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ATTRITION OF BED MATERIALS AND FUEL PELLETS FOR FLUIDIZED BED GASIFICATION APPLICATION

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ABSTRACT

This paper reports on a study of the attrition/fragmentation behavior of different bed materials and fuel pellets for application in fluidized bed gasification. Three different bed materials displaying catalytic activity, namely fresh and sintered dolomite and a Ni-alumina catalyst, were tested for their resistance to fragmentation and attrition in fluidized bed. The fresh dolomite displayed extensive particle breakage upon calcination and a large production of attrited fines during fluidized bed operation. The other two materials were much more resistant to attrition and appeared to be suitable for further long-term operation testing. The attrition/fragmentation resistance of three pelletized fuels, one based on wood and the other two on a mixture of wood and coal, was also characterized under both inert and gasification conditions. Pellet breakage by primary fragmentation upon devolatilization appeared to be rather limited for all fuels. On the contrary, attrition of carbon fines from the char particles during gasification was extensive, due to a gasification-assisted attrition mechanism.

INTRODUCTION

Gasification consists in the conversion of solid carbonaceous fuels, such as coal or biomass, into syngas via partial oxidation reactions. The syngas can be directly used as a fuel or undergo further processing to yield H_2 or liquid fuels. Because of its cleaner and more flexible nature, gasification is a favorable candidate for use in next-generation coal-based plants, especially in a carbon constrained scenario. Despite its various advantages over direct combustion, gasification faces several issues that have limited so far its widespread application. Among them, gasification is usually more capital-intensive and has often suffered for lack of reliability and availability.

Gasification of solid biomass yields a high quality syngas with a more favorable H_2/CO ratio with respect to coal and with lower energy demand, because of the larger content of hydrogen in the fuel structure. Biomass fuels, however, are characterized by a low energy specific content if compared with fossil fuels. Fuel pre-treatments like pelletization or torrefaction/compaction (1,2), are appealing techniques to increase the bulk density and energy specific content as well as to improve the fuel properties (e.g. homogenizing, stabilizing and strengthening the fuel particles).

Another option to overcome the limitation of the low energetic density of biomasses is offered by their co-processing with coal, since the latter has an almost double energetic density. This measure also turns out to be useful when the primary fuel (i.e. the biomass) is temporarily lacking because of seasonal availability. Of course, the process must be flexible towards change of the fuel properties. This is the case of fluidized bed (FB) gasification that is acknowledged to have great flexibility and high efficiency in conversion of several solid fuels, including low rank coals, into a valuable synthesis gas (3).

A research project on FB gasification, FLEXGAS - Near Zero Emission Advanced Fluidized Bed Gasification, is currently in progress with the coordination of IRC-CNR under the sponsorship of the research Fund for Coal and Steel of the European Commission. The project deals with FB co-gasification of coal and biomass/waste using in-bed catalytic materials for tar reduction, at different reactor scales. Among the various scientific activities, attention is also devoted to attrition and fragmentation phenomena that are well known to affect the reliability and efficiency of FB combustion and gasification processes. On the one side, particle attrition/fragmentation phenomena cause the elutriation of fine material from the bed that results in the loss of valuable catalyst or unconverted carbon. On the other side, they may significantly change the particle size distribution of the materials in the bed which influences the bed fluid-dynamics, heat and mass transfer coefficients and reaction rates (4,5).

This paper reports on a study of the attrition/fragmentation behavior of different bed materials and fuel pellets used in the gasification process, carried out in a lab-scale FB apparatus. Three different bed materials displaying catalytic activity, namely fresh and sintered dolomite and a Ni-alumina catalyst, were tested for their resistance to primary fragmentation and attrition upon FB processing. In addition, the attrition/fragmentation resistance of three pelletized fuels, one based on wood and the other two on a mixture of wood and coal, was also characterized under both inert and gasification conditions.

