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PRODUCTION OF FINES DURING CO-COMBUSTION OF COAL WITH BIOMASS FUELS BY FRAGMENTATION AND ATTRITION

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Abstract -Results are reported from a project funded by the RFCS Programme of the European Union. The aim is to investigate, experimentally and by modeling, the production of fine char and ash particles during co-combustion of coal with wastes and biofuels in circulating fluidized bed. Work was undertaken at installations of different scales. Polish and Colombian coals were base fuels. The additional fuels were two sewage sludges. Bed temperature, feeding system, sand particle size, devolatilisation behaviour and char burn-out were studied to verify their influence on the fine particle production. Modeling was also carried out to understand the mechanisms of fragmentation and attrition. Samples from bed and cyclone were collected to determine particle size distributions.

Introduction

Co-combustion of wastes and biomass with coal contributes to reduction of the effective emissions of greenhouse gases and to recover the energy of wastes. Co-combustion works well, but the heterogeneous nature of the added fuels may have unexpected consequences. The research work undertaken focuses on the role of the particle size distribution in circulating fluidized bed (CFB) combustors, its dependence on size diminution by attrition and the possibly increased particle concentration and heat transfer in the riser, which in turn affects steam cycle operation and other processes. CFB is a suitable technology for co-combustion, but there are problems that remain to be overcome, such as bed particle management. The size distribution of the bed material can be affected by the properties of the added fuel. Even in mono-combustion problems related to the size distribution of the bed material occur: several recently erected CFB boilers had to be modified because of deviations from the desired particle size distribution, Muschelknautz and Muschelknautz (1999).

The operating conditions of a CFB combustor strongly influence the particle behaviour and have to be considered in order to predict the extent of diminution. During combustion particles are subjected to physical forces due to the continuous movement of the fuel particles in an inert bed material, leading to the production of fines by attrition. This situation could become even more complex during co-combustion due to the different nature of the fuels used jointly. Co-fired fuels often have higher moisture and volatile contents, more porous and fragile structure, lower density and greater intrinsic reactivity than the common base fuel, coal. A boiler operator needs to be informed about the behaviour of a particular fuel added to coal. It may be possible to grind the fuel further to minimise problems associated with combustion/attrition, but this is usually not a viable method with waste fuels.

Another problem related to attrition and production of fines is the danger of excess temperatures of fine char carried away with gases. Fine char particles from wastes and biofuels are more reactive than coal char by as much as three orders of magnitude. This may rise the particle temperatures to unexpectedly high levels, leading to melting, fouling, and agglomeration with bed solids. The present project deals with waste fuels and biomass, and their conversion behaviour during co-combustion with coal. Work was undertaken to characterise combustion, fragmentation, attrition and related particle behaviour. Below, modeling and experimental activities are reported from fluidized beds of different scales.

Experimental

Materials and equipment

Tests were conducted using four fuels (Table 1): two bituminous coals - Polish and Colombian (Carbocoal) - and two municipal sewage sludges - mechanically dewatered and dried granular sludge (Biopellets) and granulated sludge (Biogran).

UN (Naples) experiments were carried out in two electrically heated 41 mm ID stainless steel FBCs. The first combustor has the freeboard unlagged to minimize fines postcombustion. It has on-line analysis of O_2, CO and CO_2 concentrations at the exhaust. A stainless steel circular basket can be inserted from the top to retrieve particles from the bed. The fluidization column can be fitted with a two-exit head conveying flue gases alternately to removable sintered brass filters for collection of fines. The second combustor has a short freeboard to minimize loss of fines caused by adhesion onto reactor walls. Fly ashes are collected by a cyclone followed by a glass-fibre filter. The particle size-distribution of elutriated fines was characterized by a laser granulometer.

INETI (Portugal) experiments were carried out in a bubbling FBC, 1.5 m high and 0.08 m ID. The fuel was supplied by two screw-feeders, the first controlling the fuel feed rate and the second introducing the fuel into the bed. The stainless steel reactor was heated by an electric furnace, with temperature control. The elutriated particles were collected by two cyclones. The air distributor was a perforated plate with 12 injectors. An analyzer recorded continuously O_2, CO and CO_2 concentrations at the exhaust.

