FLUIDIZED BED COMBUSTION: SOME AREAS OF DEVELOPMENT

by

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This paper describes a few selected topics within the field of development of fluidized bed combustion: desulfurization in pressurized applications, scale-up of combustors and influence of size of fuel.

Introduction

Fluidized bed is a highly adaptable device and even in the limited field of combustion there are many different opportunities of design, dependent on energy supply and demand, and legislation and taxation policies in various countries. Therefore, subject to different requirements, fluidized combustors vary in type and application. Some typical examples are:

• the many, small 10–100 MW_{th} units intended for heat or heat and power production in industrial and district heating plants in the Nordic countries.

• the few, large circulating fluidized bed (CFB) units intended for utility production of electricity in single Rankine cycle (in France, Korea, USA and especially in Poland) and the pressurized combined cycle plants.

• plants for industrial and utility production of electricity using difficult coals (especially in China).

In these plants properties of fuel and environmental concern, although extremely different in different countries, have been the two principal factors which that have influenced the development of plants. Here, only a few examples of development issues can be touched. Those selected are desulfurization in pressurized plants, scale-up of combustors in general (not only pressurized) and the importance of the size of fuel.

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Desulfurization under pressured conditions

The use of limestone for in-furnace desulfurization is one of the advantages of fluidized bed combustion (FBC). The goal is to reduce limestone consumption for a given degree of desulfurization and, in the combined cycle application, which will be treated here, to avoid calcium sulfide (CaS) in the effluent ashes. The most important features of desulfurization with limestone in atmospheric FBC have recently been summarized by Lyngfelt and Leckner (1999). In addition, during last years the desulfurization properties of limestone for use in pressurized FBC (PFBC) have been intensively studied. The behavior of uncalcined limestone in a PFBC, and sulfidation of limestone in a gasifier followed by sulfation and combustion of the remaining char have received attention. If this desulfurization, avoiding flue gas treatment, is successful, the advantage of using fluidized bed conversion in high efficiency coal conversion increases further.

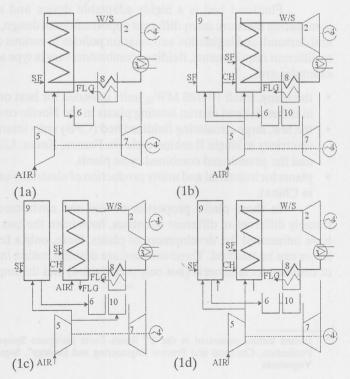
The PFBC is now well known, Fig. 1a. The principal drawback of this conversion method is the temperature limitation of the bed, giving a low (800 °C) gas turbine inlet temperature and consequently a limited efficiency for electricity production. However, there are several combinations of gasifiers and combustors having the purpose of enhancing the efficiency of coal utilization (which is considered to be one way to reduce the amount of CO₂ emitted per kWh of electricity produced). During last South-East European Symposium Olsson (1997) described a few combinations of gasifier and combustor, proposed but not yet introduced commercially:

Figure 1. Various arrangements of pressurized fluidized bed combustion without (1a) or with gasifier (1b, c, d). (1a) PFBC, conventional layout. (1b) Same as 1a but with a gasifier (9) and a gas combustion chamber (10). (1c) Same as 1b but the combustor (1) is operated at atmospheric conditions, and it is not connected to the gas turbine (7).

(1d) Similar to 1b but the flue gases from the combustor (1) go to the gasifier (9) which in this case is the main conversion unit.

1 – combustor, 2 – steam turbine, 3 – condensor, 4 – generator, 5 – compressor, 6 – particle separator, cyclone in Fig. 1a and ceramic filter in Fig. 1b, c, d and, once it works properly, also, in 1a.

