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# **FLUIDIZATION 2003**

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#### EMISSIONS FROM CO-COMBUSTION OF COAL, WOOD AND SLUDGE IN CFB

by

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#### ABSTRACT

This paper reports on gaseous emission from co-combustion of sewage sludge with coal or wood in circulating fluidised bed (CFB). Dried, or in some cases "wet" (mechanically dewatered), municipal sewage sludge was investigated. The fuels coal and wood were used in contrast to each other. The emissions of CO, NO, N<sub>2</sub>O, SO<sub>2</sub> and HCl were measured from various sludge fractions and air supply methods in two CFB combustion units: the 12 MW<sub>th</sub> boiler at Chalmers University of Technology and the laboratory scale unit at the Technical University Hamburg-Harburg. The two units were scaled according to approximate scaling rules and give comparable emissions. It was shown that the emissions were moderate despite the considerable quantities of fuel nitrogen and sulphur in the sludge. For commercially interesting sludge fractions (<25 % of the energy supply) the European emission limits could be fulfilled, except in some quite extreme cases: sulphur emission from co-combustion of sludge and wood (mostly because of an odd emission rule) and for chlorine, where the emission limits are not well established. In all cases fluidized bed combustion has shown itself worthy its reputation: if the sludge handling systems are adequate, there are no further problems related to fluidization.

## **INTRODUCTION**

Fluidized bed is suitable for combustion of various fuels and it has been applied for cocombustion already from its introduction. During recent years the possibility of replacing fossil fuels with biofuels and to utilize the energy content of wastes has become of interest. Then, it is important to predict the consequences of fuel mixing on boiler design and on harmful emissions. For this purpose the fuel properties are decisive: volatile content, major pollutant precursors (N, S, Cl), major ash components (alkali, Si, etc.), and trace elements, all have their particular effects. This study is concerned with co-combustion of municipal sewage sludge with other fuels, such as coal or wood waste. Both the fate of heavy metals, including emissions of gasified species, and the "conventional" gaseous emissions were studied. The latter emissions are the topic of the present report that to a large extent is based on an investigation made in co-operation with the University of Hamburg-Harburg [1]. The potential problem is that municipal sewage sludge contains large quantities of nitrogen and some sulphur that may give rise to emissions of sulphur and nitrogen oxides. It is therefore interesting to know the effects of various fuel mixtures on these emissions and how they can be influenced by fluidized bed combustion. The purpose of this work is to show the consequences of co-combustion on the emissions in fluidized bed and to investigate the effect of various measures on the emissions.

### EXPERIMENTAL BACKGROUND

The investigation was carried out in two plants, the 12  $MW_{th}$  circulating fluidized bed (CFB) combustor located at Chalmers Technical University (CTH) and the pilot scale CFB unit at

the Technical University Hamburg-Harburg (TUHH). A schematic sketch of the plants is given in Figure 1.



Figure 1. The CFB test facilities at CTH in Göteborg (left) and at TUHH in Hamburg (right): (1) combustion chamber, (2) cyclone, (3) particle return line, (4) bed material hopper, (5) particle seal, (6) heat exchanger, (7) windbox, (8) fuel feed, (9) primary air supply, (10) secondary air addition into combustion chamber, (11) secondary air addition after cyclone, (12) after-burner chamber, (13) probe for flue gas extraction.

The combustion chamber (1) of the CTH unit has a square cross-section of about 2.25  $m^2$  and a height of 13.6 m. Fuel is fed to the bottom of the combustion chamber through a fuel chute (8). The circulating solids are separated in the cyclone (2) and transported through the particle return leg (3), the loop seal (5) and the external heat exchanger (6) back into the combustion chamber. Primary combustion air (9) is supplied to the wind box (7) below the gas distributor, whereas secondary air may be added either into the combustion chamber (10) or downstream of the cyclone (11). The exit duct is refractory lined and serves as an afterburner chamber (12). The dimensions of the CTH unit are close to commercial scale, and the results obtained are transferable to industrial units. Investigations in the pilot scale unit at TUHH could complement and extend the boiler measurements. This unit consists of a cylindrical combustion chamber (1) with a diameter of 0.1 m (cross-section area 0.008  $m^2$ ) and a total height of 15 m. The fuel is fed into the dense bed of the CFB via a screw feeder (8). The afterburner (12) has a diameter of 0.3 m and a length of 4.25 m, giving a residence time of up to 8 s. For emission measurements, gas was withdrawn from a sampling port (13) at half of the length of the after-burner, resulting in a total gas residence time of about 2.7 s under the operating conditions applied.