EXPERIMENTAL

A stainless steel atmospheric bubbling FB combustor 40 mm ID and 1 m high was used for fragmentation and attrition experiments. A 2 mm thick perforated plate with 55 holes 0.5 mm in diameter disposed in a triangular pitch was used as gas distributor. A 0.6 m high stainless steel column for gas preheating and mixing was placed under the distributor. Two semicylindrical 2.2 kW electric furnaces were used for heating the fluidization column and the preheating section. The temperature of the bed, measured by means of a chromel-alumel thermocouple placed 40 mm above the distributor, was kept constant by a PID controller. Gases were fed to the column via high-precision digital mass flowmeters. Further details of the apparatus are reported elsewhere (4).

Two different reactor configurations were used for the tests. In the first configuration, used for particle fragmentation experiments, the top section of the fluidization column was left open to the atmosphere. When testing the bed materials, a bed of sand (0.71-0.8 mm, 180 g) was fluidized with air at 0.3 m/s. Experiments were carried out by injecting batches (40 g) of dolomite or catalyst particles into the bed,

kept at 800°C, from the top of the column. After about five minutes, required to completely calcine the material, the bed was discharged, and the dolomite/catalyst particles were gently sieved out from the sand in order to investigate the particle size distribution of the produced fragments. In the pelletized fuel tests a basket equipped configuration was used. A bed of sand (0.3–0.4 mm, 180 g) was fluidized with nitrogen at 0.3 m/s. During the run a stainless steel circular basket was inserted from the top to retrieve the particles from the bed. A basket mesh of 0.8 mm was used, so that the sand could easily pass through the net openings. Following the procedure proposed by Chirone et al.(6), experiments were carried out by injecting single fuel particles into the bed kept at 800°C from the top of the column. After about three minutes, required to completely devolatilize the fuel, the resulting char was retrieved by means of the basket in order to investigate the number and size of the produced fragments. The experiment was repeated with more than 30 particles in order to collect a statistically significant number of fragments.

In the second configuration, used for fines elutriation rate (attrition) experiments, a two-exit brass head was fitted to the top flange of the column. By operating a valve it was possible to convey flue gases alternately to two removable sintered brass filters. Batches (20 g for dolomite/catalyst tests, or 2.0 g for pre-devolatilized fuel pellet char tests) of materials were fed to the bed (0.3–0.4 mm sand, 150 g). The bed was kept at 800°C and fluidized at 0.3 m/s with air (bed material tests), or at 0.8 m/s (fuel char tests) either with nitrogen (inert conditions) or with a N_2-CO_2 mixture with 60% CO_2 concentration (gasification conditions). Elutriated fines were collected by means of the two-exit head by letting the flue gas flow alternately through sequences of filters (one was in use while the previous one was replaced) for definite periods of time. In order to prevent hydration of the collected material, each filter was quickly put in a drier after being used, where it was cooled down before it was weighed. The difference between the weights of the filters before and after operation, divided by the time interval during which the filter was in operation, gave the average fines elutriation rate relative to that interval. Fines collected in the filters during the fuel char tests were further analyzed to determine their fixed carbon content. The assumption underlying this procedure was that the residence time of elutriable fines in the reactor could be neglected and that elutriation rate could be assumed equal to the rate of fines generation by attrition at any time.

Materials

Fresh and sintered dolomite (0.212-0.400 mm) and a Ni-γ-Al_2O_3 catalyst (0.125-0.25 mm) were tested in the FB. Dolomite and Ni-γ-Al_2O_3 were selected because of their catalytic activity, relevant to gasification (7). The Ni-based catalyst (5.5%wt. Ni) was prepared by wet impregnation with nickel-nitrate of a commercial γ-Al_2O_3 (PURALOX SCCA-150/200) (8). The sintered dolomite was a commercial product (Pentadol 0-1) roasted at 2000°C. Table 1 reports the properties of the bed materials.

The fuel particles characterized in the fluidized bed were: commercial spruce wood pellets; home-made pellets composed either of 70% pine wood – 30% German brown coal or 70% pine wood – 30% Polish bituminous coal (by weight). The starting materials for the home-made pellets were all in the particle size range 0.5-2.0 mm. All the pellets had cylindrical shape with a diameter of 6 mm and an average length of 20 mm. Table 2 reports the properties of the fuel pellets.

Table 1 Properties of the bed materials.

