Samförbränning av torkat avloppsslam med kol och trä i en panna med fluidiserad bädd

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Abstract:	This project deals with the emissions from co-combustion of sewage sludge with coal and wood in circulating fluidised bed (CFB) combustors. The increasing ash flows and the presence of trace elements in the flue gas and ashes leaving the boiler is also included in the study.
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Förord

Denna rapport utgör redovisningen av VA-Forsk projekt 20-108 med titeln: "Advanced air staging techniques to improve fuel flexibility, reliability and emissions in FB cocombustion". Projektet bygger på förbränningsförsök i fullskala med samförbränning av avloppsslam med kol eller trä i forskningspannan på Chalmers tekniska högskola. Denna förbränningsanläggning är av typen cirkulerande fluidiserad bädd. Resultaten jämförs med en motsvarande mätkampanj genomförda i en liten pilotanläggning vi Tekniska Universitet i Hamburg-Harburg. I huvudsak handlar rapporten om gasformiga emissioner till omgivningen som ett resultat av inblandningen av avloppsslam. De ökande askflödena som slaminblandning leder till samt förekomsten av tungmetaller i utgående askflöden behandlas också. Initiativet till projektet togs ursprungligen av VärmeForsk i samarbete med sin systerorganisation i tyskland VGB. Realistiska försök i fullskala är dock kostsamma och bidrag från Statens Energimyndighet samt från det aktuella VA-Forskprojektet har varit en förutsättning för att kunna slutföra den svenska delen av projektet. Redovisningen i sin helhet med en utökad sammanfattning på både engelska och tyska återfinns i VärmeForsk rapport 756. Denna rapport kan beställas via länken: http://www.varmeforsk.se/databas/databas index.html

Speciellt tack

Arbetet har genomförts i samarbete mellan Sverige och Tyskland finansierat av VGB PowerTech e.V./VGB Research Foundation och VärmeForsk Service AB. Den tysksvenska referensgruppen för detta projekt tackas speciellt för dess aktiva medverkan. Ledamöterna i den svenska referensgruppen har varit: G. Borgström (VärmeForsk), L. Hammar (Sydkraft), A. Kullendorff (Scandinavia Energy Project), M. Lundberg (Kvaerner Pulping), R. Lundborg (ÅF Energikonsult), G. Sfiris (Vattenfall), C. Tullin (Swedish National Testing and Research Institute). Ledamöterna i den tyska referensgruppen har varit: N. Heidenhof (Stadtwerke Duisburg), J. Jacobs (VGB), W. Köhler (VW Kraftwerke), L. Müller (VGB), B. Röper (RWE Rheinbraun), G. Rosenauer (Lenzing), M. Trost (Stadtwerke Flensburg).

Som ett komplement till huvudfinansieringen från VGB PowerTech e.v./VGB Research Foundation och VärmeForsk Service AB har den svenska delen av projektet erhålligt stöd från vatten och avloppsföreningens forskningsorgan VA-Forsk. För att kunna genomföra projektet vid forskningspannan på Chalmers kraftcentral har basanslag från Statens Energimyndighet utnyttjas.

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Bilaga: Advanced Air Staging Techniques to Improve Fuel Flexibility, Reliability and Emissions in Fluidized Bed Co-Combustion

SAMMANFATTNING

Denna rapport gäller ett projekt utfört gemensamt av tekniska universitetet i Hamburg– Harburg (TUHH) och Chalmers tekniska högskola (CTH). Projektet handlar om emissioner från sameldning av avloppsslam och kol eller trä i pannor med cirkulerande fluidiserad bädd (CFB). Följande resultat bör beaktas vid sameldning:

- Avloppsslammets höga askhalt resulterar i väsentligt ökade askflöden.
- Även om alkalikoncentrationerna är höga i avloppsslammets aska är koncentrationerna inte kritiska, och tendenser till beläggningar observerades inte.
- Spårämnesflödet ökar med ökande slamandel. Emissionen av metaller i rökgasen var emellertid väl under befintliga gränsvärden (EUs). Spårämnena tenderar att anrikas i flygaskan.
- I allmänhet kan mycket låga omvandlingsgrader av bränslekväve till NO och N₂O på 2–4 % uppnås.
- Med kol som basbränsle ger alternativ luftstegning, dvs. sekundärlufttillförsel efter partikelseparationen i cyklonen, till och med lägre NO emission än normal stegning utan att väsentligt påverka CO och SO₂ emissionerna. Alternativ luftstegning minskar dessutom N₂O emissionen. En luftfaktor i förbränningsrummet på 1,05 är optimal vid en total luftfaktor på 1,2.
- Med trä som basbränsle, dvs. för högflyktiga bränslen, påverkar luftstegning över huvud taget inte NO_x emissionerna.
- Mätningarna visar att en optimal gasuppehållstid för emissioner i CFB-pannor är 6 till 7 sekunder. Sådana uppehållstider kan uppnås tack vare CFB-pannornas stora cykloner, som delvis kanske kan ersätta en stor efterbrännkammare.
- En panna med cirkulerande fluidiserad bädd kan köras flexibelt med bränsleblandningar på upp till åtninstone 25 % slam (energiandel) utan att överskrida rådande emissionsgränser.

SUMMARY

This is a report from the joint project between Technical University of Hamburg– Harburg (TUHH) and Chalmers Technical University (Chalmers Tekniska Högskola – CTH) regarding emissions from co-combustion of sewage sludge and coal or wood in circulating fluidized bed (CFB) combustors. For operation under co-combustion the following results should be considered:

- The high ash content of the sewage sludge results in significantly increased ash flows.
- Although high alkali metal concentrations are found in the sewage sludge ash, no critical concentrations were reached and tendencies to fouling were not observed.
- The trace metal input rises with increased sludge fraction. However, emissions of metal compounds were well below legal limits. The trace metals tend to accumulate on the fly ash.
- In general, very low fuel nitrogen conversions to NO and N_2O of 2–4% are achievable.
- With coal as a base fuel alternative air staging with secondary air supply after solids separation attains even lower NO emissions than normal staging without strongly affecting CO and SO₂ emissions. Alternative staging also reduces N₂O emissions. An optimum for the excess air ratio in the riser of 1.05 was found for a total excess air ratio of 1.2.
- The higher the volatile content of the fuel is, the less effective the NO reduction due to air staging becomes.
- The measurements suggest that the optimum gas residence time regarding the emissions in CFB combustors is around 6 to 7 s. These times are achieved in commercial scale plants due to their large cyclones that perhaps partly can replace a large afterburner chamber.
- The circulating fluidized bed boiler can be operated in a very flexible way with various fuel mixtures up to an energy fraction of sludge of 25 % without exceeding legal emission limits.

PROJEKTRESULTAT

Fördelar och nackdelar med sameldning. Sameldning av avfall i pannor med cirkulerande fluidiserad bädd (CFB) som normalt använder konventionella bränslen (kol eller trä) har både ekonomiska och ekologiska fördelar. Användning av biomassa och avfall i sameldning med kol är ett bidrag till minskning av CO₂ utsläpp genom att fossila bränslen ersätts. Sameldning kan minska elproduktionskostnaderna och ökar därmed lönsamheten för ett kraftverk. Alltså, sameldning är ett miljövänligt och ekonomiskt fördelaktigt sätt att destruera avfall. Emellertid, beroende på typ och sammansättning av avfallet, finns det också några potentiella risker med sameldningen, t.ex. ökning av NO_x / SO_x emissionerna på grund av en ökad tillförsel av kväve och svavel i bränslet, ökning av både gas- och partikelformade tungmetallutsläpp och ökning av risken för deposition och korrosion p.g.a. större alkaliinnehåll i askan. Dessa aspekter med fokus på gasformiga utsläpp behandlas vidare i denna rapport.

