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Devolatilization and Attrition Behavior of Fuel Pellets during Fluidized-Bed Gasification

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ABSTRACT: Devolatilization, fragmentation, and attrition phenomena occurring to fuel pellets during fluidized-bed gasification have been studied. Three pelletized fuels, one based on wood and the other two based on a mixture of wood and coal, were characterized in a lab-scale fluidized-bed reactor under gasification, inert, and combustion conditions, for comparison. Similar and relatively long devolatilization times were observed for the three types of pellets. Pellet breakage by primary fragmentation upon devolatilization appeared to be rather limited for all fuels, indicating that fuel pelletization gives sufficient mechanical strength to the particles. On the contrary, secondary fragmentation and attrition by abrasion of char particles during gasification were extensive, suggesting a gasification-assisted attrition enhancement effect. This mechanism, associated with the low reactivity of the generated fines, made the loss of carbon by fine elutriation during char gasification much more significant than that found under combustion conditions. Larger carbon losses were associated with fuel pellets with a lower reactivity.

■ INTRODUCTION

Gasification consists of 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.[1] Despite its advantages over direct combustion, gasification faces several issues that have thus far limited its widespread application. Among them, gasification is usually more capital-intensive and has often suffered for the lack of reliability and availability. In addition, gas cleaning is a typical problem in gasification.

Gasification of solid biomass yields a high-quality syngas with a more favorable H_2/CO ratio with respect to coal and with a 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 to fossil fuels. Fuel pretreatments, such as pelletization or torrefaction/compaction,[2,3] are appealing techniques to increase the bulk density and energy-specific content, to improve the fuel properties (e.g., homogenizing, stabilizing, and strengthening the fuel particles), and to simplify the design of handling and storage devices.

Another option to overcome the limitation of the low energetic density of biomasses is offered by their co-processing with coal, because 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 toward the 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[4] and allows the use of catalysts and sorbents directly in the reaction chamber for improving the gas quality.[5,6]

Upon devolatilization and primary fragmentation, a fragile char particle is generated that undergoes attrition and secondary fragmentation; all of these phenomena are well-known to affect the reliability and efficiency of FB combustion and gasification processes.[7,8] On the one side, these phenomena may significantly change the particle size distribution of the fuel in the bed, which influences the rate and mechanism of fuel particle conversion, as well as the particle heat- and mass-transfer coefficients. On the other side, they may cause the elutriation of fine material from the bed that results in the loss of unconverted carbon. With this respect, it has been underlined that the relevance of attrition and fragmentation phenomena is emphasized when using high-volatile fuels (biomass and waste) instead of coals, because highly porous and friable chars are formed upon devolatilization.[3,7]

Even if several attrition studies have been carried out under FB combustion conditions, focused on either coal[8] or alternative raw or pelletized fuels,[3,7,9] only very limited activity has been reported under gasification conditions.[10,11] Furthermore, information about devolatilization of pellets in FB is sparse.