TUHH (Hamburg) tests were run in a pilot-scale CFB combustor, Klett et al. (2004). The riser has an inner diameter of 102 mm and is 15 m high. The combustor is equipped with a cyclone and an after-burner chamber. It is electrically heated from the walls to prevent heat losses. Emissions (O_2, CO, CO_2, NO_x, SO_2) were recorded at the cyclone outlet. Suction probes were used for solids sampling inside the riser and for solids mass flux measurements. In addition, a heat transfer probe sensed changes in bed-to-wall heat transfer.

CTH (Chalmers) tests were conducted in the 12 MW_{th} CFB boiler at Chalmers University of Technology. The plant and the equipment for feeding sludge have been presented by Åmand and Leckner (2004). Typical operating conditions for a CFB boiler were used: Bottom bed temperature 850 °C, excess air ratio 1.2, primary air 56% of total air, gas velocity top 5 m/s, furnace pressure drop 6 kPa, and sand mass mean diameter 0.3 mm.

Procedures

UN measured single particle combustion, primary fragmentation, elutriation rate and *primary ash particle size distribution* (PAPSD). The fuels investigated were sieved in two size ranges: 2.4-4.0 mm and 4.8-6.4 mm. All tests were carried out injecting the fuel into a bed of sand, kept at 850°C. The first two kinds of experiment used the basket set up, Chirone et al. (1991). Operation of the basket allowed to weigh, size and photograph the fuel particles at different fixed degrees of carbon conversion (single particle combustion experiments) and determine number and size of the fragments produced upon devolatilisation (primary fragmentation experiments). Elutriation rate experiments used the two-exit head for collection of elutriated fines in the filters that were weighed and analyzed for fixed carbon content. This procedure allowed time-resolved measurement of carbon elutriation rates, according to Chirone et al. (1991). The PAPSD of the fuels was measured according to a modified version of the procedure of Cammarota et al. (2001), based on batch operation of the combustor with a short freeboard.

INETI conducted devolatilisation and combustion tests. The fuels were sieved in two size ranges: 1.25-2.0 mm (devolatilisation) and 1.00-1.25 mm (combustion). In all tests the fuel was injected into a bed of sand kept at a temperature between 800°C and 900°C. Three sand sizes were used for devolatlisation tests: 0.3-0.4, 0.5-1.0, 1.0-1.3 mm. Only the smallest size range was employed for combustion studies. The fluidization velocity was 1.0 m/s in the devolatilisation tests and 0.5 m/s in combustion tests.

TUHH experiments were carried out firing Polish coal or dewatered and dried municipal sewage sludge (Biopellets, Table 1). The temperature in the riser was set to 850°C, the gas velocity at the riser's top was adjusted to 4 and 5 m/s, respectively. With an air ratio of 1.2 this leads to a fuel mass flow of 3 to 4 kg/h during coal combustion and to 4 to 7 kg/h in the case of sludge combustion. The coal was crushed to particle sizes below 5 mm. The initial bed material was quartz sand with a surface mean diameter of 150 μm. In order to keep a constant riser pressure drop of 7500 Pa, sand was added or ash was removed. The solids suction probes determined the local solids flow rate, and the heat transfer probe sensed changes in the bed-to-wall heat transfer, both at 6.2, 10.5 and 14.5 m above the distributor.

CTH fired Polish coal alone or together with municipal sewage sludge, (Biopellets, both mechanically dewatered and pre-dried). In co-combustion tests the dry mass fraction of sludge was varied up to 67%, corresponding to a fraction of energy from sludge of 46%. Apart from the co-firing tests, a reference case with pure coal was repeated with the coal sieved into a fine fraction and a coarse fraction, burned independently. The bed material was silica sand and no lime was added. Samples were taken at three locations in the boiler (0.6, 3.7 and 7.9 m above the bottom of the combustion chamber) to determine particle size distribution. Also gas concentration profiles using gas extraction probes were obtained.

Modeling Work

Models were developed by the groups in Naples, Hamburg and Ansaldo on different levels of sophistication and with different goals.

