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PHOSPHOROUS IN ASH FROM CO-COMBUSTION OF MUNICIPAL SEWAGE SLUDGE WITH WOOD IN A CFB BOILER: A COMPARISON OF EXPERIMENTAL DATA WITH PREDICTIONS BY A THERMODYNAMIC EQUILIBRIUM MODEL

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ABSTRACT

Combustion tests have been carried out in a circulating fluidised bed (CFB) boiler co-firing wood and municipal, digested and mechanical dewatered sewage sludge from two Swedish wastewater treatment plants, using either iron sulphate or aluminium sulphate as precipitating chemicals. Experimental data on the concentration of various phosphorous compounds in the ashes are compared with theoretical equilibrium predictions. A clear relationship is not found between the employed precipitation chemical and the formation of phosphorous compounds in the ash. Hematite ($Fe_2O_3(s)$) is predicted to be the stable iron compound, and even in the sludge precipitated by iron sulphate, the aluminium content is sufficient to form aluminium phosphate ($AlPO_4(s)$) in similar amounts as in the sludge precipitated by aluminium sulphate. Lime addition to the bed gives a decreased formation of $AlPO_4(s)$ in favour of calcium orthophosphate ($Ca_3(PO_4)_2(s)$), which interferes with the sulphur capture efficiency of lime.

INTRODUCTION

In Sweden about one million ton of dewatered sewage sludge is produced annually. The sewage sludge contains large amounts of nutrition, for instance phosphorous, plus some metals and toxic organics. The nutrition originates from farms and is transported via grain, crops, milk and meat to the drains of the community. To retain the agriculture in a sustainable state it is necessary to recycle or by other means return the nutrition to the land, but without risk for enrichment of metals in the soil.

Deposition of sewage sludge is becoming more difficult because of stricter environmental demands on disposal sites. The first of January 2005 it will be forbidden to deposit sludge and other organic wastes in Sweden. Thermal processing of sewage sludge is a disposal method practised in several European countries [1]. Incineration results in reduction of the disposal volume, thermal destruction of toxic organics and recovery of the energy contained in the sludge. The quantities produced in most sewage treatment plants in Sweden are considerably smaller than what is normally burned in a power station. This is why co-combustion with, for instance, wood in existing plants could be a suitable technique. The phosphorous is accumulated in the ash and could be recovered, by leaching [2] or by other methods [3-4], none of which, however, has been commercialised.

The aim of the current work is to study co-combustion of sludge and wood with focus on the influence of precipitation chemicals and addition of lime on ash formation and desulphurisation efficiency. Identification of the specific phosphorous compounds formed is of interest for judging the performance of the leaching process.

Sludge contains higher concentrations of sulphur than wood pellets, and this leads to increased SO_2 emission, especially at high feed ratios of sludge [5]. The calcium content in the fuel ash binds sulphur and reduces the SO_2 emission, but other substances may interfere with the sulphur capture [6]. Data from ref. [7] show that the capture of sulphur by lime does not perform well during co-firing of sewage sludge with wood as base fuel. The SO_2 emission was almost equal to the theoretical SO_2 emission, no matter if lime was added to the bed or not. Two possible explanations were presented. The first suggests that lime is mainly present in the bottom of the combustion chamber, whereas the SO_2 from the high volatile fuels is formed higher up in the furnace and thereby does not have an opportunity to react with the lime. The second suggestion explains the poor sulphur capture by a parallel reaction, which deactivates the lime that is otherwise available for reaction with SO_2 [7]. Phosphorous could be involved in such a reaction [8].

The approach in the present work is to collect data and solid samples from measurements in a full-scale boiler and then perform equilibrium analyses on the system to determine elements in the ash.