Bränsleegenskaper. Undersökningen är inriktad på sameldning av torkat avloppsslam med kol eller trä som basbränslen. Några försök har också utförts med mekaniskt avvattnat (vått) avloppsslam och papperfiberslam. Typiska sammansättningar för de olika bränslena visas i Tabell 1. Avloppsslamm A är ett torkat slam från Himmerfjärdsverket utanför Södertälje och avloppsslamm B ett mekaniskt avvattnat slam från Hamburg. Avloppsslamm C består av medelvärden från 3 kommunala avloppsslam nämnda i referens [a]. Dessa värden kan därför anses vara representativa för ett allmänt rötat kommunalt avloppsslam. Anmärkningsvärt är det höga askinnehållet och de höga kväve och svavelkoncentrationerna, som kan ge upphov till höga svavel- och kväveoxidemissioner. Slammens undre värmevärde relaterat till torr askfri substans är omkring 21 MJ/kg medan

	Kol II	Trä II	Avlopps	Avlopps	Avlopps
			slam A	slam B	slam C
Överslagsanalys					
Vatten, % rå	8.7	9.1	15.9	76.6	76.0
Aska, % torr	15.7	0.8	42.1	43.2	45.1
Flykt, % torr&askfri	35.3	81.2	91.1	92.4	91.4
Elementeranalys, % to	rr&askfri				
С	82.5	50.5	53.2	49.7	52.4
Н	5.0	6.0	7.1	8.0	7.3
0	9.9	43.4	30.6	33.9	31.5
S	0.9	0.02	1.9	1.5	2.1
Ν	1.7	0.14	7.1	6.9	6.7
Cl	0.07	0.01	0.05	0.08	n.m.
Undre värmevärde, MJ	/kg				
H _u , torr&askfri	31.4	18.9	20.9	23.9	22.4
H _u , rå	24.2	16.8	9.4	1.5	1.1
rå=som levererat				n.m. = obel	kant

Tabell 1: Sammanfattning av typiska bränslesammansättningar

[a] *NO_x- und N₂O-Emissionen bei der Verbrennung von Klärschlämmen.* Arbeitsbericht des ATV/BDE/VKS-Fachausschusses 3.3, Korrespondenz Abwasser 7/96, 44, pp. 1260-1263

värmevärdet relaterat till råbränsle styrs av vatteninnehållet och kan variera mellan 2 MJ/kg för ett mekaniskt avvattnat slam med 65–70 % vatten till 10 MJ/kg för ett torkat slam med endast 10 % vatten. Slamaskornas sammansättning (Tabell 2) visar på höga halter av fosfor och kalium vilka kan återvinnas för användning som gödningsmedel.

Driftegenskaper. Undersökningar har utförts i två ganska olika CFB-anläggningar på CTH och TUHH. Men även om pannan på CTH är 200–300 gånger större i effekthänseende än forskningsanläggningen på TUHH stämmer emissionsdata mätta i de två anläggningarna väl överens. Försöken varade alltifrån en halv dag till flera dagar. Samma bränslen användes i de två anläggningarna (Tabell 1). Det visade sig att torkat slam kunde användas tillsammans med basbränslena utan tekniska problem. Sameldningen fungerade bra med de befintliga bränslehanteringssystemen och endast små skillnader i anläggningarnas egenskaper observerades trots slamtillsatser med upp till 50 % av energiinnehållet i tillfört bränsle. Detta förutsätter att askhanteringssystemen kan hantera de ökade ask-flödena som uppstår p.g.a. slammets askinnehåll som är omkring 45 % räknat på bränslets torrsubstans. Vid en energiandel på 50 % från slam blir askflödet nästan tre gånger det för ren kolförbränning och nästan 40 gånger det från ren träförbränning. På grund av askans höga järninnehåll är askan rödfärgad.

Askegenskaper, beläggningar och korrosion. Alkaliinnehållet (K, Na) i askan från avloppsslamm (Tabell 2) kan leda till misstankar om ökad beläggningstendens och korrosion på värmeöverförande ytor. Askan innehåller emellertid även ämnen med hög smältpunkt såsom Al₂O₃ och SiO₂ och totalt är askans smältpunkt omkring 1100–1200°C [a] vilket endast är något under en typisk smältpunkt för bituminöst kol som kan vara 1300°C. Alkaliinnehållet relaterat till eneriinnehållet i det ingående flödet når inte kritiska värden nämnda i litteraturen [b] och därför kan man anta att beläggningsproblem är osannolika. Korrosionsrisken på grund av HCl i gasen är inte högre än vid normal kolförbränning, eftersom klorinnehållet i de undersökta slammen inte är högre än i kolet. Därmed, och på grund av att inga tecken till beläggningar observerades under de relativt korta försök som genomfördes, kan man bara konstatera att inga tendenser till beläggningar observerades.

	kol II	trä II	slam A
Κ	557 ± 179	$448~\pm~5$	$1820~\pm~164$
Na	137 ± 6	90 ± 16	602 ± 45
Al	2767 ± 1097	178 ± 52	18400 ± 1341
Si	47667 ± 14434	1000 ± 0	48800 ± 1789
Fe	8167 ± 3175	568 ± 377	73800 ± 2863
Ca	5467 ± 1097	1275 ± 96	27200 ± 1643
Mg	$2366 ~\pm~ 404$	193 ± 19	$4180 ~\pm~ 295$
Р	1733 ± 1154	$315 \ \pm \ 182$	38800 ± 1644
Ti	300 ± 0	$2875 \ \pm \ 2249$	538 ± 84

Tabell 2:Asksammansättningenhos två basbränslen och torkat
avloppsslam. Koncentrationer i mg/kg torr aska

[[]b] J. Werther, M. Saenger, E.-U. Hartge, T. Ogada, Z. Siagi: *Combustion of agricultural residues*, Progress in Energy and Combustion Science (2000), 26, pp. 1-27



Figur 1: Gasemisioner från CTH pannan och TUHH anläggningen vid förbränning av rena bränslen (□ Polskt kol I CTH, ○ träpellets II CTH, ■ Polskt kol II TUHH,
träpellets II TUHH). Koncentrationerna är i mg per m³ vid standardförhållanden vid 6 vol-% O₂ och torr gas; λ_{total}=1.2, sekundärlufttillförsel efter cyklon. "N" är "normal" stegning λ_{primary}=0.7 med sek. lufttillförsel i förbränningsrummet.

Optimala luftstegningsförhållanden. En alternativ luftstegningsmetod har undersökts där sekundärluften tillsätts så sent som efter cyklonen. Jämfört med normal stegning med sekundärlufttillförsel i förbränningsrummet så har det visat sig att denna avancerade teknik ger ännu större förbättring av kväveoxidreduktionen än normal stegning vid förbränning av kol, utan att väsentligt påverka emissionen av CO eller svavelrening med kalksten i förbränningsrummet (Figur 1). Det optimala luftöverskottet i förbränningsrummet är omkring 1,05. Ytterligare minskning av lufttillförseln till förbränningsrummet, dvs. mild förgasning, tenderar att öka emissionerna igen.