Although the TUHH combustor is significantly smaller in diameter than the CTH boiler, it has been shown [2] that the emissions are practically the same as those from the CTH boiler,

if suitable similarity rules are obeyed. The similarity criteria can be summarized by the following conditions that should be approximately the same in both units:

- bed material, fuel and additive (for instance, limestone)
- gas residence time in the hot region
- fluidizing velocities
- riser pressure drop
- bed temperature

The time needed to transport the reactants for reaction compared to the reaction time is taken as a similarity criterion (a Damköhler number),

Da = transport time/reaction time (1)

In the vertical direction, the transport time is the time required for the vertical transport of the gases from the bottom, for instance, from the air inlet to the height H above the bottom, or to the top of the combustor  $H=H_o$ . In case of fly-char the corresponding height would be  $nH_o$  where n is the number of times the char particle of a given size passes the combustor.

Because of similar fuel, bed material and temperature in the test plant and in the boiler to be modeled, the reaction times are similar (if the mixing conditions are assumed to be similar) and only the transport times need to be considered. The average gas residence time

(2)

(3)

(4)

(5)

$$\tau = H_1 / u_1 = H_{11} / u_{11}$$

yields a relationship between height H and fluidization velocity u in the two plants, index I and II,

$$u_{11} = H_{11} u_1 / H_1$$

which is of importance for the choice of the height of the plant  $H_{oll}$  if the transport time is smaller than the reaction time ( $Da_v < 1$ ). The adjustment of the fluidization velocity may appear to be in conflict with the requirement of equal total excess air ratios. However, this is not the case as long as the fuel mass flow can be adapted. This latter approach was used in the comparison between the plants. Also the average solids concentration in the test plant should be ajusted to become similar to that in the boiler  $\bar{c}_{sll} = \bar{c}_{sl}$  in order to have similar vertical distributions of solids in the two reactors. The measured total pressure drop of the riser can express this

$$\Delta \mathbf{p} = \boldsymbol{\rho}_{s} \mathbf{g} \mathbf{H}_{o} \mathbf{c}_{s}$$

which gives

$$\Delta p_{\rm ll} = \Delta p_{\rm l} H_{\rm oll} / H_{\rm ol}$$

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

The processes in the horizontal direction could not be scaled because the width of the two reactors was given and the same fuel was used. This had two consequences [2]: firstly, mixing was more efficient in the smaller reactor, and the overall progress of conversion was faster,

secondly, the impact of secondary air was more gradual in the larger furnace than in the narrow test reactor. These differences were relatively seen small and the final emissions were rather similar. The second effect mentioned (that of secondary air) was eliminated during the present tests, because all air to the furnace was supplied through the primary air nozzles and the remaining air downstream of the cyclone (Positions 9 and 11 in Figure 1).

The properties of the fuels are summarized in Table 1.

Table 1 The fuels	
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Fuel type	coal	wood (pellets)	sewage sludge A dried	sewage sludge B wet	sewage sludge C wet						
						Proximate analysis					
						Water (wt-%, raw)	9.0	8.1	19.0	73.0	76.6
Ash (wt-%, dry)	17.5	0.4	37.9	46.0	43.2						
Volatiles (wt-%, daf)	32.7	81.7	90.6	90.3	92.4						
Ultimate (wt-%, daf)											
С	84.9	50.2	53.2	52.1	49.7						
Н	5.0	6.1	7.1	7.1	8						
0	7.7	43.6	30.6	33.2	33.9						
S	0.7	0.01	1.9	1.6	1.5						
Ν	1.6	0.12	7.11	6.05	6.9						
Cl	0.08	0.002	0.05	0.09	0.08						
Lower heating value (MJ/kg)											
H <sub>u</sub> , raw	24.7	17.2	9.8	2.6	1.5						

daf = dry and ash free

The base fuels were either Polish coal or wood pellets. Pellets were used to as a homogeneous and well defined high-volatile fuel with low concentrations of nitrogen, chlorine and sulphur. The sludges were Swedish municipal sewage sludge (A), dried after digestion and burned in both plants, German (B) or Swedish (C) digested and mechanically de-watered municipal sewage sludges that can not be transported and therefore were used in the respective plants. The composition of the sludges is almost identical. One should particularly pay attention to the high nitrogen and sulfur contents of the sewage sludges.