EXPERIMENTAL BACKGROUND

The boiler

The boiler used in the experiments was the 12 MW_{th} circulating fluidized bed boiler (CFB), located at Chalmers University of Technology (Fig. 1.). The boiler is built for research but has all the features of a commercial unit. The combustion chamber (1) has a square cross-section of about 2.25 m^2 and a height of 13.6 m. The base fuel was fed to the bottom of the combustion chamber through a fuel chute (2). In order to feed mechanical dewatered sewage sludge a rebuilt cement pump was installed (21). Primary combustion air was supplied to the wind box (3) below the gas distributor, and secondary air was supplied on one level (4). The other secondary air ports (5-7) were not used. The circulating solids were separated in the primary cyclone (9) and transported through the particle return leg (10) and the loop seal (11) back into the combustion chamber. During combustion of only wood pellets, the circulating material also passed through the external heat exchanger (12). After the exit duct of the cyclone (8), the flue gases were cooled down to 150°C in the convection path and fly ash was separated from the flue gas, first in a secondary cyclone (13) and then in a bag filter (14).

Solid samples of the bed material were taken in two locations, (H2) and (10). Fly ash samples were taken both after the secondary cyclone (13) and the bag filter (14).

The fuels

The wood pellets were produced from domestic trees like pine and birch by AB Svensk Brikettenergi. The additional fuels were municipal sewage sludge from two wastewater treatment plants in Sweden, Ryaverket and Nolhagaverket. Both plants produce digested and mechanically dewatered sewage sludge but employ different precipitation chemicals for removal of phosphorous. Ryaverket is the second largest wastewater treatment plant in Sweden, taking care of wastewater from 775 000 inhabitants of the city of Göteborg and its surroundings. Iron sulphate ($Fe_2(SO_4)_3$) is employed for phosphorous removal. Nolhagaverket treats wastewater from 42 000 inhabitants in Alingsås and uses aluminium sulphate ($Al_2(SO_4)_3$) as precipitation chemical for phosphorous removal.

The content of moisture, combustibles and ash were analysed by a MAC 400 Proximate Analyzer 785-700 system. The main components were analysed by x-ray fluorescence and trace elements by ICP-MS (inductive coupled plasma with a mass spectrometer as detector) in an accredited laboratory. The properties of the fuels investigated are given in Table 1. The degree in trace element contamination of the sludge is shown in Table 2. Sludge from Ryaverket is somewhat more contaminated than sludge from Nolhagaverket.

The load during co-combustion should be equal to that during mono-combustion. The flow of sludge is, for that reason, increased with a higher rate than the flow of wood is decreased. The relative increase of ash, trace- and main elements with waste fraction depends partly on the higher content of these compounds in sludge compared to wood, and partly on the lower heating value of the sludges compared to wood.

The experimental procedure

The experimental procedure comprises four tests as can be seen in Table 3. In test A2 and B2 the fuel mix was wood pellets and sludge. No lime was added to the combustion chamber. The sludge originated from Ryaverket in test A2 and from Nolhagaverket in test B2. In tests C2 and D2:2 lime was added to the bed to investigate the effect on mass balances and emissions. The operating conditions of the tests are given in Table 4. The arrangements of the research boiler allow to keep the operating conditions similar in all tests, only changing the amount of sludge added and type of sludge. The operating conditions were chosen to represent a typical case of a CFB boiler with a bottom bed temperature of 850°C, an excess air ratio of 1.2, split of air supply into primary and secondary air, inlet of secondary air at 2.2 meter and a load corresponding to a fluidising velocity of 5 m/s at the top of the combustion chamber.

The thermodynamic modeling

To gain a fundamental understanding of the combustion chemistry, experimental investigations and advanced analyses in combination with equilibrium studies have been proven useful [9], [10] and [11]. An equilibrium analysis has its limitations and does not consider physical phenomena such as kinetics, mass transfer, particulates (aerosol formation, surface reactions, adsorption and absorption) or local conditions (temperature, residence time and mixing). The result depends not only on the quality of the thermodynamic data but also on the selection of input data and the elimination of compounds that are unlikely to form. The knowledge on the chemistry in combustion processes is not complete, and the databases available are limited. Hence, the results of a calculation should be regarded with caution. The calculation gives an estimation of the chemical composition and may clarify some of the influencing parameters on, for example, sulphur capture.