SF – Solid Fuel, FLG – Flue Gas, W/S – Water/Steam, CH – Char



- (1) Mild gasification of coal in a gasifier and combustion of the product gases in a gas turbine combustion chamber in order to rise the gas turbine inlet temperature. The char residue from the gasifier is burned in the PFBC, which also serves as a heat source for a steam cycle. The gas is to be cleaned in a high-temperature ceramic filter (Robertson, 1987; McClung *et al.*, 1993), Fig. 1b.
- (2) Similar to 1b but the char from the gasifier is burned in an independent atmospheric CFB combustor connected to a steam cycle (Daves *et al.*, 1995), Fig.1c.
- (3) Same as 1b but the flue gas from the PFBC is introduced into the gasifier, which in contrast to the case of 1b is the main conversion unit. The gas leaves the gasifier together with the produced gas and is cooled to about 500 °C before it is filtered, thus avoiding a not yet reliable high-temperature (800–900 °C) filter (6). Like in the other cases, the gas is burned in the gas-turbine combustion chamber to attain a temperature level suitable for the gas turbine (Mori, 1998; Ikeda *et al.*, 1999), Fig 1d.

Desulfurization is one of the critical processes in this application. Limestone or dolomite is introduced into the gasifier and into the combustor to capture the sulfur released from the fuel. The desulfurization process is limited by thermodynamic restrictions and by product layers formed in the particles by reactions. Furthermore, in a gasifier a few hundred ppm of the H_2S produced will not be reacted because of thermodynamics, but the rest of the H_2S reacts with calcined CaO or half-calcined dolomite, Fig 2.

In the gasifier the principal overall reactions are

$$CaCO_3 \rightarrow CaO + CO_2$$
 (calcination) (1)

$$CaCO_3 \cdot MgCO_3 \rightarrow CaCO_3 + MgO + CO_2$$
 (half-calcination) (2)

$$CaO + H_2S \rightarrow CaS + H_2O$$
 (sulfidation) (3)

$$CaCO_3 + H_2S \rightarrow CaS + H_2O + CO_2$$
 (direct sulfidation) (4)

The last reaction is only useful with half-calcined dolomite, so in general calcination is desired, and this means gasifier temperatures above 900 to 950 °C, depending on the CO_2 partial pressure.

In the combustor the conventional sulfur capture reactions will take place if limestone or dolomite is added. The CaS delivered from the gasification plant will be sulfated without release of SO_2 if the temperature is 9000 °C according to

$$CaS + 2O_2 \rightarrow CaSO_4$$
 (5)

At temperatures >9000 $^{\circ}$ C and low O_2 concentrations SO_2 can be released from the CaSO₄ formed

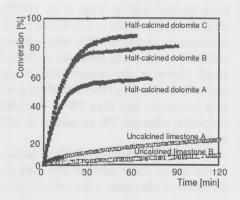


Figure 2. Conversion of CaS under gasification conditions, from Yrjas et al.

$$CaS + 3CaSO4 \rightarrow 4CaO + SO_2$$
 (6)

and perhaps also at still higher temperatures

$$CaS + 3/2 O_2 \rightarrow CaO + SO_2 \tag{7}$$

The high molar volume of CaSO₄ leads to formation of a product layer that prevents penetration of further oxygen to react with the core of CaS in a particle. Also, the exothermal reaction (5) may initially rise the temperature of the particle to favor reaction (6), Davies et al. (1994), but eventually a core of CaS may remain unreacted, surrounded by a shell of CaSO₄ and perhaps also CaO (the latter reacts to some extent with the SO₂ released. Abbasian *et al.* (1991) summarize their results regarding sulfation claiming that an optimum conversion from sulfide to sulfate is obtained in the temperature range of 815 to 9000 °C. Furthermore, the conversion is better for partially sulfided particles, *i.e.* the calcium to sulfur molar ratio (Ca/S) should be high in the gasifier. He also claims, in agreement with subsequent investigations, that the conversion to CaSO₄ is higher for dolomites having a high fraction of MgCO₃ (cf. differences between dolomites in Fig. 2).

The laboratory investigations indicate that it may be difficult to convert all CaS in the combustor at the same time as a low amount of wastes is produced from the process (low Ca/S and no MgO). Only a few pilot scale tests are reported in the literature. Foster Wheeler uses a gasifier at 925 °C and a PFBC at 870 °C. They attain a sulfur capture of 98.5% (probably at Ca/S = 2) using "proprietary sulfur capturing techniques" (Robertson et al., 1997), so it is not clear what was actually done. No information concerning the CaS content in the ashes was given. However, in a previous work, (Conn et al., 1995), a conversion of sulfide to sulfate of only 74 to 82 percent was attained. It was believed that the conversion would be higher in a larger unit.