This paper reports on a study on the devolatilization, fragmentation, and attrition of three pelletized fuels, one based on wood and the other two based on a mixture of wood and coal, tested under gasification conditions in a lab-scale FB apparatus. The devolatilization time, the propensity to segregate on the bed surface, and the tendency to fragment were investigated, providing qualitative/quantitative data for modeling. Additional experiments under oxidizing and inert conditions were carried out to better understand the mechanisms of pellet attrition and to underline the differences in the pellet behavior upon changing the reaction environment.
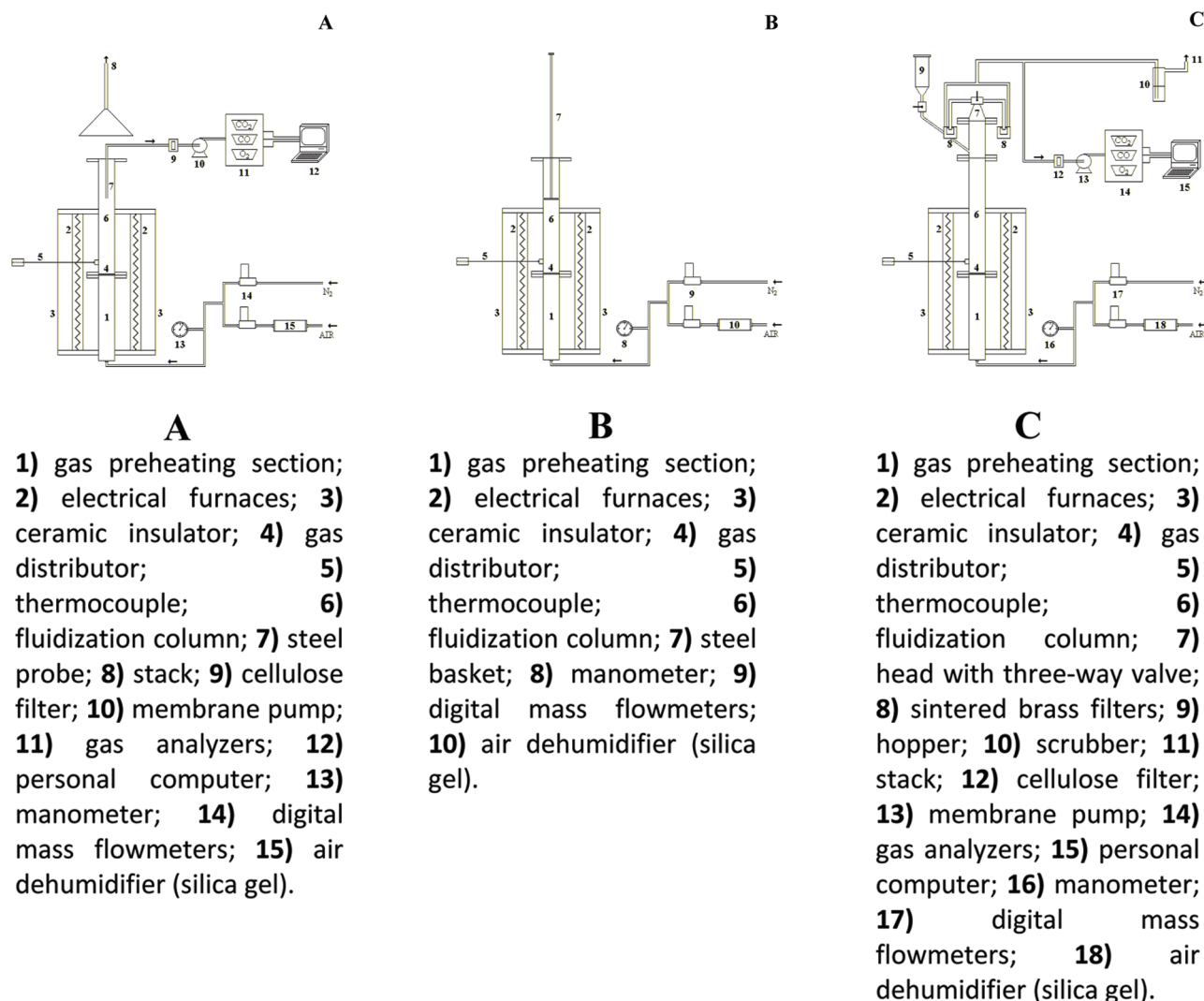
■ EXPERIMENTAL SECTION

Apparatus and Procedures. An electrically heated stainless-steel atmospheric bubbling FB combustor, 40 mm inner diameter and 1 m high, was used for the experiments (Figure 1). Details of the apparatus are reported elsewhere.[7]

A

1) gas preheating section; **2)** electrical furnaces; **3)** ceramic insulator; **4)** gas distributor; **5)** thermocouple; **6)** fluidization column; **7)** steel probe; **8)** stack; **9)** cellulose filter; **10)** membrane pump; **11)** gas analyzers; **12)** personal computer; **13)** manometer; **14)** digital mass flowmeters; **15)** air dehumidifier (silica gel).

B

1) gas preheating section; **2)** electrical furnaces; **3)** ceramic insulator; **4)** gas distributor; **5)** thermocouple; **6)** fluidization column; **7)** steel basket; **8)** manometer; **9)** digital mass flowmeters; **10)** air dehumidifier (silica gel).

C

1) gas preheating section; **2)** electrical furnaces; **3)** ceramic insulator; **4)** gas distributor; **5)** thermocouple; **6)** fluidization column; **7)** head with three-way valve; **8)** sintered brass filters; **9)** hopper; **10)** scrubber; **11)** stack; **12)** cellulose filter; **13)** membrane pump; **14)** gas analyzers; **15)** personal computer; **16)** manometer; **17)** digital mass flowmeters; **18)** air dehumidifier (silica gel).

Figure 1. Experimental apparatus: (A) open-top configuration, (B) basket-equipped configuration, and (C) two-exit head configuration.

Three different reactor configurations were used for the tests. The first configuration (Figure 1A) was used for particle devolatilization experiments. The top section of the fluidization column was left open to the atmosphere. A stainless-steel probe was inserted from the top of the column (at 0.6 m above the distributor) to convey a fraction of the exit gases to the gas analyzers. A paramagnetic analyzer and two non-dispersive infrared (NDIR) analyzers were used for online measurement of O_2, CO, and CO_2 concentrations, respectively, in the exhaust gases. Tests were carried out with a 180 g bed of silica sand, either 0.20–0.40 or 0.40–0.80 mm in size (U_{mf} = 0.03 and 0.13 m/s, respectively), at 800 °C and with the bed operating under a bubbling fluidization regime at a superficial gas velocity of 0.3 m/s. Air was used as fluidizing gas. Experiments were carried out by feeding single pellets from the top of the column and following the progress of devolatilization by measuring the CO_2 concentration at the outlet. About 10 repetitions were performed for each kind of fuel pellet. Alternatively, some tests were carried out by keeping the fuel pellet inside the bed, at a depth of around 100 mm from the bed surface, by means of a stainless-steel cage.