The chemical equilibrium calculations performed are based on the minimization of Gibbs free energy. The computer program FactSage and the module EQUILIB was used and the thermodynamic data was collected from the database F*A*C*T [12]. The program permits the determination of the chemical equilibrium state of a system that is defined with regard to temperature, pressure and total amounts of elements or compounds.

The purpose of this equilibrium study was partly to estimate the distribution of species with special focus on the fate of phosphorous and the interference on sulphur capture by lime, and partly to investigate the influence of the increased flow of aluminium and iron on the equilibrium composition. The identification of specific phosphorous compounds is of significance for the interpretation of the leaching results in ref. [2].

A schematic figure of the equilibrium reactor model representing the combustion chamber with a mean temperature of 850°C at atmospheric pressure is seen in Fig. 2. It is simple and should be regarded as a first step towards the prediction of the equilibrium composition in this system.

The model contains major- (C, H, N, S, Cl and O) and minor elements (K, Al, Si, Fe, Ca, Mg and P). Gas, pure solids and pure liquids are considered. At this stage no solution species are taken into account and N_2 is assumed to be inert.

RESULTS

The ash analysis

The closure of the mass balances in the reference case, only wood was burnt, and in the cases with mixed fuels is seen in Fig. 3. The closure is good or fairly good in the mixed cases. The higher value in the reference case can be explained by the low ash content in wood leading to a modest ash flow which introduces a large uncertainty in the ash balance. The same balances are carried out for all elements.

The measured flows of ash at different locations of the boiler are seen in Fig. 4. The bars “Inert in” are based on the solid flows (fuel ash, silica sand and lime) added to the boiler. The bars denoted “Inert out” are measured values of inert material leaving the boiler (fuel ash, silica sand and sulphur capture products e.g. $CaSO_4$). The following bars, “Bed ash” and ”Fly ash”, are simply the total ash flow divided into the two ash streams. The fly ash can be divided further into streams of secondary-cyclone and bag-filter ash (see Fig. 1., no. 13 and 14).

The main elements remain in the ash. Only a small fraction (>0.04 mole %) of potassium is found in the flue gas. The analysed concentrations of the main elements in the bottom bed sample are seen in Fig. 5. The concentrations reflect the fuel composition and the specific operational conditions in each test. The concentration of, for instance, iron is high in test A2 and C2 due to the precipitation chemical employed. Correspondingly, the concentration of aluminium is high for the tests B2 and D2:2. The lime addition is reflected in the concentration of calcium in tests C2 and D2:2. The concentration of silica in the bed ash is high, since silica sand is used as bed material.

Phosphorous, calcium and sulphur in ash

The thermodynamic data in commercialised data bases for phosphorous are criticized to be incorrect and insufficient. The equilibrium composition calculated should, for that reason, be regarded with care.

Phosphorous is found mainly as aluminium phosphate ($AlPO_4(s)$) and calcium orthophosphate ($Ca_3(PO_4)_2(s)$) in the ash at 850°C, see Fig. 6. Iron orthophosphate ($FePO_4(s)$) is not formed according to the model calculations, and since there is enough Al present even in the sludge precipitated by iron sulphate, $AlPO_4(s)$ is formed. The precipitation chemical employed does not seem to affect the formation of phosphorous compounds to any great extent. The lime addition on the other hand, seems to have a greater influence on the formation. More lime available leads to higher fractions of $Ca_3(PO_4)_2(s)$. There is an affinity between phosphorous and calcium. The amount of calcium available in the system affects the formation of phosphorous compounds. An increase of available calcium gives a decreased formation of $AlPO_4(s)$ in favour of $Ca_3(PO_4)_2(s)$.

Calcium is added to reduce sulphur emissions by capture of sulphur as calcium sulphate ($CaSO_4(s)$).

According to the calculation the compound will be found up to 700°C. At higher temperatures $Ca_3(PO_4)_2(s)$ is the preferred, stable compound.