The approximate operating conditions in both plants were as follows:

- Riser temperature was kept at 850 °C despite the wide variations of fuels
- Furnace air ratio was 1.0 with intended variations up to 1.2
- Total air ratio, 1.2
- Calcium to sulphur molar ratio, slightly above 2
- Total furnace pressure drop, 70 mbar

Air staging is a well-known measure to control NO. In conventional ("normal") air staging both primary and secondary air are supplied to the riser (furnace) of the CFB boiler (Position 10 in Figure 1). In advanced air staging, applied during the present tests, the second stage is located after the separation of the solid particles from the flue gas (Position 11 in Figure 1), while the whole riser is operated under near-stoichiometric conditions (Fig. 2).



Fig. 2. Comparison of normal (left) and advanced air staging (right).  $\lambda$  is air ratio.

By this method more air is supplied to the lower part of the combustion chamber than during normal staging. The increase of the amount of air to the bottom part is beneficial for sulphur capture with limestone. The oxygen is almost entirely consumed at the top of the riser section, and this has been proven to reduce  $N_2O$  emissions. Also the NO emission decreases. The effect of advanced air staging on emissions of SO<sub>2</sub>, N<sub>2</sub>O, NO and CO was first investigated for coal combustion [3].

The tests were run with either no staging (all air through the bottom nozzles) or advanced staging (stoichiometric conditions in the riser and addition of excess air downstream of the cyclone for burnout).

#### RESULTS

A comprehensive picture of the results is given by Figure 3, showing the influence of sludge addition on the NO emission for the different modes of air supply and different base fuels.



Figure 3. Comparison of advanced staging ( $\blacksquare$ ) and no-staging ( $\Delta$ ) during co-combustion in the TUHH combustor. (Concentrations in mg/m<sup>3</sup> under standard conditions, based on 6 vol %  $O_2$  and dry basis). The vertical scale is shortened.

The NO emissions range from 100 mg/m<sup>3</sup> to 1000 mg/m<sup>3</sup>, depending on the conditions. The upper left figure shows a case of addition of sludge with coal as "base fuel" and the lower left figure represents the same case but with wood replacing the coal. In the lower right diagram wood replaces the sludge in the upper left diagram and is burned together with coal. The upper right diagram shows results from the "wet" sludge. Because of the low heating value of wet sludge only a mass fraction of less than 0.4 could be run.

Starting with the cases without sludge (at a co-fuel ratio of zero) it is evident that the air supply method has a great impact when coal is involved: the difference between the runs with and without advanced staging is very large (all conditions but the air supply were unchanged). On the other hand, in the case of pure wood there is no impact of air staging at all. This situation prevails in the entire range of wood-sludge fractions (lower left diagram) and in fact, also, the more the sludge ratio increases, the less impact of air staging is seen. The conclusion is that the more volatiles that are present, the less important becomes air staging.

The influence of the fuel nitrogen content becomes evident when comparing the coal/dried sludge case (upper left diagram) with the coal/wood case (lower right diagram), in which the sludge was replaced with wood. The high nitrogen content of the sludge makes the NO emission increase as the fraction of sludge increases, whereas in the wood case, because of the low nitrogen content of wood, the NO emission decreases with increasing share of wood.

The wet sludge (upper right diagram) behaves in a different way compared to the dried sludge. An explanation is not yet found.



Figure 4. Comparison between emissions from the two plants (CTH - full lines and TUHHdashed lines) and between coal/sludge (\* and  $\Box$ ) and wood/sludge ( $\Delta$  and o).