A comprehensive test on combustion of CaS-containing char from a gasifier has been carried out by Sage *et al.* (1997). The atmospheric CFB combustor used was operated at 3 m/s, 900 to 970 °C and 6 to 37% excess air. Remarkably good desulfurization efficiencies of above 96% were obtained for Ca/S of about 2, and the combustion efficiency was equally good, above 99%. The influence of air supply and temperature was small. However, the requirement of complete CaS conversion was difficult to fulfil. In line with the results of the laboratory investigations, dolomite proved to be more favorable that limestone; less than 1% of the sulfur remained in the form of sulfide in tests with dolomite, whereas 7% remained in tests with limestone, especially in the coarse bed material.

The tendency of shells of particularly CaSO₄ to prevent the reaction of the CaS found in the inner parts of the lime particles seems to be the general reason for the difficulty of obtaining a complete reaction of the CaS. However, the reaction processes are complex and limestone types differ considerably from each-other, and more studies are needed to optimize limestone size and type for high CaS conversion. Moreover, it seems to be necessary to assess in more depth the environmental consequences for disposal of spent material containing CaS.

Scale-up

Are pilot plant results like those of Sage *et al.* (1997) valid for a large commersial plant? It is not possible to satisfy both chemical and fluid dynamic scaling criteria. Normally, fluid dynamic scaling is sacrificed carried out and chemical scaling is deemed to be satisfactory if the operating parameters are the same as in a large plant, and as stated by Sage *et al.* (1997) "the gas residence time (is) typical of full scale units". These criteria usually result in tall but narrow test reactors.

Usually the chemical scaling can be facilitated by choosing a number of important parameters equal in the test plant (II) and in the full scale plant (I): bed temperature, total excess-air ratio, primary stoichiometry, fuel and bed material. The fluidization velocity can be chosen in the same range in the two plants, supported by flue gas recirculation if necessary, to attain similar fluidization regimes (see discussion below).

Here the combustor Damköhler number Da (Damköhler, 1937) is proposed as a criterion to determine the combustion scaling. In general terms this number can be defined as

$$Da = transport time/reaction time$$
 (8)

Instead of the original four Damköhler numbers, here a vertical (Da_{ν}) and a horizontal (Da_h) number will be defined, describing vertical and horizontal conversion processes in the combustor.

In the vertical direction the transport time is the time required to transport the gases from bottom to the exit of the combustor, a distance H. Considering that similar fuel, bed material and temperature are maintainedkept the same, both inin the test plant and in the plant to be modeled, the reaction times should be identical (if mixing conditions are assumed to be similar) and only the transport times (residence times) need to be adjusted. The average gas residence times

$$t_{\text{max}} = H_{\text{I}}/u_{\text{I}} = H_{\text{II}}/u_{\text{II}} \tag{9}$$

yield a relationship between height H and fluidization velocity u,

$$u_{\rm II} = H_{\rm II} u_{\rm I} / H_{\rm I} \tag{10}$$

which is of importance for the choice of the height of the test plant $H_{\rm II}$ as long as ${\rm Da}_{\nu} < 1$. This is what was indicated by Sage *et al.* (1997) and explicitly stated by Knöbig *et al.* (1998). Knöbig and coworkers also proposed to adjust the average solids concentration in the combustion chambers to become $\bar{c}_{s {\rm II}} = \bar{c}_{s {\rm I}}$ in order to have similar vertical amounts of solids in the two reactors.

$$\Delta p = \rho_s g H c_s \tag{11}$$

which gives

$$\Delta p_{\rm II} = H_{\rm II} \, \Delta p_{\rm I} / H_{\rm I} \tag{12}$$

since the particle density is $\rho_{sI} = \rho_{sII}$ for the same type of bed material.