Under the conditions of the tests, the equilibrium calculations reveal that available sulphur can form $CaSO_4(s)$ already at 175°C. At 600°C sulphur forms gaseous compounds, and the amount of $CaSO_4(s)$ decreases to be zero at 725°C. At 850°C calcium is found in the ash as $Ca_3(PO_4)_2(s)$ and sulphur in the flue gas.

The fate of precipitation chemical

Iron is found mainly as hematite ($Fe_2O_3(s)$) in the ash. The equilibrium composition of aluminium compounds has a more complicated character.

A comparison between the four tests at 850°C is seen in Fig. 7, which shows deviations due to both the precipitation chemical and the lime addition. The distribution of aluminium in test A2 shows that 18% is bound in mullite ($Al_6Si_2O_{13}(s)$), 28% in cordierite ($Mg_2Al4Si_5O_{18}(s)$), 41% in aluminium phosphate ($AlPO_4(s)$) and 13% is bound in potassium aluminosilicate ($KAlSi_2O_6(s)$). The effect of adding lime to the bed is shown by test C2. The halving of the $AlPO_4(s)$ formed to 19% confirms the decrease of the compound seen in Fig. 6 and is linked to the formation of $Ca_3(PO_4)_2(s)$ when more Ca is available. The formation of $Al_6Si_2O_{13}(s)$ and $Mg_2Al4Si_5O_{18}(s)$ are increased in this test, to 36% and 30% respectively, and $KAlSi_2O_6(s)$ is practically unchanged. Aluminium is more available in test B2 compared to test A2 where the highest share, 66%, is found as $Al_6Si_2O_{13}(s)$. The remaining Al is divided between $Mg_2Al4Si_5O_{18}(s)$ (9%), $AlPO_4(s)$ (19%) and $KAlSi_2O_6(s)$ (6%). Adding lime (test D2:2) increases the formation of $Al_6Si_2O_{13}(s)$ further to 84% and $AlPO_4(s)$ is decreased to less than 2%. The two remaining compounds are basically unchanged. Abundance of aluminium seems to favour the formation of $Al_6Si_2O_{13}(s)$.

DISCUSSION and CONCLUSIONS

The following conclusions can be drawn:

- All main elements in the input streams are found in the ash with exception of a small fraction of potassium.
- The equilibrium analysis did not show any clear relation between the precipitation chemical employed at the wastewater treatment plant and formation of phosphorous compounds in the ash. The reason for this is probably that the iron phosphate formed when using iron sulphate as a precipitation chemical is oxidised to hematite ($Fe_2O_3(s)$) when the sludge is burned. $Fe_2O_3(s)$ is calculated as the stable iron compound. The aluminium content in the sludge precipitated by iron sulphate from Ryaverket is sufficient to form $AlPO_4(s)$ in similar amounts as in the sludge precipitated by aluminium sulphate from Nolhagaverket.
- An increase of available calcium gave a decreased formation of $AlPO_4(s)$ in favour for $Ca_3(PO_4)_2(s)$.
- Calcium sulphate ($CaSO_4(s)$) is calculated to be the preferred compound up to 700°C but in the presence of phosphorous $Ca_3(PO_4)_2(s)$ is the stable calcium compound at higher temperatures. Even if P is removed in the calculations, calcium silicates are predicted instead of calcium sulphate, which contradicts experience on sulphur removal by lime.
- The predicted increase of $Ca_3(PO_4)_2(s)$ formation during addition of lime interferes with the sulphur capture.
- Iron is predicted mainly to occur as hematite ($Fe_2O_3(s)$) in the ash. The equilibrium composition of aluminium compounds has a more complicated character, and not only $AlPO_4(s)$ is formed.

ACKNOWLEDGEMENTS

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TABLES

Table 1. Fuel analysis.