Bed Material	Chemical analysis (%wt.)		Density (kg/m^3)	Particle size range (mm)	Minimum fluidization velocity (cm/s)
Dolomite	Al_2O_3:	1.0	1600	0.212-0.400	3.0
	$CaCO_3$:	58.0			
	$MgCO_3$:	41.0			
Sintered dolomite	SiO_2:	1.0	1500	0.212-0.400	4.1
	Al_2O_3:	0.5			
	$CaCO_3$:	58.5			
	$MgCO_3$:	39.5			
	Fe_2O_3:	0.5			
Ni/γ-alumina	Ni:	5.5	1800	0.125-0.250	0.7
	γ-alumina:	94.5			

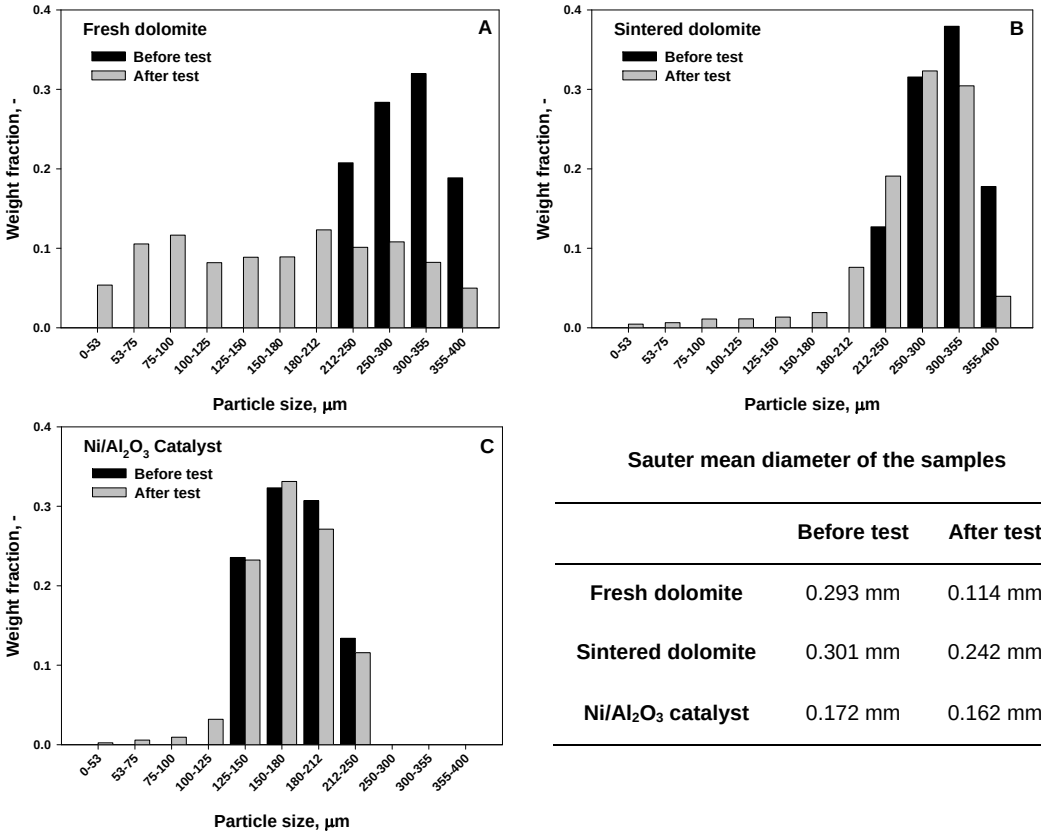
Table 2 Properties of the fuel pellets.

Fuel:	Spruce-wood pellets	70% wood - 30% German coal pellets	70% wood - 30% Polish coal pellets
Pellet size, mm (diameter x length)	6x20	6x20	6x20
Proximate analysis (as received)			
Moisture, %wt.	8.4	9.3	8.2
Volatiles, %wt.	74.2	66.8	61.4
Fixed carbon, %wt.	17.1	22.8	27.6
Ash, %wt.	0.3	1.1	2.8
Ultimate analysis (dafb)			
Carbon, %wt.	49.4	55.1	58.7
Hydrogen, %wt.	5.9	5.7	5.8
Nitrogen, %wt.	<0.1	0.1	0.4
Oxygen, %wt.	44.6	39.1	35.1
Lower heating value, MJ/kg	18.5	21.7	23.4