Trots det högre kväveinnehållet relaterat till den brännbara delen av aloppsslammet, fem gånger högre motsvarande kväveinnehåll i kol, kunde kväveoxidemissionen hållas låg under sameldning med kol vid avancerad stegförbränning (Figur 2). Basbränslenas emissionsegenskaper spelar en avgörande roll, särskilt vid energiandelar slam under 25 %. Vid högre andelar, uppåt 50 %, börjar ett inflytande av slammets egenskaper att märkas, särskilt innehållen av flykt och kväve.



Figur 2: Inflytande av förbränningsrummets luftfaktor vid sameldning i TUHH anläggningen. Kol II/tort slam A (■ 100% kol II, □ 15% torrt slam A, ▲ 25 % torrt slam A, ● 50 % torrt slam A). "N" står för "normal" stegning. Övriga förhållanden som i Figur 1.

De flyktiga bränslebeståndsdelarnas betydelse. I motsats till kolfallet visar det sig att luftstegning i CFB inte spelar någon större roll för högflyktiga bränslen, såsom trä eller slam. Vid ren kolförbränning har luftstegning ett stort inflytande för minskning av kväveoxidutsläppen, men vid ökande andel av slam eller trä blir blir inverkan av stegning allt mindre (Figur 3). Skälet kan vara att koks har en betydelse för reduktion av kväveoxider (både NO och N₂O) i förbränningsrummet, och kokskoncentrationen i bädden blir liten vid förbränning av bränslen med hög andel flyktiga beståndsdelar (såsom biobränslen och slam). Syrekoncentrationen i bädden påverkar kokskoncentrationen, men då koks-koncentrationen är liten blir en ändring av kokskoncentrationen ändå inte så betydelsefull för NO koncentrationen så mycket som vid en hög koncentration då ändringen märks tydligare. Denna hypotes förklarar varför inflytandet av stegförbränning är litet vid sam-eldning med trä, eftersom både slammet och träbränslet endast ger små mängder koks.



Figur 3: Effekt av stegning vid sameldning (TUHH riggen. Koncentrationer som i Figur 1, λ_{total}=1.2, luftstegning med λ_{primary}=1.05 och sekundärluft efter cyklonen. ▲ ingen stegning, ■ advancerad stegning).

Både trä och kol är bra basbränslen, men p.g.a. deras olika flyktinnehåll kan man observera mindre skillnader i förbränningsförhållandena. De är:

• Reduktion av NO är bättre med kol än med trä (trots deras olika bränslekvävehalt). Som sagt ovan, för trä som basbränsle spelar arrangemanget för luftillförsel mycket mindere roll for NO-minskning än för kol.

- CO emissionerna är mindre än för kol (under förutsättning att eldstaden är korrekt dimensionerad och att driftförhållandena inte är orimliga).
- N₂O emissionerna är obetydliga vid träförbränning men väsentliga vid kol, men emissionerna i de två fallen blir mer lika då slamandelen ökar väsentligt.
- Svavelrening med kalksten är något mindre effektiv för trä än för kol som basbränsle.

De två basbränslena har sina fördelar och nackdelar, men i allmänhet kan sägas att båda är i stort sett lika bra som basbränslen.

Även om det torkade slammet A och det våta slammet B har en liknande sammansättning relaterat till torr och askfri substans och lika stor andel flyktiga beståndsdelar, så är inverkan av stegning något olika för de två typerna av slam: tillsats av torkat slam ger en stort sett oförändrad NO emission, medan vått slam visar en drastisk NO minskning i fallet utan stegning. Å andra sidan, vid stegad luftillförsel uppförde sig torkat och vått slam likartat vad beträffar NO emissionen medan N₂O emissionen var något högre för vått slam.



Figur 4: NO emissioner och bränslekvävets omvandlingtill NO eller N2O. Kol II/torrt slam A och trä/tort slam A, Förhållanden som i Figur 3 (TUHH: ■ basbränsle kol,
basbränsle trä; CTH: □ basbränsle kol, ○ basbränsle trä).

Omvandling av bränslekväve. Avloppsslam innehåller stora mängder kväve och höga kväveoxidemissioner kan uppstå, speciellt vid monoförbränning.Om allt bränslekväve i slam A skulle omvandlas till kväveoxid (NO) skulle emissionen bli 8130 ppm NO (16690 mg NO_2/m^3) medan en fullständig omvandling av bränslekvävet i det använda kolet skulle ge en emission på 1180 ppm NO (2420 mg NO_2/m^3). Emellertid är reduktionen i en CFB panna också hög, särskilt i närvaro av koks, och emissionerna är moderata vid energiandelar slam på mindre än 25 %; omvandlingen av bränslekvävet är då endast några procent. Omvandlingen till N₂O var också relativt låg (Figur 4).

Inflytandet av högre bränslekvävehalt syns tydligt i form av högre koncentrationer av NO och N₂O vid högre slamflöden.

Svavelrening. Avloppsslam innehåller svavel, som till stor del kommer från omvandling av proteiner men som till del är en återstod från svavel som tillsätts för slambehandlingen. Det aktuella avloppsslammet hade en svavelhalt som var dubbelt så hög som kolets. Svavelrening är alltså nödvändig. Vid samförbränning med kol i CFB är svavelrening p.g.a. slamtillsatsen inget problem, eftersom anläggningen högst sannolikt redan är utrustad med svavelrening genom kalktillsats, och svavelet från slammet innebär bara en marginell ökning av den totala svaveltillsatsen som hanteras med en likaledes marginell ökning av kalktillsatsen. I en träeldad panna, å andra sidan, finns det ingen utrustning för avsvavling, och svavlet måste avlägsnas från slammet före förbränningen eller genom installation av utrustning för avsvavling. Svavelrening med kalkstenstillsats till bädden är den klassiska metoden i fluidiserad bädd och den kan användas också vid trä som basbränsle, men föreliggande resultat visar att avsvaling med kalktillsats blir något mindre effektivt vid högflyktiga bränslen än för kol. Vad beträffar lufttillförsel så är drift utan sekundärluft mest fördelaktig om man bara beaktar emmissionsbeteendet vid avsvavling. (Lufttillförseln måste också beaktas i anknytning till optimering av tryckfall ock lastreglering.)

Spårämnen. De mest betydelsefulla spårämnena, som också är beaktade i utsläppsnormer, är Cd, Tl, Hg, Sb, As, Pb, Cr, Co, Cu, Mn, Ni och V.



Figur 5: Relativ ökning av de tillförda flödena av aska, Hg, Cr, Cu, V, Tl, Cd, Pb, Mn, Sb, As och Co som funktion av slamtillförseln vid kol som basbränsle. Observera att de vertikala axlarna har olika skalor.

Gasformiga såväl som partikelformiga emissioner av alla dessa ämnen har mätts. I allmänhet ökar tillflödet av tungmetallerna med ökande slamfraktion eftersom halterna är högre i slammet än i basbränslena (Figur 5). Kvicksilvertillförseln ökar med en faktor 15 om 50 % av bränslets energiinnehåll är slam jämfört med monoförbränning med kol. Relativt sett blir ökningen större jämfört med trä än jämfört med kol. I allmänhet finner man högre koncentrationer i flygaskan än i bottenaskan, men det beror på att bädden till stor del består av sand som visar mindre tendens att passera cyklonen i form av flygaska. Även de flyktiga metallerna tenderar att visa låga koncentrationer i bädden, såsom Hg, som inte kan detekteras alls i bädden, men i detta fall beror de låga koncentrationerna på att metallerna förflyktigats och följt med gasen.