Tests	Sewage sludge A2 and C2	Sewage sludge B2 and D2:2	Wood pellets A2 to D2:2
Presipitation chemical	$Fe_2(SO_4)_3$	$Al_2(SO_4)_3$	—
Proximate analysis			
Water (wt-%, raw)	72.0	77.8	8.0
Ash (wt-%, dry)	46.0	42.6	0.4
Combustibles(wt-%, dry)	54.0	57.4	99.6
Volatiles (wt-%, daf)	94.4	85.3	81.7
Ultimate analysis (wt-%, daf)			
C	52.6	50.2	50.3
H	7.2	7.3	6.1
N	5.4	5.0	43.5
S	1.4	1.2	0.01
Cl	0.1	0.1	0.09
O	33.3	36.2	0.01
Lower heating value (MJ/kg)			
Hu, daf	20.50	19.50	18.80
Hu, raw	2.78	2.24	17.20
Ash analysis (g/kg dry ash)			
K	13.3	10.7	82
Na	7.28	6.9	6.7
Al	73.3	193	12.2
Si	127	115	79.7
Fe	160	42.1	20.9
Ca	40.0	38.0	164
Mg	9.9	5.3	26.4
P	58.6	61.3	12.7
Ti	4.4	9.6	0.7

daf= dry and ash free, raw= as received

Table 2. Trace- and main elements analysis.

Tests	Sewage sludge A2 and C2	Sewage sludge B2 and D2:2	Wood pellets A2 to D2:2
Presipitation chemical	$Fe_2(SO_4)_3$	$Al_2(SO_4)_3$	—
Trace elements (mg/kg dry fuel)			
Hg	1.2	0.77	0.02
Cd	0.86	0.53	0.11
Pb	38	17	0.35
Cr	31	43	0.64
Cu	394	266	2.6
Mn	296	299	119
Co	6.9	2.8	0.04
Ni	21	13	0.20
As	5.9	5.2	0.12
Sb	13	15	0.06
V	23	23	0.06
Tl	9.4	10	0.06
Zn	652	385	15
Se	13	15	5.0
Main elements g/kg dry fuel			
K	6.6	4.9	0.50
Na	3.6	3.1	0.04
Al	35	88	0.08
Si	62	52	0.49
Fe	76	19	0.13
Ca	20	17	1.01
Mg	4.8	2.4	0.16
P	28	28	0.08
Ti	2.1	4.3	0.004

Trace elements and inorganic componens calculated on dry fuel. n.a.: not analysed

Table 3. Test matrix.

Tests	Type of sludge	Base fuel	Precipitation chemical	Lime addition
A2	sewage sludge	wood pellets	$Fe_2(SO_4)_3$	none
B2	sewage sludge	wood pellets	$Al_2(SO_4)_3$	none
C2	sewage sludge	wood pellets	$Fe_2(SO_4)_3$	to the bed
D2:2	sewage sludge	wood pellets	$Al_2(SO_4)_3$	to the bed

Table 4. Operating conditions.

	Wood/Sludge Ryaverket Test A2	*Wood/Sludge Nolhagaverket Test B2*	*Wood/Sludge Ryaverket Test C2*	*Wood/Sludge Nolhagaverket Test D2:2*	*Wood pellets*
Load, MW_{th}	6.1	6.0	6.0	6.4	6.5
Bed temp., °C (bottom)	848	843	852	850	849
Bed temp., °C (top)	866	880	852	856	861
Exit temp. of after burning chamber, °C	850	855	865	861	865
Total riser pressure drop, kPa	6.4	6.4	6.5	6.3	6.3
Calcium addition molar ratio Ca/S	0.0	0.0	1.7	2.7	0.0
Ca/S with Ca in fuel included	1.9	1.9	3.7	4.7	2.81
Excess air ratio	1.22	1.22	1.23	1.24	1.19
Primary air flow/total air flow,%	57	50	59	59	55
Superficial flue gas velocity at top of riser U_{top}, m/s	5.3	5.2	5.2	5.4	5.3