RESULTS

Bed materials

Figures 1A-C show the particle size distribution (PSD) of the fresh and sintered dolomite and of the Ni-γ-Al_2O_3 catalyst before and after fragmentation tests at 800°C in the lab-scale FB. Figure 1 also reports the Sauter mean diameter of the different samples before and after the tests. Fresh dolomite (Fig. 1A) undergoes extensive fragmentation by thermal/mechanical shock during the test, as demonstrated by the amount of particles with a size below 0.212 mm (fragments) being of the order of 65%wt. On the other hand, sintered dolomite (Fig. 1B) is much more resistant to fragmentation, the amount of particles with a size below 0.212 mm after the test being less than 15%wt. The sintering process appears to be very effective in strengthening the dolomite particle structure. Also the catalyst particles (Fig. 1C) appear to be very resistant to thermal/mechanical fragmentation. The PSDs before and after the test are almost equal, with a very limited tail of fragments below 0.125 mm. These findings are confirmed by the comparison of the Sauter mean diameters.



Sauter mean diameter of the samples

	Before test	After test
Fresh dolomite	0.293 mm	0.114 mm
Sintered dolomite	0.301 mm	0.242 mm
Ni/Al_2O_3 catalyst	0.172 mm	0.162 mm

Figure 1 Results of bed materials fragmentation tests (T=800°C; U=0.3 m/s).

Figures 2A-B show the rate of fines elutriation from the bed (E) as a function of time, for the fresh and sintered dolomite and for the Ni-γ-Al_2O_3 catalyst during the attrition FB tests. The E vs t curve shows the typical decreasing trend, until a steady-state value is reached (9). The initial higher fines generation is due to the rounding off of the rough particles by mechanical removal of surface asperities. Consistently with the fragmentation results the fresh dolomite shows a larger fines elutriation rate, indicating that this material is more fragile than the other two. Both sintered dolomite and catalyst particles have a relatively good resistance to attrition. In comparing the results, however, it must be considered that the catalyst particles have a smaller particle size than the dolomite ones, and that the attrition rate in a FB is reported to be proportional to the inverse of the average particle size (9). In conclusion, fresh dolomite appears not to be appropriate for a reliable process, whereas sintered dolomite and Ni-γ-Al_2O_3 catalyst are suitable for further long-term operation testing.

Pelletized fuels

Primary fragmentation tests have been carried out by monitoring the number and size of fragments produced by each fuel pellet upon FB devolatilization. Table 3 reports an overview of the results of primary fragmentation experiments for the three fuels. Results are expressed using the following quantities: d_0 = Sauter mean diameter of the initial pellets; S_f = primary fragmentation probability, the number of fuel pellets that undergo fragmentation divided by the total number of pellets fed;

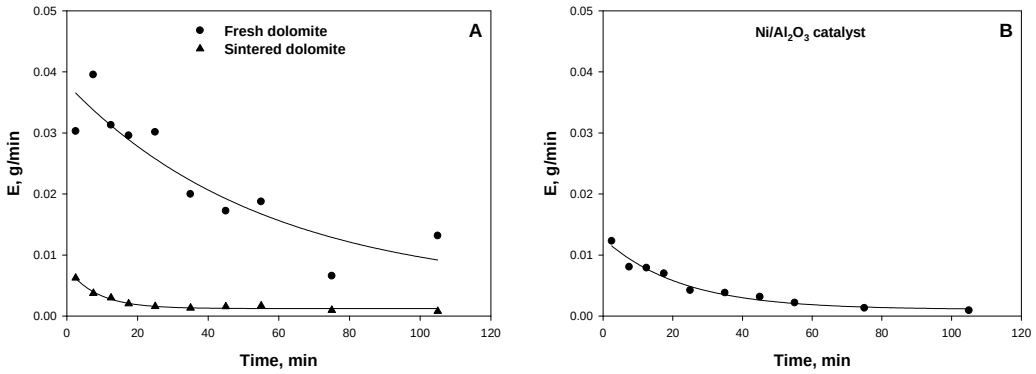


Figure 2 Results of bed materials attrition tests (T=800°C, U=0.3 m/s).