Tungmetallemissionerna från CTH-pannan var under de gränsvärden som fastställts av den europeiska unionen även vid höga slamflöden och oberoende av basbränslet (Figur 6). Orsakerna till de goda resulteten är inte kända och bör undersökas ytterligare. Troligen kan den höga partikelbelastningen i CFB pannans gaskanaler ha haft en positiv inverkan på emissionerna av Hg och Cd.



Figur 6: Tungmetallemissioner jämförda med emissionsgränser föreslagna av den europeiska kommissionen för sameldning av slam A och trä (vänster) och bituminöst kol (höger).

Cyklonens och efterbrännkammarens betydelse. Förbränningsförloppen är ganska lika i de två försöksanläggningarnas brännkammare, särskilt som lufttillförseln till brännkammaren sker i botten, och sekundärluft endast tillförs nedströms brännkammaren där omblandningen är intensiv på grund av virvelbildningen i cyklonen, som ger gasen en kraftig rotation i utloppet från cyklonen. Av dessa skäl finns inga "makroskopiska" skillnader i omblandningen jämfört med dem som uppstår vid normal luftstegning, då blandningen är nästan instantan i den smala TUHH reaktorn, medan den endast sker gradvis i CTH pannan [c]. Det mesta tillförda syret förbrukas i förbränningsrummens bottendel och följdaktligen äger den största delen av förbränningen rum där. Även om mindre skillnader i förbränningsförloppen i de två eldstäderna observeras, jämnas dessa ut i cyklonerna och de resulterande emissionerna skiljer sig inte mycket. Slamtillförseln

[[]c] L.-E. Åmand, A. Lyngfelt, M. Karlsson, B. Leckner: Fuel Loading of a Fluidized Bed Combustor Burning Bituminous Coal, Peat or Wood Chips, Report A 97-221, CTH, revised Dec. 1999

påverkade de beskrivna förloppen obetydligt, men det fanns en klar inverkan av slamtillsatsen på kväveoxidkoncentrationsprofilerna i förbränningsrummen.

På grund av blandningsbegränsningar kan man ofta se höga koncentrationer av oförbrända gaser i de övre delarna av CFB pannors förbränningsrum. Naturligtvis är effekten tydlig i den vida CTH pannan, men den var synlig till och med i den smala TUHH reaktorn, och även vid förbränning av kol. När gaserna når cyklonen förbättras omblandningen och förbränningsintensiteten ökar. Utbränning erhålls om uppehållstiden vid hög temperatur är tillräcklig, och om cyklonen inte är tillräcklig behövs mer utrymme.



Figur 7: Emissioner mätta vid samförbränning av kol II med (a) torkat slam A och (b) vått slam B jämfört med gränser enligt utsläppsnormer; koncentration per m³ vid standard tillstånd torra gaser och glidande O₂ koncentration enligt EU direktiv (öppna symboler) och tyska normer (fyllda symboler); λ_{primary}=1.05, sekundärlufttillsats efter cyclonen (□■ TUHH anläggningen, ○● CTH anläggningen). Energiandelen vått slam B inkluderar stödbränsle.

Efter att ha utökat förbränningsutrymmet med efterförbrännare efter cyklonen fick de använda försöksanläggningarna en gasuppehållstid som är i samma storleksordning som i en stor kommersiell anläggning (Detta utfördes som en förberedelse för det aktuella projektet). Efterbrännkamrarna kunde utnyttjas för sen tillförsel av skundärluft och därmed kunde den avancerade luftstegningen tillämpas. Gaskoncentrationerna mättes längs gasflödets väg. För CO, som får representera de flyktiga bränslebeståndsdelarna, registrerades en stark reduktion efter förbränningsrummet. Detta noterades även för NO, men, åtminstone vid slamtillsats, ökade istället oväntat nog N2O koncentrationen i efterbrännkammaren. Ammoniak eller oförbränt koks är inte kända att vara betydande föregångare till, men de är de enda kandidaterna till N₂O bildning som återstår i och efter cyclonen. Skälet till N₂O ökningen behöver utredas ytterligare. Emellertid är den allmänna slutsatsen att en tillfredställande utbränning och emissionsminskning erhölls efter en gasuppehållstid på 6–7 sekunder från bränslets tillförsel. Kommersiella CFB pannor uppnår dessa långa uppehållstider p.g.a. deras stora cykloner. Alltså kan efterbrännkammaren till del kompenseras av cyklonen som kan utnyttjas effektivt som ett system för slutförbränning och för reduktion av utsläpp, speciellt vid samförbränning med högflyktiga bränslen.

Emissionsgränser. Tyska såväl som EU normer överskreds inte då energiandelarna slam var mindre än 25 % både med trä och med kol som basbränsle (Figur 7). Detta är en mycket tillfredställande slutsats eftersom vid praktisk tillämpning av sameldning kommer sannolikt endast mindre andelar av tillsatsbränslen vara av intresse. Även om det inte finns någon bestämmelse som begränsar N₂O emissioner så är det viktigt att uppskatta nyttan av låga N₂O emissioner av miljöskäl. Endast med avancerad stegning kunde N₂O emissionen hållas låg vid sameldning med kol.

Avfall med lågt värmevärde, t.ex. vått avloppsslam, behöver tillsatsbränsle för att brännas. Det är inte uppenbart hur ett sådant tillsatsbränsle skall behandlas vid tillämpning av reglerna för emissionsbegränsning. Både definitionerna av energiinnehållet och justeringen av luftfaktor och luftförvärmning påverkar tolkningen av reglerna och kan vara avgörande för om en anläggning skall klassas som en avfallsförbränningsanläggning eller som ett kraftverk, något som i sin tur har en väsentlig betydelse för emissionsbestämmelserna.

Advanced Air Staging Techniques to Improve Fuel Flexibility, Reliability and Emissions in Fluidized Bed Co-Combustion

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1 BACKGROUND AND OBJECTIVES

1.1 Incentives of co-combustion

Co-combustion of biomass or wastes with coal or other primary fuels has many potential advantages: the effective emission of CO₂ is reduced by replacing some coal with waste, efficient utilization of the energy in waste by converting it to electricity in a coal power station and, of course, the primary purpose-destruction of waste. There are also potential risks: some biofuels may lead to slagging and fouling in the combustor or to bed agglomeration in a fluidized bed, some wastes lead to enhanced emissions of heavy metals and, finally, an augmentation of the gaseous emissions may occur, especially during combustion of sewage sludge. If sewage sludge is to be used as an additional fuel, investigation of the related emissions becomes particularly important because of the large content of nitrogen in the fuel, which in a hypothetical extreme case of dried sewage sludge (if all nitrogen were converted to NO) could give rise to an additional emission of 100 to 200 ppm NO per percent energy of sludge added. Fluidized bed combustion is probably the most advantageous method available for cocombustion because of its fuel flexibility and the possibility to influence the processes of formation and destruction of emissions. Therefore the present work is dedicated to the emission performance of co-combustion of dried sludge in circulating fluidized bed (CFB). In a previous work [1] air supply to the combustor has been shown to have a significant influence on the flue-gas pollutant concentrations (emissions). This is further investigated here with particular emphasis on the impact of sludge and other highvolatile fuels together with coal or wood. To better understand the processes leading to emissions and to get a picture of the formation and reduction processes in the boiler, infurnace measurements of gaseous concentration are presented. In addition, some comments will be made regarding the influence of bed material and ashes on the emissions.