The two important reactions in the horizontal direction, devolatilization and char combustion take place as the fuel particles are dispersed from thea fuel feed point across the cross-section of the furnace. If the time needed for this transport is small compared to the conversion time, that is, if $Da_h < 1$, there will be an even distribution of fuel (volatiles and char) to meet the even distribution of air from the air distributor. If $Da_h \gg 1$ fuel and air will not be evenly distributed, and there will be a deficit of air on the fuel feed side of the combustor's cross-section and an air excess on the other side. Usually, the transport time is small in a narrow test unit but large in a wide commerscial plant. This causes an obvious problem of scaling, since the test plant can be tall to fulfill the height criterion, but it can never be wide.

Example

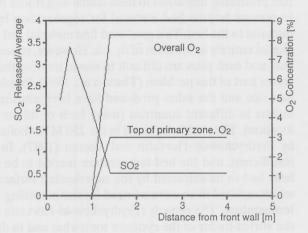
Sulfur capture is sensitive to the combustion conditions in a furnace. Thermodynamics reveal that, particularly in combustors operated at atmospheric pressure, the conversion of CaO and SO₂, as well as that of CaS, goes towards CaSO₄ if sufficient oxygen is present. Under oxygen-deficient conditions, which could occur locally even at overall air excess, the reverse reactions yielding SO₂ may take place, especially in the presence of CO and H₂. As shown by Lyngfelt and Leckner (1989), the "oxygen depletion theory" (Dennis and Hayhurst, 1985) produces the well known temperature dependence of sulfur capture (giving a maximum sulfur capture at 850 °C and a considerable decay of sulfur capture at higher temperatures), caused by an increasing combustion of gases in the particle phase (where the lime is) at increasing temperatures. This combustion removes oxygen and impedes sulfur capture. Even a release of already captured sulfur could occur (Lyngfelt and Leckner, 1998). This phenomenon takes place in a bubbling bed and should also be effective in the dense bottom bed of a CFB. The effect becomes worse when the distribution of fuel and air is imperfect.

As mentioned above, the experiments of Sage *et al.* (1997) yielded a surprisingly high sulfur capture at the high temperature employed (above 900 °C). The question was: Is this result representative also for a large plant? A tentative explanation is as follows: char was burned and char contains a negligible amount of volatiles. The char combustion is slow compared to volatile combustion and the fuel should have been perfectly distributed in the small 0.15 m diameter test plant, $Da_h < 1$. Most likely, there was an even distribution of fuel and air over the cross-section of the riser. In this case, under predominantly oxidizing conditions in the bed, sulfur capture is efficient also at higher temperatures than 850 °C. Such a result would then be representative for a larger plant as long as the Damköhler number for that plant does not exceed one too much. This could be the case for a plant burning char, but a similar situation would be more difficult to achieve for a plant burning volatile coal.

A number of different coals were investigated by Alliston and Wu (1996) in a test plant. Later they compared the results with desulfurization using the same limestones and the same fuels in large commerscial plants. They found that for low volatile fuels the agreement between desulfurization in the test plant and the large plant was better than

for bituminous coals, which always showed a higher desulfurization efficiency in the small plant than in the large commerscial boilers. The behavior of the large plants can be tentatively explained by the measurement results quoted from Alliston and Wu (1996), Fig 3.

Figure 3. Release of SO₂ in the vicinity of a fuel feed point (at zero). Adapted from Alliston and Wu (1996)



This figure illustrates very well the scaling problem related to the horizontal distribution of fuel: in the vicinity of the fuel feed point at the side of the combustion chamber the oxygen is consumed by the volatiles. SO_2 is released from $CaCO_4$ present in the bed (originating from previously captured sulfur in oxidizing regions) and produces high SO_2 concentrations in this oxygen-starved region, leading to a reduction in the over-all sulfur capture efficiency.

The general conclusion is again: scaling can be achieved in the vertical direction, but it is not possible to obtain a reasonable scaling of the width of a commerscial plant for all fuels; a test plant is by necessity narrow (otherwise it is no longer a test plant).

Bed material management

After fragmentation, attrition and combustion the materials supplied to the bed produce a certain particle size distribution in the bed. This size distribution results in a corresponding vertical suspension density distribution, which affects heat transfer to the walls and determines bed temperature: if the size distribution becomes biased towards large particles, the particle suspension density in the heat transfer region decreases and the bed temperature increases.