The second configuration (Figure 1B) was used for primary fragmentation experiments. In this configuration, the top section of the fluidization column was also left open to the atmosphere and the basket technique described by Chirone et al.[8] was applied. A bed of sand (0.2–0.3 mm, 180 g) was fluidized with nitrogen at 0.3 m/s. During the run, a

Table 1. Properties of the Fuel Pellets

	spruce wood pellets	70% wood/ 30% German coal pellets	70% wood/ 30% Polish coal pellets
pellet size (mm) (diameter × length)	6 × 20	6 × 20	6 × 20
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

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. Experiments were carried out by feeding single fuel particles into the bed kept at 800 °C from the top of the column. After devolatilization was finished (5 min), the resulting char was retrieved by means of the basket to investigate the number and size of the produced fragments. The experiment was repeated with more than 30 particles to collect a statistically significant number of fragments.

The same apparatus configuration was used for secondary fragmentation experiments under gasification conditions (60% CO_2 concentration in N_2), at the same operating conditions ($T = 800$ °C, and $U = 0.3$ m/s). In these experiments, 3−4 pre-devolatilized char particles were fed into the bed from the top of the column. At definite times, the char was retrieved from the bed with the basket and the fragments were weighed and photographed. The run ended when no more carbon was found in the basket; i.e., the fragments were consumed by gasification and attrition down to a size smaller than the mesh-opening size.

In the third configuration (Figure 1C), used for attrition experiments, a two-exit brass head was fitted to the top flange of the column. When a valve was operated, it was possible to convey flue gases alternately to two removable sintered brass filters. Batches (2.0 g) of pre-devolatilized char were fed to the bed of sand (0.3−0.4 mm, 180 g). The bed was kept at 800 °C and fluidized at 0.8 m/s with either nitrogen (inert conditions), a N_2−CO_2 mixture with a 60% CO_2 concentration (gasification conditions), or with a N_2−O_2 mixture with a 4.5% O_2 concentration (combustion conditions). Elutriated fines were collected by means of the two-exit head by letting the flue gas flow alternately through sequences of filters for definite periods of time. 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 fine elutriation rate relative to that interval. Fines collected in the filters 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 the elutriation rate could be assumed equal to the rate of fine generation by attrition at any time.

Fuel Pellets. The fuel pellets used in the experiments were commercial spruce wood pellets or homemade mixed pellets composed of either 70% pine wood/30% German brown coal (GBC) or 70% pine wood/30% Polish bituminous coal (PBC) by weight. The starting materials for the homemade pellets were all in the particle size range of 0.5−2.0 mm. All of the pellets had cylindrical shape, with a diameter of 6 mm and an average length of 20 mm. Table 1 reports the properties of the fuel pellets. As expected, the wood pellets have the largest volatile matter content and the lowest ash content. The fixed carbon content of the mixed pellets increases with the fixed carbon content of the original coals.

■ RESULTS

Devolatilization Experiments. In these experiments, single fuel pellets were fed in the bed at 800 °C, fluidized by an air flow at 0.3 m/s. Oxidizing conditions with a large excess air ratio were used in the devolatilization tests, resulting in an easier analysis of the gas effluents, because the volatiles promptly burned and formed CO_2 as they left the bed. With this respect, it is likely that, under gasification or inert conditions, the devolatilization behavior of the pellets is very similar to that experienced under combustion conditions. In fact, during high-temperature FB devolatilization, the emitted volatiles form a shield that surrounds the fuel particle and prevents significant chemical reaction with bulk species (e.g., O_2, CO_2, and H_2O), independent of the oxidizing/reducing environment around the particle.[12]

The devolatilization behavior of the pellets was preliminary characterized by visual observation of the bed surface during the
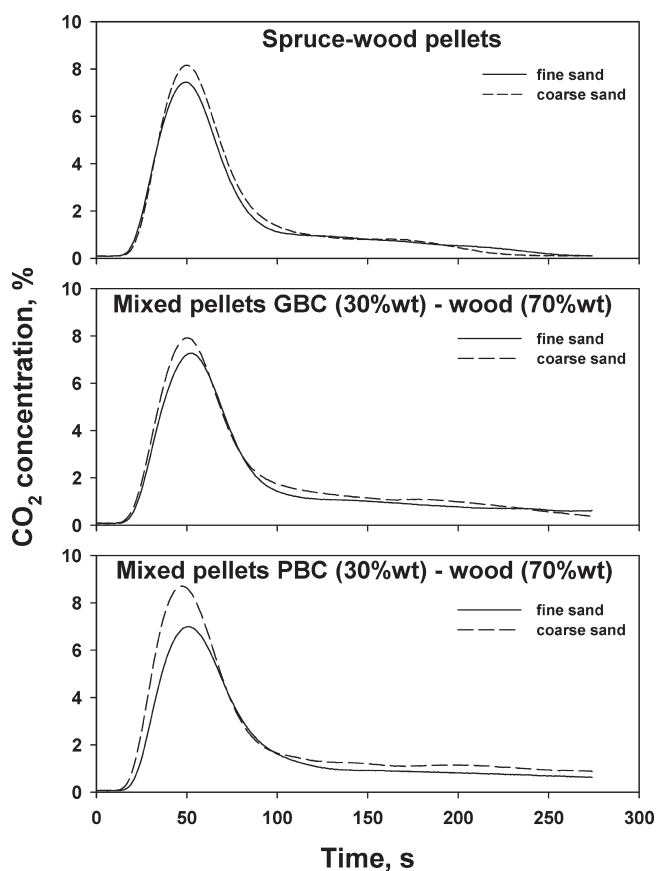
Figure 2. CO_2 concentration profiles during devolatilization tests in air of single fuel pellets (length =14 mm) with different inert bed particle sizes: fine sand, 0.2−0.4 mm; coarse sand, 0.4−0.8 mm. $T = 800$ °C, and $U = 0.3$ m/s.