The general incentives for co-combustion can be summarized as follows:

- Co-combustion of biomass is a contribution to the solution of the global CO₂ problem
- Co-combustion of residues may increase profitability of utility stations
- Co-combustion makes use of existing plant capacities
- In general, the required throughputs are small compared to coal throughputs in power stations (e.g. sewage sludge production in Hamburg is 76,000 t dry matter per annum)
- Governmental regulations will limit future disposal of waste (Germany: TASi [2])

The problem areas of co-combustion are, depending on the composition of the fuel:

- Emission of NO and other conventional pollutants
- Emissions (particulate or gaseous) of heavy metals originating from the added fuel.
- Alkali-content of the added fuel may cause sintering, deposits and corrosion
- Emissions of dioxins and furans from certain types of additional fuel.

All these areas are important for co-combustion, but this project focuses on nitrogen compounds (NO, NO₂, N₂O, NH₃, HCN), resulting emissions and in-furnace measures for emission limitation, especially the influence of air supply.

1.2 The idea of advanced air staging



Fig. 1-1: Comparison of conventional (left figure) and advanced air staging (right figure).

Air staging is a well-known measure used for NO_x -control. In conventional ("normal") air staging both primary and secondary air are supplied to the riser of the CFB boiler. The riser (furnace) can therefore be divided into two sections. In the first stage, below the secondary air injection, oxygen-lean conditions favor the destruction of NO by high CO and char concentrations. Downstream of the secondary air injection port, in the second stage, an oxygen-rich atmosphere leads to burnout of the carbon monoxide and other unreacted combustible gases.

During advanced air staging, the second stage is located after separation of the solid particles from the flue gas, while the whole riser is operated under near-stoichiometric conditions (Fig. 1-1). By this method more air is supplied to the lower part of the combustion chamber compared to normal operating conditions. This increase of the amount of air to the bottom part is beneficial for sulfur capture with limestone. On the other hand, the oxygen is depleted at the top of the riser section during advanced air staging, which has been proven to reduce N_2O emissions. Also the NO emission decreases. The effect of advanced air staging on emissions of SO₂, N_2O , NO and CO was first tested in the Chalmers boiler for coal combustion [1].

1.3 Objectives of the present project

The purpose of this project is to study reduction of gas emissions during co-combustion of sludge with coal or wood. The following topics will be investigated:

- Release, formation and decomposition of nitrogen compounds, especially for cocombustion of coal or biomass with waste fuels having high nitrogen content.
- Optimal operation conditions for low emissions (NO_x, CO, SO₂).
- The effect of release of volatiles. This is important, since most of the combustible part in biomass and waste fuels consists of volatiles.
- Release of mercury and other trace metals.

2 EXPERIMENTAL

2.1 Experimental facilities

The plants used in the present investigation are the 12 MW_{th} CFB combustor located at Chalmers Technical University (CTH) and the pilot scale unit at the Technical University Hamburg–Harburg (TUHH). A schematic sketch of both plants is given in Fig. 2-1. The combustion chamber (1) of the CTH unit has a square cross-section of about 2.25 m² 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 and the external heat exchanger 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 a commercial scale. This means that results obtained are transferable to industrial units. Investigations in the boiler are complemented by measurements in the pilot scale unit operated at TUHH. This pilot scale unit consists of a cylindrical combustion chamber (1)



Fig. 2-1: The CFB test facilities at CTU 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.

with a diameter of 0.1 m (cross-section area 0.008 m²) and a total height of 15 m. The fuel is fed into the dense bed of the CFB via a screw feeder (8). The after-burner chamber (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 chamber, resulting in a total gas residence time of about 2.7 s under the operating conditions applied. Although this device is significantly smaller in diameter than the CTH boiler, it has been shown in a previous investigation [3] that the emissions are practically the same as those from the CTH boiler, if suitable similarity rules are obeyed in the operation. The similarity criteria can be summarized by the following conditions that should be approximately the same in both risers:

- bed material, fuel and additive
- gas residence time
- fluidizing and flue gas velocities
- riser pressure drop
- temperature

2.2 Gas analysis systems

The boiler at CTH as well as the test facility at TUHH are equipped with daily calibrated gas analysis systems for monitoring both local in-furnace and flue gas concentrations of O_2 , CO_2 , CO_2 , CO_2 , NO_2 , NO_2 , N_2O , H_2O and C_xH_y . To detect precursors of the nitrogen oxide emissions such as NH₃ and HCN an FTIR instrument was applied.



Fig. 2-2: Gas sampling and analyzing equipment at TUHH. FTIR instrument supplied by CTH.

	DTGS – detector	MCT – detector
volume of gas cell, cm ³	2000	500
optical path length, m	6.4	0.15
scans per minute, -	12	43
concentration range of calibration gases		
H ₂ O	1.40-35.9 vol-%	5.00-30.0 vol-%
CO_2	2.95-25.0 vol-%	2.95–25.0 vol-%
CO	125 ppm – 9.0 vol-%	913 ppm – 9 vol-%
CH_4	5 ppm – 1.5 vol-%	898 ppm – 5 vol-%
C_2H_4	5 ppm – 1.1 vol-%	0.1 - 1.0 vol-%
C_2H_2	19.0–9660 ppm	989–9890 ppm
NH ₃	2–984 ppm	492–984 ppm
N_2O	3.6–254 ppm	25–250 ppm
HCN	5.3–374 ppm	39–388 ppm
HNCO	10–924 ppm	103–1247 ppm

Tab. 2-1: Characteristics of the two detectors of the Bomem MB 9100 FTIR instrument

Fig. 2-2 shows the setup of the measurement system at TUHH. Gas samples were withdrawn at the centerline of the riser and the afterburner chamber, and sucked by a pump through a cyclone, two electrically heated filters and a sampling line. In the case of the hydrocarbon flame ionization analyzer, the water vapor in the flue gas was removed with a permeate drier. In the conventional analysis system the sampled gas was dried in a gas-cooler to avoid condensation in the analyzers. The gas was then supplied to each analyzer at a rate of 1 l/min. The concentration of O₂ was measured on the basis of the paramagnetic effect, whereas CO₂, CO, NO, N₂O and SO₂ were measured by nondispersive infrared absorption spectroscopy. Since ammonia is highly soluble in water and reacts with SO₂ below a temperature of 330 °C, NH₃ was measured with a third sampling line. A SO₂ scrubber was placed prior to the analyzer to remove the SO₂ in the sampling gas, and a second SO₂ analyzer was placed in the sampling line to check the function of the scrubber. The scrubber, as well as the NH₃ analyzer, was heated to 160°C to avoid condensation. UV absorption spectroscopy was used for detecting H₂O.