This influence of bed material size was discovered soon after the introduction of CFB and has been clearly explained (Herbertz et al., 1989): the small particle side of the particle size, spectrum is determined by the fractional efficiency of the gas-particle

separator, whereas the coarse particle side of the spectrum is found in the bottom bed and has to be adjusted by bed drainage. Various types of bed material regeneration devices have been invented to remove the coarse particles and send back the fine fractions to the bed after regeneration to avoid loss of bed material.

Despite this general knowledge, problems have occurred after changing from a fuel producing fine ashes to fuels containing stones (Grief et al., 1997). The solution was to remove bottom bed material for regeneration by crushing and return the crushed material to the bed. This produced fine material and caused the ashes to leave the boiler almost entirely in the form of fly ash. However, stones could pile up on the bottom of the bed, and such piles are difficult to move: treating the fuel before it is fed into the boiler solves part of this problem (Tham et al., 1997). Problems related to the properties of the fuel size and the ashes produced have been encountered recently in many large CFB boilers in different countries (using fuels of different properties, characteristic of the location). The problems found in the 250 MWe boiler in Gardanne have been described by Frydrychowski-Horvatin and Vostan (1997). In this boiler the heat transfer was insufficient, and the bed temperature tended to be high. To control bed temperature, heat had to be extracted by the superheated surfaces in the external heat-exchangers, which resulted in an extreme need for steam cooling, in order to reach the desired steam temperature. The remedy (Frydrychowski-Horvatin and Vostan, 1997) was to lengthen the vortex-finder of the cyclones somewhat and to displace it into an eccentric position in the cyclone according to Muschelknautz and Muschelknautz (1996). These measures enhanced the cyclone efficiency on the small particle size side, the suspension density rose in the upper part of the furnace, the heat transfer in the furnace increased, less heat had to be removed in the external heat exchangers, and the operation of the boiler improved

The example given has already become a "classical" solution and has been applied to alleviate similar problems also in other boilers (Muschelknautz and Muschelknauz, 1999). From these examples it is evident that the situation can be handled by design measures. But is there enough knowledge for a conscious optimization of particle size? The following items are of interest in this context:

- (1) Size of fuel and limestone. Large fuel particles require less communition work and furthermore improve the distribution of fuel across the cross-section of the furnace, but more efforts have to be spent on bed material regeneration. Optimization of bed material size requires improved knowledge on fragmentation and attrition and on dispersion of fuel under actual boiler conditions. The present abundant information is mostly limited to laboratory conditions.
- (2) Segregation effects. The movement of large particles in a bed of small particles applied to fuel particles in a boiler bed and to separation of large particles from small particles in a classifier. Again, work has been done in this field, but the results are difficult to apply at the desired operation conditions.
- (3) Gas-particle separators. The vortex finder in cyclones has been a constant source of preoccupation, either it is too long and erodes or it is too short and does not perform well. Also, many different shapes have been proposed for separators. The square separator of Foster Wheeler (Hyppänen *et al.*, 1997) is remarkable. If the

efficiency is satisfactory in such a device, new possibilities arise to avoid the expensive cyclone. However, tools are needed for sensitivity analyses: numerical fluid dynamic calculations are suitable for this type of investigation. Single particle tracking gives some information, but the goal of development of computational techniques is to handle both particle-particle interactions and particle size distribution.

In conclusion, optimization of particle size (fuel, sorbent, bed) would relieve several secondary problems caused by local unmixedness in FBC boilers, Such an optimization needs adequate information on fuel and sorbent behavior inside the combustor and in the external particle separation devices.

Conclusion

During this decade there has been a dramatic increase of knowledge on FBC design as well as of detailed phenomena. However, too little scientific attention has been directed to the fluidized bed processes taking place on the scale of a ccombustion chambers of commercial boilers, and processes such as volatile combustion and fuel dispersion are not well known on this scale. The combustion chamber processes are important for the evaluation of the emission and combustion behavior of the boiler.

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