FIGURES

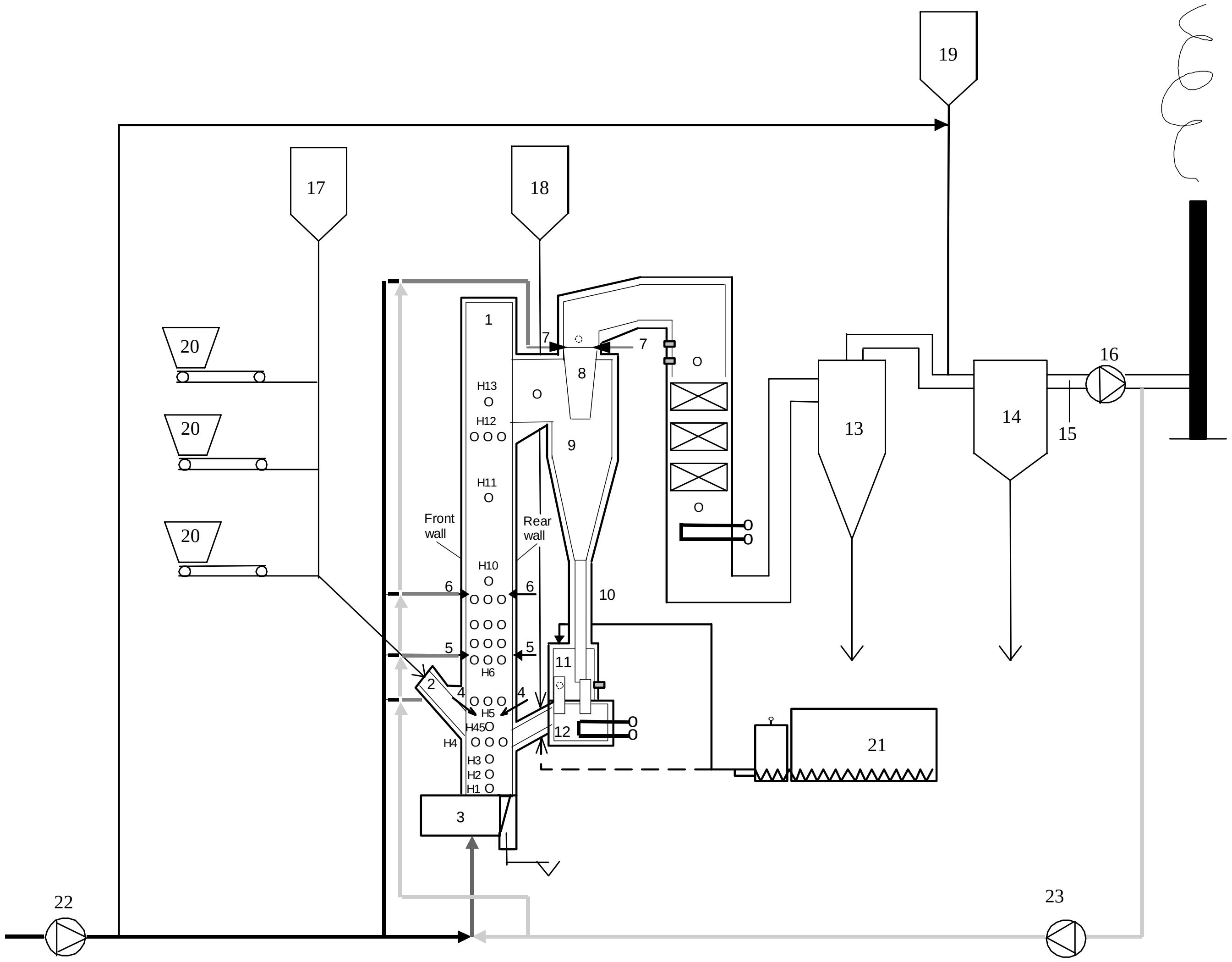

Figure 1. The 12-MW_{th} FBC boiler at Chalmers University of Technology. (1) combustion chamber, (2) fuel feed chute, (3) wind box, (4) secondary air inlet at 2.1m, (5) secondary air inlet at 3.7 m, (6) secondary air inlet at 5.4 m, (7) secondary air inlet into cyclone exit duct, (8) cyclone exit duct, (9) hot primary cyclone, (10) particle return leg, (11) particle seal, (12) heat exchanger, (13) cold secondary cyclone, (14) bag filter, (15) gas-extraction probe for emission monitoring, (16) flue gas fan, (17) sand bin, (18) lime bin, (19) hydrated lime bin, (20) fuel bunkers, (21) sludge pump (22) air fan, (23) flue gas recirculation fan.