Ansaldo's goal was prediction of carbon conversion in CFB reactors on the basis of the residence time in the reactor of particles of different sizes, subject to chemical reaction and size reduction (e.g. combustion and comminution). Solids react according to a shrinking core model, with carbon consumed through reaction, leaving a layer of ash surrounding the unreacted core. The ash layer on the particle surface is eroded by abrasive attrition, forming fines that are not further attritable. The particle population is described by particle and core sizes, and the particle balance equations accommodate a population distribution, expressed as a function of these sizes. Solids in the exit stream, originating from mother particles with different size, staying in the reactor for different times, may have the same particle size and/or core size, while particles in the exit stream having the same size may have originated from mother particles with different size and may have different age, i.e. different unreacted core size. The exit particle and core size distributions are then computed by adding all contributions from different mother particles and ages to the same particle and core size fraction. The computation of particle size and core size distribution requires an analysis of the residence time in the reactor, i.e. a description of gas-solid mixing in the reactor through an adequate hydrodynamic model. This is considered by a simplified mixing model treating series-of-mixed-flows. The fraction of particles with a given particle and core size can be calculated by combining the single particle model and the residence-time distribution model. The carbon conversion efficiency is calculated from integration of data over all particle and core size fractions.

In Naples a model has been developed for quantitative assessment of the size distribution of the inert material in the bed, established at steady state in a CFBC. The model is based on the following assumptions:

1. The bed solids consist of fuel ash only, and no sorbent or inert material is added.
2. Primary Ash Particles (PAP) are liberated from the carbon matrix by the combined action of devolatilization, combustion and attrition of fuel and of its char. The size distribution of such particles, *PAPSD*, is an inherent property of a given fuel.
3. PAPs may further undergo attrition during their lifetime in the reactor. It is assumed that liberation of PAPs from the carbon matrix by fuel attrition takes place over a short time-scale compared to the average residence time of the bed material.
4. The ash attrition rate is proportional to the exposed surface area of the ash and to the superficial velocity of the excess gas. The ash attrition yields very fine particles, rapidly lost at the exhaust, which do not contribute to the ash hold up. The ash leaves the combustor either as bottom ash or as fly ash at the exhaust.

Two issues deserve to be mentioned because of their criticality:

- Axial mass flux/segregation profile of polydisperse fluidized suspensions in the transport regime: although a few studies are available in the literature (e.g., Nowak et al., 1997; Zhang et al., 1998), constitutive equations are lacking. An approximation has been adopted in the model to express the net solids flux, based on an elutriation equation according to Geldart (1981). The relevant gas superficial velocity has been assumed as that corresponding to the total (primary+secondary) air feed.
- The dependence of attrition rate on particle size and on superficial velocity of the gas is an open issue, and it is assumed that the velocity relevant to attrition is that due to primary air.

The model developed by the Hamburg group complements the work done in Naples by going a bit deeper into the details. For example, a distinction is made between attrition mechanisms acting on the particles in a bubbling fluidized bed, in the jet zone near the gas distributor, or during the passage through the cyclone. Furthermore, the stress history of an individual particle is taken into account, allowing a fresh particle that enters the system to have a higher rate of attrition than a particle which has already undergone attrition for some time, whose weak parts and edges are broken off. The population balance for the fluidized bed system consists of modules describing the CFB riser, the cyclone, and the return leg. Neglect of initial fragmentation of the fuel particles and attrition of the reacting particles simplifies the solution. Following the concept of the PAPSD (e.g. Cammarota et al., 2002), the ash particle size distribution resulting after burnoff of the carbon is taken as input to the model of the population balance. The model equations, including the fluid dynamics of the CFB and the cyclone, have been described by Klett et al. (2004).

Some Results

Attrition/conversion determines the fate of fuel particles and ash. Figure 1 shows the features of transformations: primary fragmentation of fuel (1 and 2), rate of char burnout and fines generation by char attrition (3), liberation of PAPs upon complete char burn-off (4 and 5) and subsequent attrition of the ash (6). The two sludges showed limited, if any, primary fragmentation upon devolatilisation, whereas the primary fragmentation of the two coals, especially the Columbian one, was significant. Sludge char combustion followed the shrinking core reaction pattern with a coherent skeleton of ash. Both coals burnt as shrinking particles.

The following observations were made regarding attrition of sludges:

---Under inert conditions the carbon elutriation rate has an initial peak due to rounding off of surface asperities. The rate becomes steady after about 5 min. A principle illustration is seen in Figure 2. This feature has been included in the models.

---Under oxidizing conditions the carbon elutriation rate becomes negligible soon after the combustion process starts. This results from two parallel phenomena. On the one hand, the unconverted carbon core shrinks as combustion proceeds, and accordingly the carbon content at the outer surface of the particle decreases. On the other hand, attrited carbon fines undergo extensive afterburning during their residence time prior to elutriation, favored by the high intrinsic combustion reactivity of both sludges.