The Bomem MB 9100 FTIR instrument from CTH was used to perform complementary measurements during both in-furnace and emissions measurements. The FTIR instrument is equipped with two different detectors, the characteristics of which are found in Tab. 2-1. There are two reasons to have a double set-up of gas cells and detectors. The large multi-path cell with the optical path-length of 6.4 meters leads to too high absorbances from H₂O, CO₂ and hydrocarbons when measuring combustion gases from the furnace. A smaller cell with only 15 cm optical path-length reduces the absorbances down to levels needed for evaluation. The second reason is that a MCT detector has a much higher sensitivity in the spectral regions used for evaluation of HCN, a species that is important for this project. As a consequence, the small cell and MCT-detector were used to perform in-furnace measurements, whereas the DTGS detector was applied for emissions. Monitoring of the flue gas emissions by means of a DTGS detector is a reliable and often applied alternative to conventional IR measurements.



Fig. 2-3: Gas analysis system A at CTH used for analysis of flue gas in the stack. (1) gas sampling probe; (2) heated ceramic filter; (3) heated gas sampling line; (4) moisture removal; (5) gas sampling pump; (6) safety filter; (7) mass flow controller, total flow; (8-12) individual volumetric flow controllers; (13) CO₂ analyzer; (14) O₂ analyzer; (15) NO analyzer; (16) SO₂ analyzer; (17) CO analyzer (18) SO₂ washer (sodium carbonate) ;(19) moisture removal; (20) N₂O analyzer.

When axial gas concentration profiles were recorded in the test facility of TUHH, an additional independent sampling system was dedicated to control the emissions of O_2 , CO_2 and CO. The continuous monitoring of the oxygen concentration enables operation of the reactor under steady-state conditions, while the other sampling systems record the evolution of the gas concentrations along the flue gas path inside the furnace.



Fig. 2-4: Gas analysis system (FTIR) used for analysis of flue gases from the combustion chamber (1) gas sampling probe with ceramic filter; (2) heated gas sampling line; (3) heated ceramic filter; (4) heated gas sampling pump; (5) heated gas sampling line; (6) oven with filter and volumetric flow controller; (7) heated gas sampling line; (8) Fourier transformed infrared analyzer; (9) heated gas sampling line; (10) moisture removal; (11) gas sampling pump; (12) gas chromatograph H₂.

CTH operated measuring systems similar to TUHH. In Fig. 2-3 the analysis system for continuous monitoring of the stack emissions at CTH is shown. O_2 was measured by a paramagnetic analyzer, while non-dispersive infrared analyzers were used for CO, SO_2 and N_2O . The NO instrument is a chemiluminiscence analyzer, and the CO_2 instrument utilizes the thermal conductivity principle. N_2O is cross-sensitive towards SO_2 at high concentrations. SO_2 is therefore removed prior to the analysis of N_2O , no. 18-20 in Fig. 2-3.

The system for in-furnace measurements at CTH is shown in Fig. 2-4 and Fig. 2-5. The same gas-sampling probe was used to withdraw combustion gas from the furnace to supply either the FTIR system or the gas analysis system B. The total flow rate through the gas sampling probe was not sufficient for supplying both systems in parallel, and therefore the measurement had to be performed sequentially during day time and evenings. During the nights both the gas analyzing system B and the FTIR were sampling flue gas from the stack as a backup to the gas analysis system A. The heated oven of the FTIR system (no. 6 in Fig. 2-4) makes an easy shift possible between the DTGS and MCT detectors of the FTIR, see Tab. 2-1. In connection to the FTIR system a gas chromatograph (no. 12 in Fig. 2-4) is available to measure the hydrogen concentration (H_2).

The analyzers building up the B-system are identical or similar to those of the Asystem. However, the measurement ranges for CO and NO need to be higher when analyzing combustion gases. Also, a total hydrocarbon analyzer is added to the Bsystem to measure the sum of individual hydrocarbons passing the gas cooler (no. 4 in Fig. 2-5) as methane (CH₄) equivalents.



Fig. 2-5: Gas analysis system B used for analysis of flue gases from the combustion chamber (1) gas sampling probe; (2) heated ceramic filter; (3) heated gas sampling line; (4) moisture removal; (5) gas sampling pump; (6) safety filter; (7) total mass flow controller; (8-11) individual volumetric flow controllers; (12) HC_{tot} analyzer; (13) NO analyzer; (14) SO₂ and O₂ analyzer; (15) SO₂ and CO analyzer; (16) CO-low analyzer; (17) CO₂ and CO-high analyzer



Fig. 2-6: Equipment for measurement of trace element emissions.

The measured concentrations are reported "as measured" when showing results from the furnace. Results from stack measurements are recalculated to be based on a common oxygen concentration of 6 vol-percent. The concentrations are given in mg per m^3 dry flue gas at standard temperature and pressure.

The emissions of trace elements in the flue gases leaving the boiler were determined by a standard procedure for waste incinerators. This involves isokinetic sampling in position located before the flue gas fan using dust collection on a heated filter. Trace elements in the gas phase were trapped in acidic solutions (in the case of Hg in acidic KMnO₄-solution) kept in wash bottles through which the flue gas was passed, Fig. 2-6. Special attention was taken measuring Hg. Apart from being included as one of the trace elements, Hg was also measured in position (14) by an on-line flue-gas measurement system (SEMTECH Hg 2000 from Boliden Contech, Fig. 2-7). This instrument measures elemental Hg (Hg°), but by reducing the ionic form of Hg in for example HgCl₂ (Hg²⁺) by SnCl₂ prior to the entrance of the gas into the SEMTECH analyser, it is possible to also measure the total concentration of Hg (Hg^{tot}). This is called the gaseous concentration of Hg.



Fig. 2-7: Equipment for on-line measurement of elemental and total concentration of Hg in the flue gas duct.

2.3 The fuels investigated

The properties of the fuels are summarized in Tab. 2-2. The base fuels were bituminous coal and wood. Coal I and coal II are both bituminous coals from Poland. Wood I was supplied in the form of chips, whereas wood II consisted of pellets. The co-combustion fuels were Swedish municipal sludge, which after digestion was dried to a residual water content of 13 % (sludge A), and German municipal sludge (sludge B), which after digestion was only mechanically de-watered and had a water content of approximately 77 %. The fiber sludge was supplied by Stora Enso's pulp and paper mill in Skoghall, located close to the city of Karlstad in Sweden. The raw fiber sludge had a water content above 70 wt-percent. Since it is not possible to feed such a wet material directly into the Chalmers boiler, the fiber sludge was pre-dried in a fluidized bed pilot plant dryer [4], where it was dried to a residual water content of about 25 %.