n_1 = primary fragmentation multiplication factor, representing the number of generated fragments per unit fuel pellet fed to the reactor; d_1 = Sauter mean diameter of the char particles after devolatilization. Results show that for all the three fuels limited primary fragmentation occurs, with a fragmentation probability between 20 and 30% and particle multiplication factor of the order of 2-3. The slight decrease of the Sauter mean diameter upon devolatilization is caused by the combined effect of primary fragmentation and particle shrinkage (2). These results are confirmed by Figure 3, showing the cumulative particle undersize distributions (on mass basis) of the char pellets after devolatilization as compared to the initial fuel pellets size distributions. A shift of the curves towards smaller sizes is clearly visible, but the population of smaller fragments (< 5 mm) does not significantly increase.

Figure 4 shows the rates of carbon fines elutriation measured during batch attrition experiments with the three pelletized fuel chars. The carbon elutriation rate (E_C) was normalized with respect to the initial amount of fixed carbon fed to the reactor (W_{C0}). Results of experiments carried out under inert and gasification conditions (60% inlet CO_2 concentration) are compared. The curves under inert conditions are similar one to the other and follow the typical decreasing trend towards an asymptotic value. However, the extent of fines attrition is different among the three chars. Spruce-wood char appears to be harder than the other two, suggesting that the commercial pelletization procedure yields more compact pellets than the home-made one. This is reflected by the total amount of elutriated carbon upon the runs: the percentage of the initial carbon fed with the fuel char that was cumulatively elutriated during the 120 min runs in inert condition was 7, 21 and 12% for the spruce-wood, German coal – wood and Polish coal – wood chars, respectively.

Table 3 Results of pelletized fuel primary fragmentation tests.

Fuel	d_0, mm	S_f	n_1	d_1, mm
Spruce-wood pellets	7.27	0.20	1.75	5.96
70% wood - 30% German coal pellets	7.35	0.21	2.47	6.37
70% wood - 30% Polish coal pellets	7.30	0.30	3.30	6.24

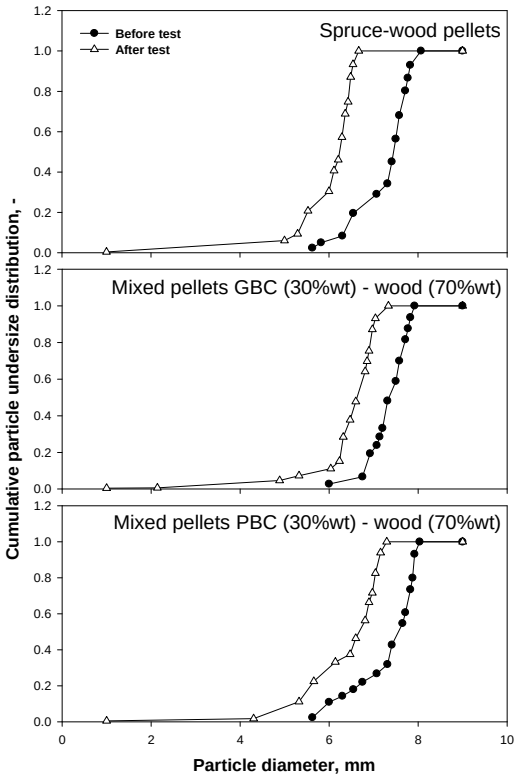


Figure 3 Particle size distribution of fuel pellets before and after primary fragmentation tests (T=800°C, U=0.3 m/s).