The impact of sludge on the emissions presented in Figure 4 can be interpreted based on previous experience:

- The CO emission is low from CFB combustion of high-volatile fuels like wood and sludge. It is always higher when coal is involved under otherwise similar conditions because of production of CO during burn-out of char in the cyclone. The falling trend with increasing sludge fraction in the coal case supports this statement.
- The conversion of fuel nitrogen to NO is small, a typically favourable consequence of CFB combustion. The influence of fuel nitrogen is seen, however, and the NO emission increases with increasing sludge fraction, as already shown in Figure 3.
- The emission of N<sub>2</sub>O is small, surprisingly small considering the high nitrogen content of the sludge. This depends to some extent on the operation with advanced staging, but tests with all air from the bottom nozzles and 100% sludge resulted in an N<sub>2</sub>O emission of only 100 mg/m<sup>3</sup>, whereas the corresponding emission of NO was in the order of 1000 mg/m<sup>3</sup>. As shown before, the emission from pure wood is very small. In this case the increment as a consequence of sludge addition is evident.
- The sulphur emission increases with sludge addition, especially during co-combustion with wood. In all cases limestone was added in a Ca/S ratio slightly in excess of 2, so the conclusion is that sulphur capture with limestone becomes increasingly less efficient when the fraction of high-volatile fuels increases (both sludge and wood).
- A comparison with a recently introduced EU-directive regarding emissions from utility plants and waste combustors [4], allowing interpolation for co-combustion of wastes with conventional fuels, proves that the measured emission values satisfy the emission limits, especially if co-combustion is applied in small, industrially interesting energy ratios, say less than 25%. There is one odd exception, however. Co-combustion of sludge and wood will cause problems because the emission limit for sulphur emission from biofuel combustion is much more severe than for coal, and second, because desulphurisation with limestone is less efficient in connection to high-volatile fuels.
- The similarity between the two plants is not perfect but sufficient to allow the same conclusions to be drawn from the corresponding sets of measurement results.

The final gaseous emission to consider is that of chlorine, as well as entirely in the form of HCl, Figure 5. These emissions are strongly linked to the chlorine content of the fuels (Table 1) and behave accordingly: the chlorine content in the coal and the sludge is higher than that of the wood and the emission increases with sludge addition from the value for 100% wood, whereas the emission level is about constant in the coal/sludge case. The emission values and the data on the chlorine content in the fuels (Table 1) are not accurate enough to express any capture of chlorine in the lime or in the ashes, and hence, the emissions approximately correspond to the chlorine concentration in the fuels.



Figure 5. Emission of chlorine from the CTH boiler during sludge addition to coal or wood.

In Europe there are emission limits for HCl from waste combustion  $(10 \text{ mg/m}^3)$ , but at present there are no rules for conventional combustion units. Their limit is often established officially at the actual measured value during normal operation. If the limit for co-combustion with sludge is obtained by interpolation between the measured value for mono-combustion down to  $10 \text{ mg/m}^3$  for a waste combustion unit, it is easy to realise that co-combustion is impossible without flue gas cleaning. Fortunately, there are simple methods to reduce the chlorine emission, for instance, lime injection in the flue gas path in connection with bag filters.

## CONCLUSION

Fluidized bed is a very suitable combustion device for co-combustion of various fuels, especially coal, with sludge. Dried sludge can be fed into the combustor by a conventional feed system but wet sludge has to have a special feed system, preferably consisting of a cement pump and connection pipes into the furnace.

The high nitrogen content of sludge does not result in high conversion to NO and  $N_2O$ , especially not, if air supply to the furnace is adequately arranged; in the present case "advanced staging" was applied.

Sulphur can be captured with conventional limestone addition in the case of coal, but limestone addition proves to be less efficient with high-volatile fuels, such as wood. A reduction of the efficiency is also noticed during co-combustion with coal, when the fraction of sludge increases.

The chlorine emission corresponds to the chlorine contents of the fuels. Its value is moderate and comparable to that of coal, but if the plant is regarded a waste incinerator because of sludge addition, much more severe emission limits are applied. Then, even because of the contribution from coal, the emission limits may be difficult to obey without flue gas cleaning.

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