tests. Attention was paid to two aspects known to be relevant with biomass fuels: the possible floating of the devolatilizing pellets on the top of the bed and the presence of volatiles burning in the splashing and freeboard sections. With regard to the first point, the pellets appeared at the top of the bed from time to time, indicating segregation in the upper zone of the bed. With respect to the second aspect, flames were noted at the bed surface for all fuel pellets, within the first 40−50 s from the particle injection. These flames issuing from the bed surface, even when the particle is immersed in the bed, are related to the volatile bubbles generated during devolatilization. The observed behavior is in line with previous findings by Fiorentino et al.[12] For biomass particles having similar size but significantly lower particle density, they reported the formation and eruption of 1−4 volatile bubbles and a flame extinction time of about 45 s.

Figure 2 reports typical CO_2 profiles obtained during the devolatilization tests for the three fuels using two different sand sizes, where $t = 0$ represents the time of pellet injection into the bed. A time delay of around 5−10 s was needed for inducing the release of the volatiles upon particle heat-up and drying. The CO_2 profiles are clearly characterized by two different stages: a first devolatilization stage, which results in a CO_2 peak caused by vigorous volatile emission, followed by the char combustion stage, displaying a moderate value of CO_2 slowly decreasing to the ambient value. The devolatilization behavior of the different fuel types with the two bed particle sizes appears to be very
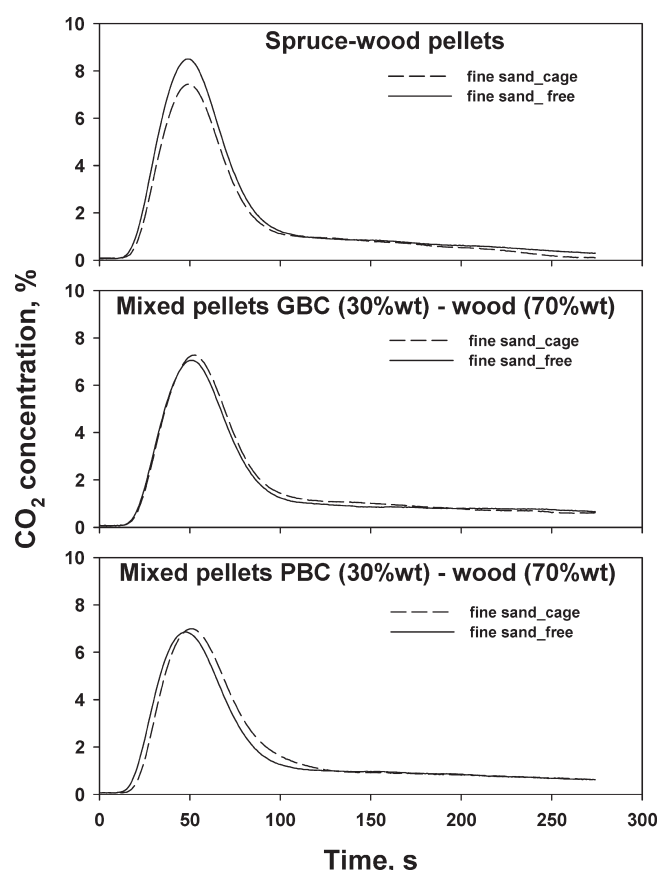
Figure 3. CO_2 concentration profiles during devolatilization tests in air of single fuel pellets (length = 14 mm) with different feeding method. Fine sand, 0.2–0.4 mm; T = 800 °C; and U = 0.3 m/s.