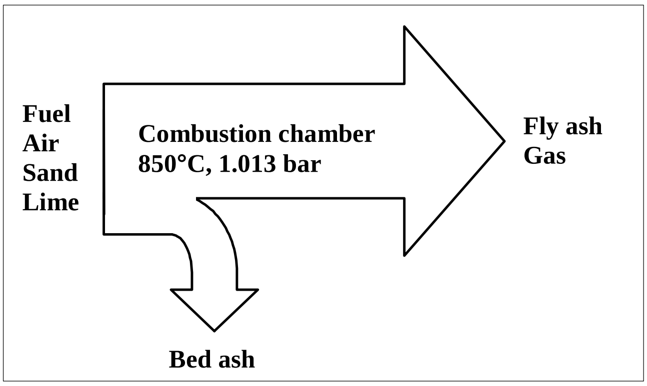

Figure 2. The equilibrium model predicting phosphorous-, aluminium- and iron compounds in the bed ash.

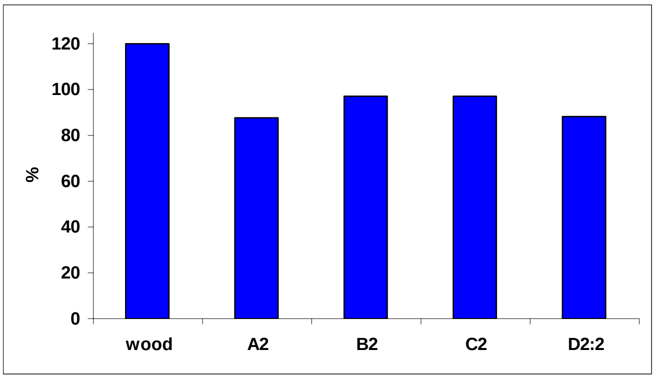

Figure 3. Mass balances for the reference case and the tests A2, B2, C2 and D2:2.

Figure 4. Measured ash flows for the reference case and the tests A2, B2, C2 and D2:2.

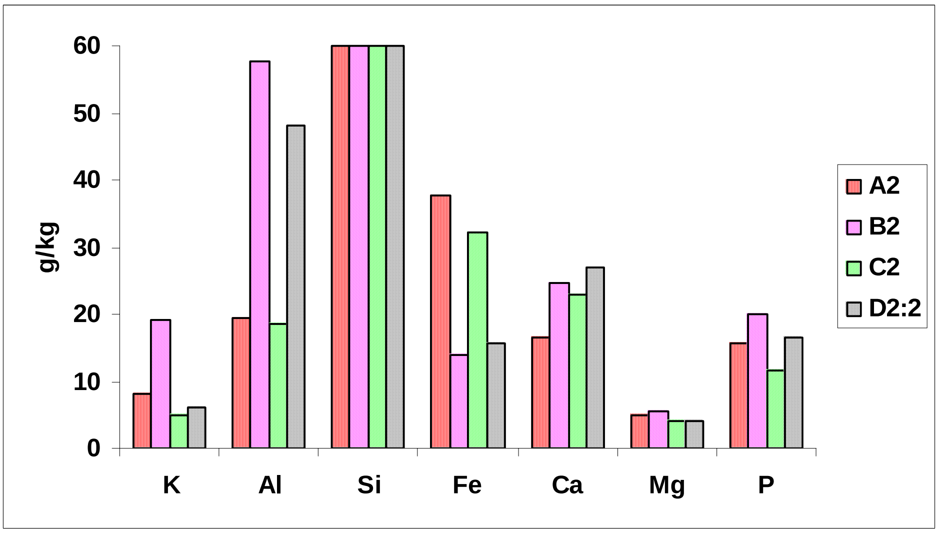

Figure 5. Analysed concentrations of main elements in bed ash from test A2, B2, C2 and D2:2. The concentration of silica is for tests A2 375, B2 336, C2 382 and D2:2 357 g/kg.

