and separated by the cyclone, whereas a small amount of ash was retrieved in the bed, being prevalently formed by impurities in the fuel (sand, fragments, etc.). Although biomass ashes can be considered as no-hazardous waste, the high ash content of the olive husks (4.2 %) is appealing for possible utilisation in alternative to disposal. Biomass ashes are potential source of

nutrients for plants, since they are rich in K, P, Mg, Na, N, S that are valuable for agriculture (Pels et al., 2005). According to ICP analysis, some of these elements are present in the ashes of olive husks.

4. Conclusions

The research demonstrated that the olive husks can be effectively burnt in fluidized bed with high combustion efficiency and very low emissions of unconverted gaseous and solid pollutants in a bed temperature range between 800 °C and 850 °C.

The fuel nitrogen is responsible of NO_x emissions that are strictly correlated to the CO concentration. The catalysts adopted during the tests as bed material seem to be poorly effective in reducing the nitrogen oxide emissions. So far, an improved mixing between bed and N species could result beneficial.

Agglomeration phenomena can be avoided by operating the bed at lower temperature and with more intense mixing, and adopting materials with low silica content. In particular, the chromite exhibited good resistance to the onset of agglomeration.

The scale-up of the FB combustion process was demonstrated in a full scale plant with wet olive husks (60 % water by mass), obtaining clean, smooth and reliable operation without any relevant problem.

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