Table 2. Parameter Values Used for Estimating the Heating Time of a Single Wood Pellet[14,15]

heat-transfer coefficient	280 (for an average sand size of 0.3 mm)
($W\ m^{-2}\ K^{-1}$)	200 (for an average sand size of 0.67 mm)
thermal conductivity of a	0.26
wood pellet ($W\ m^{-1}\ K^{-1}$)	
specific heat capacity of a	1800
wood pellet ($J\ kg^{-1}\ K^{-1}$)	
density of a wood pellet	1300
($kg\ m^{-3}$)	
initial pellet radius (m)	3×10^{-3}

similar, despite the higher heat-transfer rate associated with the fine sand. The maximum CO_2 concentrations are moderate (about 8%). The devolatilization times (in the range of 90–100 s) and the char burn-out times (>300 s) are both relatively long. Interestingly, the times corresponding to the CO_2 peaks are similar to the observed times after which no more flames appear on the top of the bed.

Figure 3 reports the CO_2 profiles obtained during the devolatilization tests for the three fuels with the two alternative feeding methods (i.e., free pellet and pellet in the cage). Similar diagrams were obtained in both cases, indicating that the fact that the pellet is forced to remain inside the bed exerts only a limited influence on the devolatilization time, even in the presence of a more effective heat transfer.
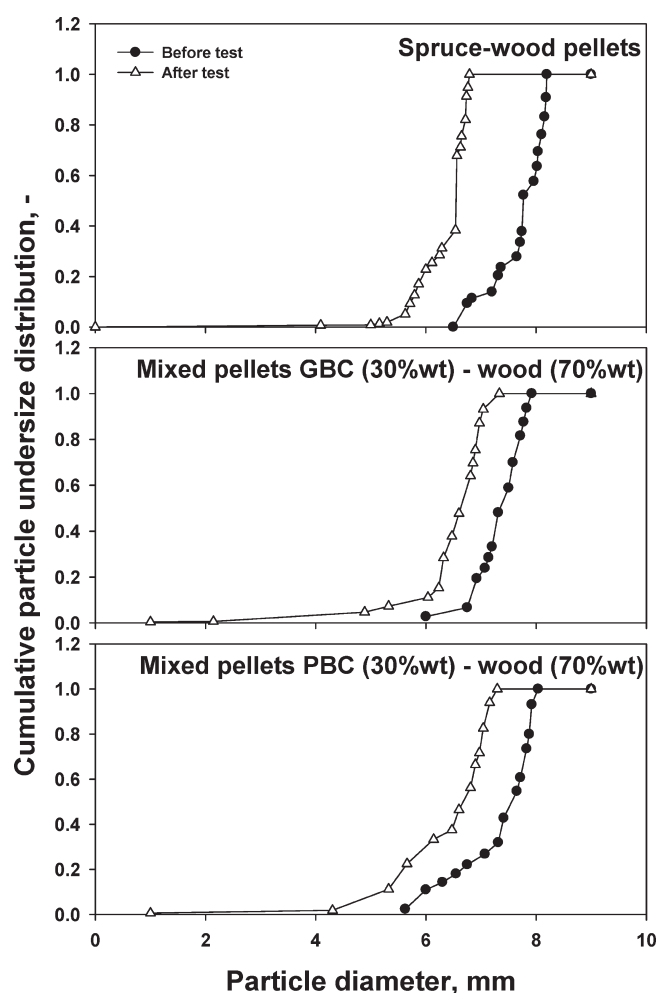
Figure 4. Particle size distribution of fuel pellets before and after primary fragmentation tests. T = 800 °C, and U = 0.3 m/s.
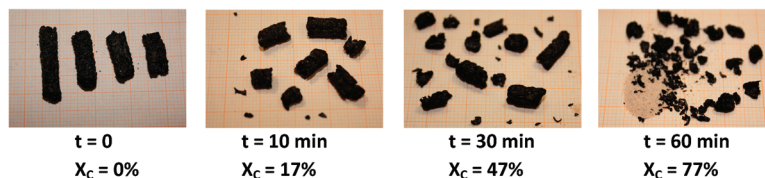
Table 3. Results of Pelletized Fuel Primary Fragmentation Tests

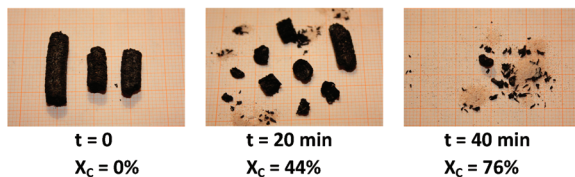| fuel	d_0 (mm)	S_f	n_1	d_1 (mm)
spruce wood pellets	6.00	0.29	1.38	4.92
70% wood/30% German coal pellets	7.35	0.21	2.47	6.37
70% wood/30% Polish coal pellets	7.30	0.30	3.30	6.24