	coal I	coal II	wood I*	wood II**	sewage sludge A	sewage sludge B	paper fiber sludge
proximate							
Water (wt-%, raw)	14.4	8.7	40.8	9.1	15.9	76.6	24.4
Ash (wt-%, dry)	8.5	15.7	0.8	0.8	42.1	43.2	11.4
Volatiles (wt-%, daf)	36.0	35.3	82.0	81.2	91.1	92.4	77.0
Ultimate (wt-%, daf)							
С	84.00	82.50	50.60	50.50	53.20	49.70	59.2
Н	5.80	5.00	6.30	6.00	7.10	8.00	8.0
0	7.00	9.90	43.00	43.40	30.60	33.90	30.7
S	1.50	0.90	<i>n.m</i> .	0.02	1.90	1.50	0.8
N	1.70	1.70	0.14	0.14	7.10	6.90	1.6
Cl	n.m.	0.07	n.m.	0.01	0.05	0.08	n.m.
Trace elements (mg/kg dry fuel)							
Hg	n.m.	0.07	n.m.	0.03	0.71	0.90	n.m.
Pb	n.m.	21	n.m.	5	30	n.m.	n.m.
Cd	n.m.	0.12	n.m.	0.17	0.93	n.m.	n.m.
Cr	n.m.	16	n.m.	5	120	n.m.	n.m.
Cu	n.m.	41	n.m.	5	350	n.m.	n.m.
Ni	n.m.	19	n.m.	2	26	n.m.	n.m.
Mn	n.m.	118	n.m.	138	222	n.m.	n.m.
Co	n.m.	5.8	n.m.	3	7	n.m.	n.m.
As	n.m.	1.6	n.m.	0.3	4	n.m.	n.m.
Sb	n.m.	0.4	n.m.	0.3	1.3	n.m.	n.m.
V	n.m.	39	n.m.	0.2	27	n.m.	n.m.
Tl	n.m.	0.09	n.m.	0.02	0.08	n.m.	n.m.
Lower heating value (MJ/kg)							
H _u , daf	n.m.	31.4	n.m.	18.9	20.9	23.9	22.7
H _u , raw	n.m.	24.2	n.m.	16.8	9.4	1.5	14.6

Tab. 2-2: Properties of investigated fuels

*) delivered as chips **) delivered as pellets *raw* = *as delivered*

n.m. = not measured

s pellets

daf = dry and ash free

The ultimate analysis of the fuels shows a content of oxygen in the sludges of around 30 %, compared to 43 % of the wood and between 7 and 11 % for the coal. The nitrogen content of the sewage sludge is about 7 %, which indicates a high potential for nitrogen oxide formation. The high sulfur content in sludge A originates to a large extent from sulfur-containing proteins in the sludge and to a minor degree from remainders of sulfur-containing compounds used in the sewage water treatment plant for flocculation of the sludge.

The bed material of CFB combustors is known to act as a catalyst in the formation and reduction mechanisms of nitrogen oxides. Therefore Tab. 2-3 presents the ash analyses of the base fuels coal II and wood II and the waste sludge A. The ash of the sewage sludge shows a high iron content that might be caused by the use of iron containing flocculation agents in the waste water treatment process.

The fraction of sewage sludge in the mixtures was calculated on the basis of its contribution to the heating value of the fuel blend. The energy fraction of the waste in the fuel mixture can therefore be determined as follows:

$$\frac{\dot{m}_{w} \cdot H_{u,w}}{\dot{m}_{w} \cdot H_{u,w} + \dot{m}_{bf} \cdot H_{u,bf}} \tag{1}$$

where \dot{m}_W and \dot{m}_{bf} are the feed rates of waste (co-fuel) and base fuel, and H_u denotes the lower heating value of the fuels as delivered. The energy fraction greatly depends on H_u . For large differences in the heating values of waste and base fuel, the energy fraction differs significantly from the mass fraction. The latter one simply relates the fuel feed rates:

$$\frac{m_{W}}{\dot{m}_{W} + \dot{m}_{bf}} \tag{2}$$

The larger the difference in the heating values of base and co-fuel the larger will be the difference between energy and mass fraction.

	coal II	wood II	sewage sludge A
Κ	557 ± 179	448 ± 5	$1820~\pm~164$
Na	137 ± 6	90 ± 16	$602 \ \pm \ 45$
Al	2767 ± 1097	178 ± 52	18400 ± 1341
Si	47667 ± 14434	1000 ± 0	48800 ± 1789
Fe	8167 ± 3175	568 ± 377	73800 ± 2863
Ca	5467 ± 1097	1275 ± 96	27200 ± 1643
Mg	2366 ± 404	193 ± 19	$4180 ~\pm~ 295$
Р	1733 ± 1154	315 ± 182	38800 ± 1644
Ti	300 ± 0	$2875 \ \pm \ 2249$	538 ± 84

Tab. 2-3:Ash composition of two base fuels and dried sewage
sludge. Concentrations in mg/kg dry ash

2.4 Experimental procedure

In the smaller and therefore more flexible test unit at TUHH the air ratio in the combustor was varied over a wide range in order to investigate the influence of air staging on the pollutant emissions. The TUHH unit was operated in measurement campaigns of 12 to 16 hours, whereas the CTH boiler was operated continuously. Operating conditions found to be beneficial for low emissions in the lab-scale unit at TUHH were therefore applied for longer periods at the CTH plant to guarantee steady state operation and to verify the emission levels. For each long test run in the CTH boiler the operating conditions were stabile with respect to air/fuel ratio within 2 hours. The SO₂, NO and N₂O values continue to change until the lime content as well as the ash content from the sludge (iron oxide) attain steady state. This could take 8-12 hours. After this primary stabilization time, the gas measurements with the suction probe were performed during additional 12–16 hours. At the end of this period solid samples were taken in order to track the trace elements in the ash. In some of the tests the gas suction measurements were not finished in time, leading to another over-night operation and further measurements for 8-12 hours the next day. In the case of TUHH, the system was allowed to adapt to new conditions for about one hour after changing the air/fuel ratio in the combustor. After this time the oxygen off gas concentrations were fairly constant and the emissions were recorded for another thirty minutes.

		TL	HH		СТН				
	mas	55	ener	gy	mas	S	energ	<i>y</i>	
	fractio	n, %	fractio	fraction, %		fraction, %		fraction, %	
	base fuel	waste	base fuel	waste	base fuel	waste	base fuel	waste	
pure fuels									
coal	100	0	100	0	100	0	100	0	
wood	100	0	100	0	100	0	100	0	
dried sludge	0	100	0	100	-	-	-	-	
paper sludge	-	-	-	-	0	100	0	100	
mixtures									
coal and	68.8	31.2	85	15	80.7	19.3	87.2	12.8	
dried sludge	53.8	46.2	75	25	-	-	-	-	
-	28.0	72.0	50	50	41.5	58.5	53.7	46.3	
coal and	79.9	20.1	85	15	-	-	-	-	
wood	41.3	58.7	50	50	-	-	-	-	
wood and	75.8	24 2	85	15	85.2	14.8	84.5	15.5	
dried sludge	-		-	-	63.1	36.9	60.8	39.2	
a lea staage	35.6	64.4	50	50	49.7	50.3	47.8	52.2	
coal and wet	41.6	58.4	92	8	_	-	_	_	
sludoe	35.8	64.2	90	10	_	-	_	_	
sinage	31.4	68.6	88	12					

Tab. 2-4:	Fuels and	fuel mixtures	combusted

Mass and energy fractions are calculated according to equations (1) and (2).

Differences between TUHH and CTH are due to fluctuating water content of the fuels.

The influence of the air staging was tested for different fuels and fuel mixtures, as summarized in Tab. 2-4. Mixtures of coal or wood and dried sewage sludge, coal and wet sewage sludge were investigated. Coal, wood and the dried sewage sludge were also mono-combusted. This large variety of fuels and fuel blends enables examination of the impact of the fuel characteristics on the flue gas emissions, mainly the volatile and nitrogen contents, but also the mixing ratio of waste to base fuel.