A completely different scenario was obtained under gasification conditions. All the curves showed a significant peak in the carbon elutriation rate at some point during particle conversion. It is worth to note that under gasification condition the tests lasted until complete carbon consumption, as witnessed by the vanishing of the CO concentration at the FB outlet. As a consequence, the total duration of the runs reflected the intrinsic reactivity of the fuels. As expected, the spruce-wood char was the most reactive, while the Polish coal – wood char was the least. The presence of the peak in the carbon elutriation rate indicates that carbon consumption in the pellets progressively weakens the char structure by pore enlargement, much like the well known combustion-assisted enhancement mechanism typically observed during FB coal combustion (6). This gasification-assisted enhancement is dependent on the parent fuel reactivity, since lower rates of carbon consumption shift towards longer times the peak position. The decreasing part of the curves at the right of the peaks are obviously caused by the progressive decrease of the carbon loading in the bed that overtakes the attrition enhancement at late stages of carbon conversion. It is important to note that the parent materials used for producing the home-made pellets were relatively coarse (0.5-2.0 mm). Thus, any effect on the char pellet attrition played by fine feedstock in the coals (i.e. < 100 μm) can be excluded, whereas the coal char reactivity and hardness play the major role.

The percentage of the initial carbon fed with the fuel char that was cumulatively elutriated upon the runs in gasification condition was 14, 29 and 38% for the spruce-wood, German coal – wood and Polish coal – wood chars, respectively. This figures show that the carbon loss by elutriation is certainly one of the critical factors during the gasification process, especially for the char containing the less reactive polish bituminous coal. On the other hand, the higher reactivity and mechanical strength of the commercial spruce-wood pellets appear to be beneficial for obtaining a larger carbon conversion during gasification.

CONCLUSIONS

In this study the attrition/fragmentation behavior of different bed materials and fuel pellets used in the gasification process was investigated in a lab-scale FB apparatus.

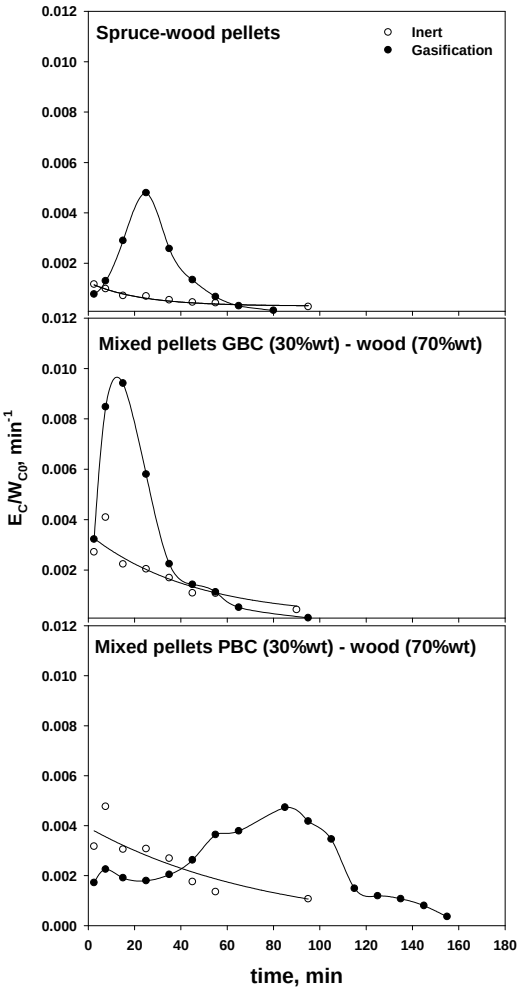


Figure 4 Results of fuel char attrition tests (T=800°C; U=0.8 m/s).

Three different bed materials displaying catalytic activity, namely fresh and sintered dolomite and a Ni-alumina catalyst, were tested. Large fragmentation was experienced for the fresh dolomite, whereas sintered dolomite and Ni-γ-Al_2O_3 catalyst appear to be resistant to thermal/mechanical shocks. Attrition tests also suggest that the fresh dolomite is practically unsuitable for a steady operation.

In addition, the attrition/fragmentation resistance of three pelletized fuels, one based on wood and the other two on a mixture of wood and coal, was also characterized under both inert and gasification reacting conditions. Pellet breakage by primary fragmentation upon devolatilization appeared to be rather limited for all fuels, indicating that the pelletization procedure is able to give sufficient mechanical strength to the particles. On the contrary, attrition of carbon fines from the char particles during gasification was extensive, due to a gasification-assisted attrition mechanism. The low reactivity of the generated fines under gasification conditions makes the loss of carbon by fines elutriation much more significant than that typically found under combustion conditions.

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