These findings suggest that the external heat transfer has a limited influence on the devolatilization rate of the pellets, which is likely to be dominated by internal heat transfer. A simplified estimate of the devolatilization time of a pellet was made by solving the unsteady heat diffusion equation valid for an infinite cylinder of constant properties and size.[13] In particular, the time required to increase the axis temperature up to 0.9 times the bed temperature was calculated, assuming that both the heat of pyrolysis and the heat flux associated with the convective flow of volatiles could be neglected.[14] The values of the parameters used for the calculations[14,15] are reported in Table 2. Assuming typical values of the external heat-transfer rate, devolatilization times of 100 and 120 s were estimated for the fine and coarse sands, respectively. These values are close to the devolatilization times observed in the experiments (Figures 2 and 3).
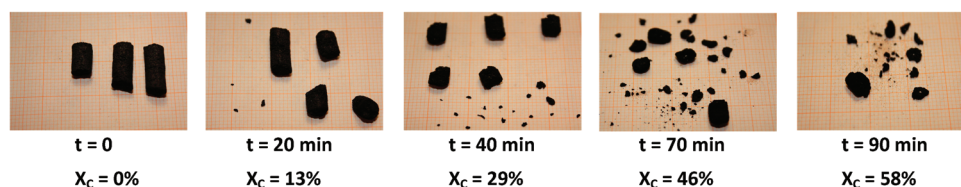
Char of spruce-wood pellets

t = 0	t = 10 min	t = 30 min	t = 60 min
X_C = 0%	X_C = 17%	X_C = 47%	X_C = 77%

Char of mixed pellets GBC (30%wt) - wood (70%wt)

t = 0	t = 20 min	t = 40 min
X_C = 0%	X_C = 44%	X_C = 76%

Char of mixed pellets PBC (30%wt) - wood (70%wt)

t = 0	t = 20 min	t = 40 min	t = 70 min	t = 90 min
X_C = 0%	X_C = 13%	X_C = 29%	X_C = 46%	X_C = 58%

Figure 5. Snapshots of char particles extracted at different gasification times during secondary fragmentation experiments (t, gasification time; X_C, carbon conversion degree). T = 800 °C, U = 0.3 m/s, and 60% CO_2.

Fragmentation Experiments. Primary fragmentation tests were carried out by monitoring the number and size of fragments produced by each fuel pellet upon FB devolatilization in nitrogen. Table 3 reports an overview of the results of primary fragmentation experiments for the three fuels. Results are expressed in terms of the following quantities: d_0, Sauter mean diameter of the initial pellets; S_f, primary fragmentation probability, i.e., the number of fuel pellets that undergo fragmentation divided by the total number of pellets fed; 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 of the three fuels, limited primary fragmentation occurs, with a fragmentation probability between 20 and 30% and particle multiplication factor in the range of 1.4−3.3. The slight decrease of the Sauter mean diameter upon devolatilization is caused by the combined effect of primary fragmentation and particle shrinkage.[3] These results are confirmed by data reported in Figure 4, showing the cumulative particle undersize distributions (on a mass basis) of the char pellets after devolatilization, as compared to the initial fuel pellet size distributions. A shift of the curves toward smaller sizes is clearly visible, but the population of smaller fragments (<5 mm) does not significantly increase, consistent with a limited particle fragmentation.

Secondary fragmentation experiments were carried out by injecting 3−4 char pellets in the bed of sand fluidized by nitrogen/CO_2 with an inlet carbon dioxide concentration of 60%. At preset time intervals, the fluidization gas was switched over to nitrogen to quench gasification. The char particles were then retrieved from the bed, photographed, and then re-injected in the bed for further conversion. A parallel CO gas concentration measurement at the exhaust enabled the calculation of carbon conversion during the run. Figure 5 summarizes the results of the tests for the three pellet types. It can be noted that,

for all of the fuel types, only limited secondary fragmentation occurs at carbon conversion below 40%, while extensive pellet shattering is evident at larger conversions. However, the gasification time needed to reach this conversion threshold is different for the three fuels, depending upon their reactivity and possibly the pelletization procedure. In particular, the German coal−wood mixed pellets appear to be the most reactive, and the Polish coal−wood mixed pellets appear to be the least reactive.

The extent of secondary fragmentation of the fuel pellets under gasification conditions appears to be more extensive than that reported for wood pellets under combustion conditions.[3] Two reasons are likely to explain this result. First, while char combustion is controlled by external diffusion of O_2, gasification is mostly controlled by intrinsic kinetics. As a consequence, in the latter case, internal reaction progressively weakens the structure inside the particle. Second, small fragments detaching from the particle surface are rapidly burned in the FB and, consequently, are not collected by the basket during combustion experiments. On the contrary, because the char reactivity toward CO_2 is much lower than toward O_2, during the gasification tests, conversion of the fragments in the bed is very limited.