---The ash attrition rate is small and the change of particle size is negligible during burn-out, at least under the fluidization conditions adopted here.

---The attrition of the two coals is consistent with that of other coals, Chirone et al. (1991).

Figure 3 compares the PAPSD measured with the four fuels. Both sludges (BP and BG) form only a minor quantity of fine primary ash particles: most particles have a size close to that of the parent fuel particles: ≅12% (BP) and ≅8% (BG) of ash material are less than 200 μm, about 65% (BP) and 80% (BG) are larger than 1 mm. The two coals have finer primary ash particles: about 20% of the particles are smaller than 100 μm. Despite of this behavior an enrichment of fine particles was found in the top region of the TUHH reactor during combustion of sludge at constant total pressure drop, Figure 4.

The average size distributions of the silica sand, coal and pre-dried sewage sludge used in the 12 MW boiler are plotted in Figure 5. The fuels are coarser than the silica sand and the question is how they influence the size distribution of the inert and char fractions in the combustion chamber. The curve for coal is an average. There was segregation in the coal hopper that made the size vary from test to test. Therefore special tests were carried out with coal sieved into large (7 mm) and small (1 mm) size fractions. Samples from the combustion chamber were sieved and the combustible part was determined. Figure 6A shows the inert part as function of height for coarse and fine coal. The mass mean particle size is coarser at the bottom than higher up in the furnace, where the size approaches that of the sand for both fuel fractions. More coarse ash builds up in the bottom bed with the coarse fraction, but the difference is larger for the combustible part of the sample, Figure 6B. The influence of sewage sludge as co-fuel is illustrated in Figure 6C and Figure 6D. As far as the dominant fraction of the bed material is concerned (the inert material) the influence of the sludge is less important than changes in particle size of the base fuel, coal. In the test with the highest co-firing fraction (67% on dry fuel basis) some impact of the sludge is seen, however. The mass mean size of the combustible fraction, Figure 6D, is more similar to the case of coarse coal combustion. There is no clear influence of the sludge.

Some results from the TUHH model are shown in Figure 7 where the modelled size distribution (lines) of the bottom bed material is seen to be fairly close to the measured data (dots) from the bed drain.

Conclusion

Some examples of results have been shown. The work is still in progress and the final work should contain a comparison of results from the partners. A good starting point is that the same fuels were used, but as indicated above, the fuels were in different state of dryness and in some cases they had to be reduced in size to fit the research equipment. However, such differences are minor, and it is believed that a unified conclusion regarding size distributions will be possible.

Acknowledgement

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Table 1 – Fuel Characteritics

	Polishcoal(PC)	Carbocoal(CC)	Biopellets(BP)	Biogran(BG)
LHV, MJ/kg	29.3	28.0	13.1	13.9
Proximate analysis, %wt (as received)				
Moisture	3.2	5.4	8.5	7.8
Ash	6.4	11.0	33.4	34.4
Volatile Matter	32.1	34.0	52.4	49.8
Fixed Carbon	58.3	49.6	5.7	8.0
Elemental analysis, %wt (dry base)				
C	82.1	70.1	34.1	34.7
H	4.6	4.9	5.2	4.8
N	1.2	4.3	4.6	4.6
S	0.5	0.9	1.2	0.9

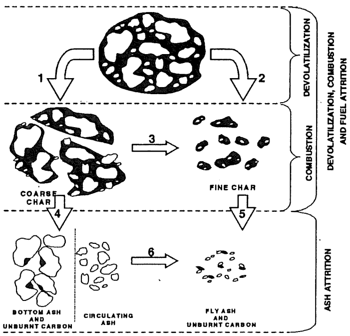

Figure 1. Outline of fuel and ash attrition during fluidized bed combustion (Cammarota et al., 2001)