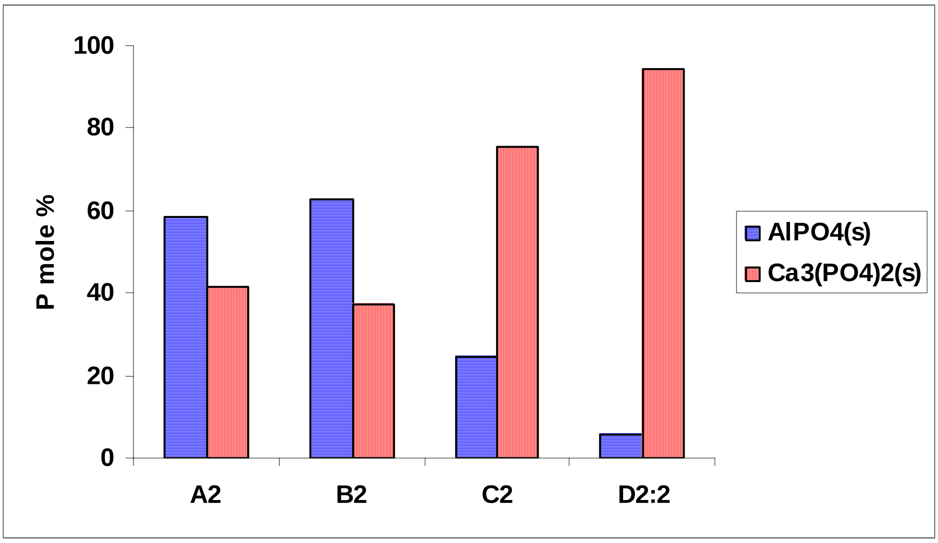

Figure 6. Predicted phosphorous species in bed ash from tests A2, B2, C2 and D2:2.

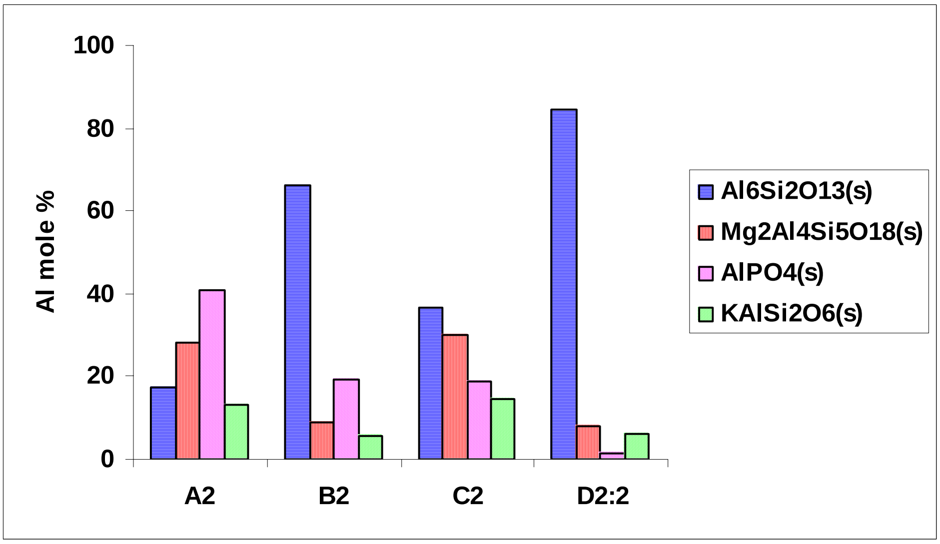

Figure 7. Predicted aluminium species in bed ash from A2, B2, C2 and D2:2.