Attrition Experiments. Figure 6 shows the rates of carbon fine 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, gasification (60% inlet CO_2 concentration), and combustion (4.5% inlet O_2 concentration) conditions are compared. The curves under inert conditions are similar to each other and follow the typical decreasing trend toward an asymptotic value.[8] However, the extent of fine attrition is different among the three chars. Spruce wood char appears to be harder than the other two, suggesting that the commercial pelletization
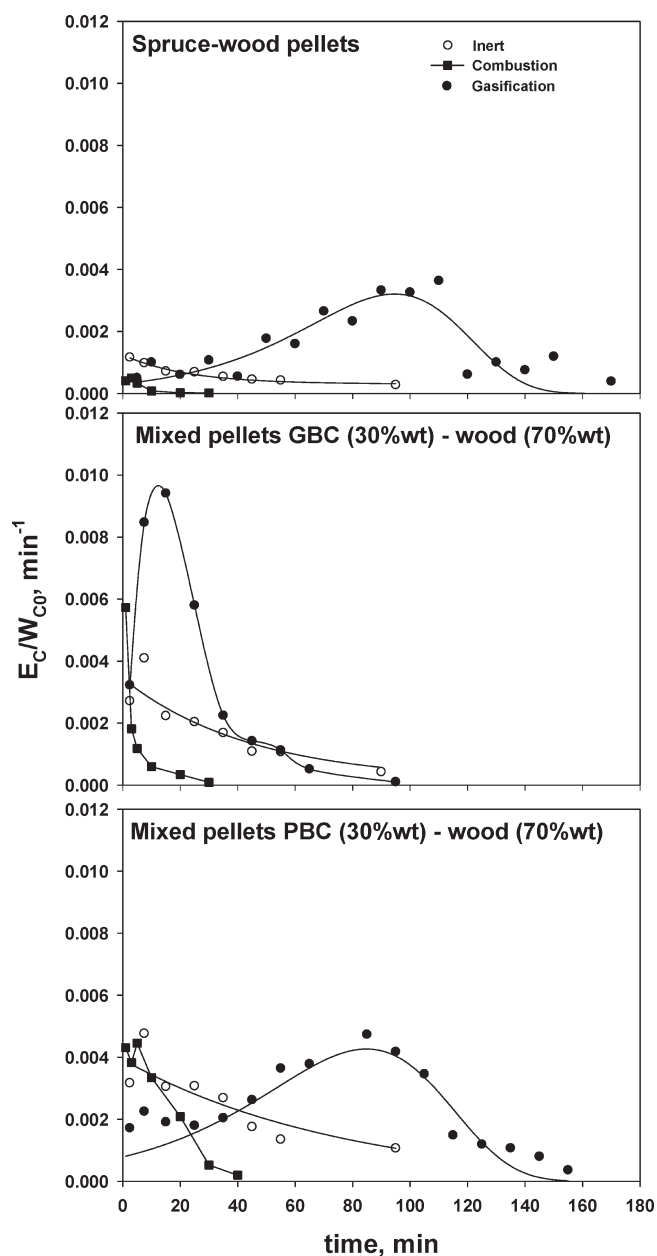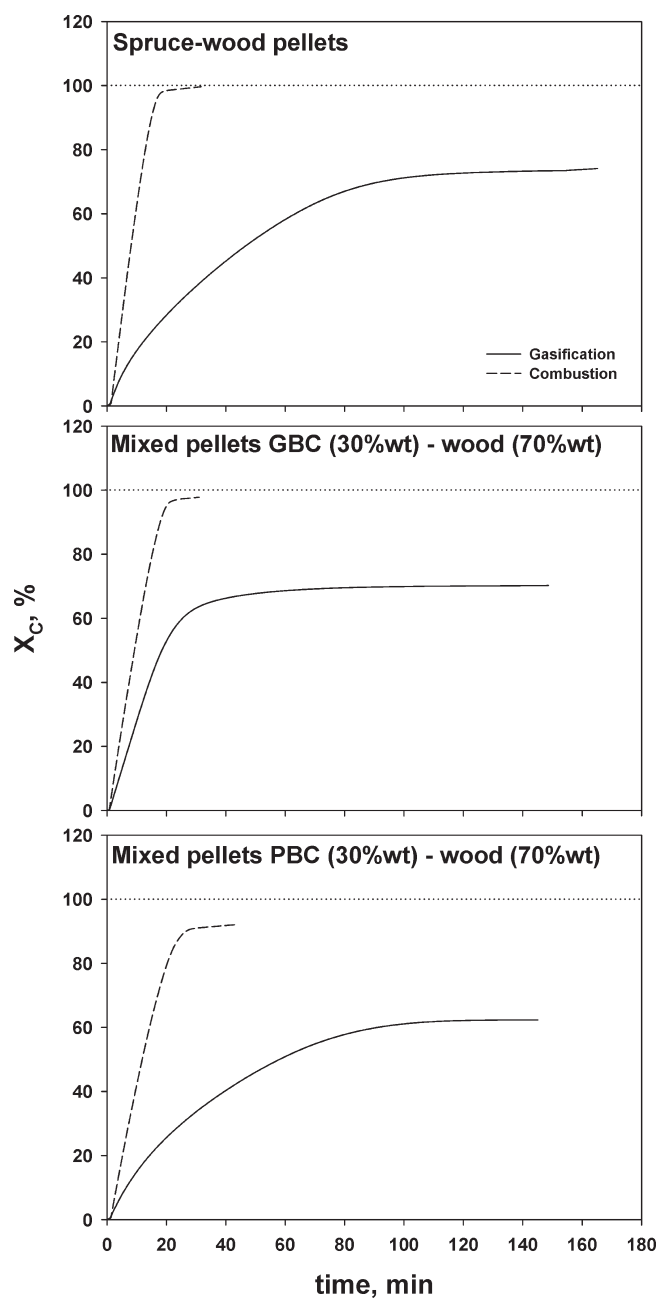
Figure 6. Results of fuel char attrition tests under inert (N_2), combustion (4.5% O_2), and gasification (60% CO_2) conditions. T = 800 °C, and U = 0.8 m/s.