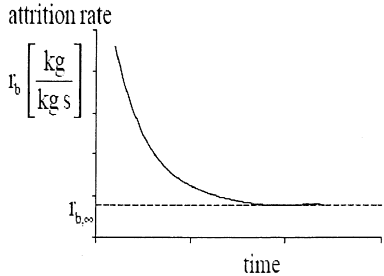

Figure 2. Initial attrition rate

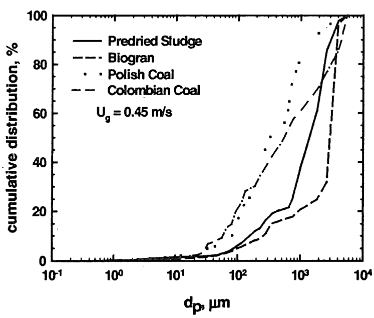

Figure 3. Primary Ash Particle Size Distribution (PAPSD) of the fuels tested

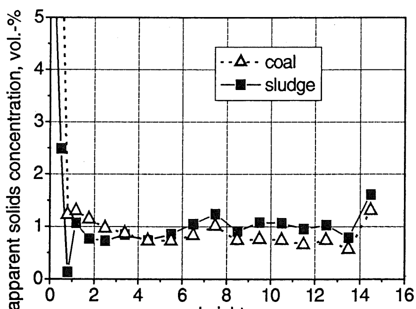

Figure 4. Vertical concentration in the TUHH unit. Total pressure drop is constant.

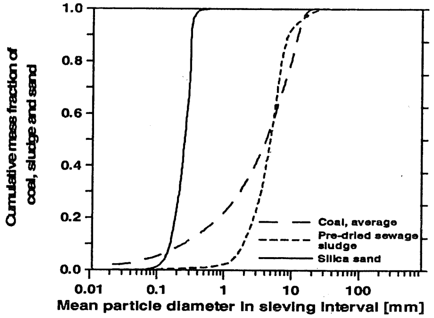

Figure 5. Average cumulative size distributions of silica sand, coal and pre-dried sewage sludge in the CTH boiler.

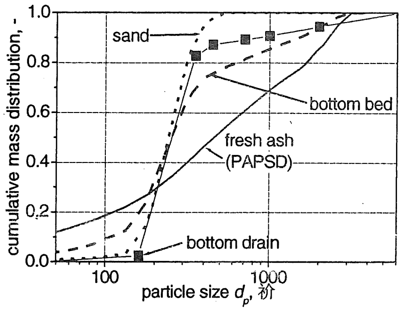

Figure 7. Steady state PSD in the TUHH unit. Measurement (dots), dashed lines calculated.

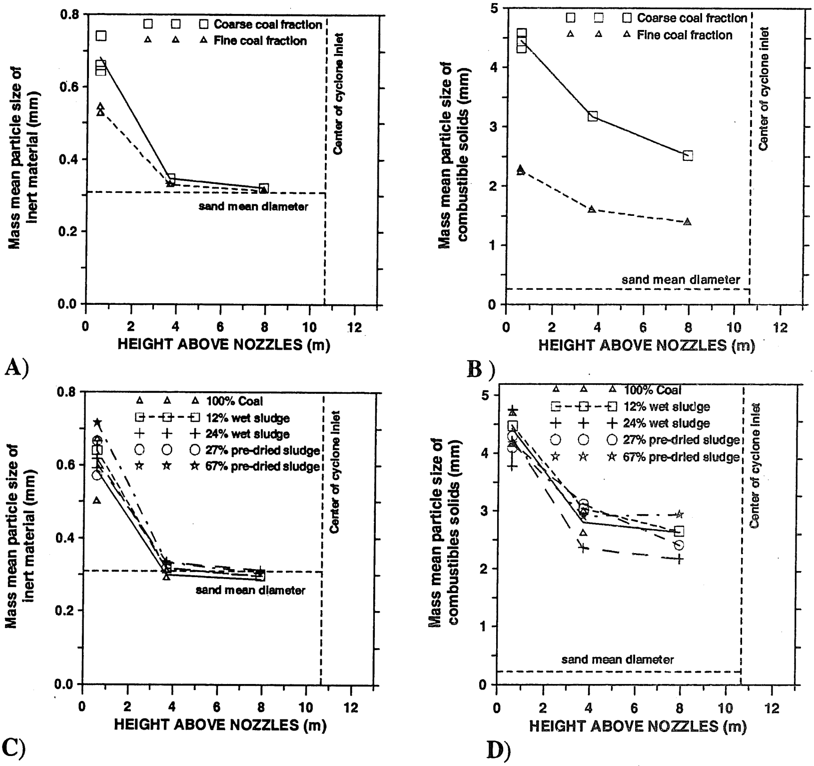

Figure 6. Mass mean particle size of the combustible fraction of the bed during coal combustion in the 12 MW boiler. A) Inert fraction using sieved coal. B) Combustible fraction using sieved coal. C) Inert fraction using the initial coal. D) Combustible fraction using the initial coal. Note that the vertical scale differs in the diagrams.