procedure yields more compact pellets than the homemade pellets. This is reflected by the lower asymptotic value of the carbon elutriation rate obtained for spruce wood pellets (2.8 × 10^{-4} min^{-1}) with respect to GBC and PBC pellets (4.3 × 10^{-4} and 1.1 × 10^{-3} min^{-1}, respectively) and by the total amount of elutriated carbon during the runs (Table 4). The percentage of the initial carbon fed with the fuel char that was cumulatively elutriated during the 120 min runs in inert conditions was 7, 21 and 12% for the spruce wood, German coal—wood, and Polish coal—wood chars, respectively.

Under combustion conditions, results in Figure 6 show the typical combustion-assisted attrition scenario found with high reactivity fuels.[3,7] The elutriation rate significantly decreases under oxidizing conditions, because the increase of the attrition rate caused by combustion is effectively counterbalanced by extensive

Table 4. Percentage of the Initial Carbon Fed with the Fuel Char Cumulatively Elutriated during the Experiments

attrition conditions	spruce wood pellets	mixed pellets GBC (30 wt %)/wood (70 wt %)	mixed pellets PBC (30 wt %)/wood (70 wt %)
inert	7	21	12
gasification (CO_2 = 60 vol % in N_2)	26	29	38
combustion (O_2 = 4.5 vol % in N_2)	0.35	2.19	7.97

Figure 7. Carbon conversion degree during char attrition tests under combustion (4.5% O_2) and gasification (60% CO_2) conditions. T = 800 °C, and U = 0.8 m/s.

afterburning of attrited fines related to their large reactivity. As expected, this phenomenon is less evident for the Polish coal—wood char, which has the lower reactivity among the three fuels. In accordance with the curves reported in Figure 6, the percentage of the initial carbon fed with the fuel char that was cumulatively elutriated during the combustion runs (Table 4) was much lower than that found under inert conditions. It was 0.35, 2.19, and 7.97% for the spruce wood, German coal—wood, and Polish coal—wood chars, respectively. Curves of measured carbon conversion during the combustion experiments are reported in Figure 7, for reference.

A completely different scenario was obtained under gasification conditions. All of the curves in Figure 6 showed a significant peak in the carbon elutriation rate at some point during particle conversion. The presence of such a peak in the carbon elutriation rate indicates that carbon consumption in the pellets progressively weakens the char structure by pore enlargement, much like the combustion-assisted enhancement mechanism typically observed during FB combustion of coal.[8] In this case, however, fine post-conversion in the reactor is much less important, because the gasification reaction has a significantly lower rate than combustion. The decreasing part of the curves at the right of the peaks is 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 homemade 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. In fact, the gasification-assisted enhancement is dependent upon the parent fuel reactivity, because lower rates of carbon consumption shift toward longer times for the peak position. The percentage of the initial carbon fed with the fuel char that was cumulatively elutriated during the runs in gasification conditions (Table 4) was 26, 29, and 38% for the spruce wood, German coal—wood, and Polish coal—wood chars, respectively.

Figure 7 reports the related curves of carbon conversion during the gasification experiments (the curves do not end at 100% conversion because of the significant carbon loss as a result of elutriation). It is worth noting that 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. Coherently with the attrition and secondary fragmentation results, the German coal—wood char was the most reactive, while the Polish coal—wood char was the least.

These results 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 side, the higher reactivity and mechanical strength of the commercial spruce wood pellets appear to be somewhat beneficial for obtaining a larger carbon conversion during gasification.

■ CONCLUSIONS

In this study, the FB devolatilization/fragmentation/attrition behavior of different fuel pellets was investigated in a lab-scale apparatus. Three pelletized fuels, one based on wood and the other two based on a mixture of wood and coal, were characterized both under gasification conditions and for comparison under inert or combustion conditions. Similar devolatilization times were observed for the three types of pellets in the range of 90—100 s.

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, secondary fragmentation and attrition of carbon fines from the char particles during gasification were extensive, especially at large carbon conversions.

A gasification-assisted attrition mechanism was proposed to explain the experimental results, similar to the well-known combustion-assisted attrition patterns already documented for coals under oxidizing conditions. The low reactivity of the generated fines under gasification conditions makes the loss of carbon by fine elutriation much more significant than that typically found under combustion conditions.

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■ ACKNOWLEDGMENT

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