For the investigation of air staging various air ratios and staging modes need to be defined. The air supply to the bottom air distributor, related to the total fuel feed rate determines the primary excess air ratio $\lambda_{primary}$. In the *advanced staging* mode secondary air was supplied as late as downstream of the primary cyclone, leaving final burn-out reactions to the afterburner chamber. This alternative staging method was compared to the commonly applied technique of *normal staging*, where the secondary air is introduced into the riser shortly downstream of the air distributor. During normal staging operation of the TUHH plant, secondary air injection into the riser takes place at 3.1 m above the distributor plate. The ratio of the height of secondary air injection to the height of the central part of the cyclone inlet is about 0.23, which corresponds fairly well to the value of 0.2 calculated for the CTH boiler where secondary air was injected 2.2 m above the distributor during normal operation. In all cases the mass flow of secondary air was adjusted so that the overall or total excess air ratio λ_{total} was around 1.20-1.23, which is identical to an oxygen concentration in the off gas of 3.6-3.9 vol-percent (dry basis).

	coal + dried sludge	coal + wood	wood + dried sludge	coal + wet sludge
load, MW _{th}	0.029 ± 0.002	0.029 ± 0.000	0.028 ± 0.001	0.026 ± 0.002
riser temperature, ° C	851 ± 11	855 ± 3	858 ± 7	847 ± 3
exit temperature of after burning chamber, ° C	845 ± 11	852 ± 7	854 ± 6	851 ± 3
total pressure drop of riser, mbar	75 ± 4	76 ± 3	75 ± 3	75 ± 3
molar ratio Ca/S	$\textbf{2.3}\pm\textbf{0.1}$	$\textbf{2.3}\pm\textbf{0}$	$\textbf{2.3}\pm\textbf{0}$	$\textbf{2.3}\pm\textbf{0}$
excess air ratio, λ_{total}	1.22 ± 0.01	1.22 ± 0.01	1.21 ± 0.01	$\textbf{1.21} \pm \textbf{0.01}$
advanced air staging				
combustor air ratio, λ_c	1.0 ~ 1.23	1.0 ~ 1.23	1.0 ~ 1.23	1.0 ~ 1.23
superficial flue gas velocity at top of riser U _{too} , m/s	4.80 ~ 5.84	4.63 ~ 5.60	4.53 ~ 5.42	4.93 ~ 5.84
U_{top} at $\lambda_{primary}$ =1.05, m/s	5.01 ± 0.08	$\textbf{4.81} \pm \textbf{0.07}$	$\textbf{4.73}\pm\textbf{0}$	$\textbf{5.14} \pm \textbf{0.01}$
normal air staging bottom bed air ratio,	0.7 ± 0	0.7 ± 0	0.7 ± 0	0.7 ± 0
λ _{primary} superficial flue gas velocity at top of riser U _{top} , m/s	5.72 ± 0.15	5.53 ± 0.1	5.42 ± 0	5.84 ± 0

Tab. 2-5: Operating conditions for advanced and normal staging at TUHH

	coal + dried sludge	wood + dried sludge
load. MWth	6.5± 0.1	6.5± 0.1
bed temp., C (bottom)	841 ± 0	841 ± 0
bed temp., C (top)	855 ± 1	857 ± 3
exit temperature of after burning	772 ± 4 (2)	797 ± 1(782)(1)
chamber, ° C		
total riser pressure drop, mbar	68.3 ± 2.9	67.9 ± 3.9
Calcium addition	$2.3{\pm}0.05$	$1.9 \pm 0.1(0)$ (1)
molar ratio Ca/S		
Ca/S with Ca in	2.6 ± 0.2	$2.5\pm0.1(0)$ (1)
fuel included		
excess air ratio, λ_{ex}	$\textbf{1.23} \pm \textbf{0.01}$	$\textbf{1.23} \pm \textbf{0.01}$
combustor air ratio, λ_{c}	1.05 ± 0.01	1.04 ± 0.01
superficial flue gas velocity at	5.3 ± 0.4	4.6 ±0.1(4.1)(1)
top of riser U_{top} , m/s		

Tab. 2-6: Operating conditions during advanced staging at CTH

(1) without sludge, (2) trend, increasing with amount of sludge

In all cases the load of the combustors, i.e. the heat input, was kept constant. The resulting operation data during the tests are presented in Tab. 2-5 and Tab. 2-6. The feed rate of fuel was adjusted until the required oxygen concentration was obtained for a fixed amount of combustion air giving a flue gas velocity of around 5.5 m/s at the top of the riser during operation with no-staging. During variations of the air ratio of the combustor in case of advanced staging, the total air feed rate was kept constant and the split between primary and secondary air supply was changed. As a consequence, the flue gas velocity at the top of the riser varied between 4.5 and 5.8 m/s. In order to verify that measurements performed at the TUHH test plant are well comparable to those at the large-scale CTH boiler, for distinct operating conditions the TUHH plant was run for 8–12 hours which is in the same range as the primary stabilization time applied at CTH. At these occasions axial gas concentration profiles along the riser height were recorded to identify the location of nitrogen oxides formation. These axial profile measurements serve as a basis for interpretation of the NO_x/N₂O formation mechanisms.

OPERATION EXPERIENCES

3



3.1 Influence of flue gas velocity on emission values

Fig. 3-1: Comparison of results under co-combustion conditions. Either the fuel feed rate (●) or the flue gas velocities (■) were kept constant. TUHH plant.

The influence of variations in the gas velocities due to air staging was tested by changing the air staging while the fuel feed rate remained constant, or by changing the fuel feed rate while keeping fluidization air flow and flue gas velocity on top of the riser constant. Fig. 3-1 shows the emissions measured in the afterburner chamber in the two cases. Obviously, the influence of load is not significant. Within the ranges employed it does not matter whether air flow or fuel feed rate is adjusted. Similar experiences have been obtained in the CTH boiler.

3.2 Influence of fuel type on the stability of operation

During combustion of high volatile fuels the oxygen concentration in the off gas of the TUHH unit varied more than for fuels with a high fixed carbon content (Fig. 3-2). However, the emissions remained fairly constant. Corresponding data from CTH are given in Tab. 2-6.



Fig. 3-2: Oxygen off gas concentration in the case of wood () and coal combustion (-.-.), advanced staging. TUHH plant.



Fig. 3-3: Typical temperature profiles during co-combustion of wood and coal. Left figure: energy fraction of coal 85 %, right figure: energy fraction of coal 50 %. TUHH plant.

A perfectly homogeneous temperature profile could not be maintained in the TUHH test facility, see Fig. 3-3. A peak near the fuel supply and slightly lower temperatures at the top, where additional heat losses due to the steel beam on which the plant is mounted, were recorded. This steel construction, although insulated as much as possible, works like a cooling fin. Corresponding data from CTH are given in Tab. 2-6.

3.4 Hot flue gas sampling

Differences between the fluid dynamics in the afterburner and in the riser section at TUHH required different gas sampling methods. Two problems became obvious: high dust concentrations and high temperatures. This might lead to production of CO from char fines collected on the hot ceramic filter of the gas sampling probe and a large error in the CO results.



Fig. 3-4: Deposits on a sampling probe within the afterburner (left) and a deposits-free filter probe in the riser (right).

In the afterburner of the TUHH plant gas velocities decrease to below 1 m/s. At such low velocities the fine fly ash particles settle down on the filter of the probe and build up deposits that cause high CO concentration in the flue gas analysis, Fig. 3-4 shows some filters. As a solution, gas was sucked from the interior of the afterburner by a small open tube with an inner diameter of 6 mm. The flue gas, as well as the fly ash, was rapidly cooled in the subsequent flue gas-sampling cyclone and fly ash, including unburned char particles, was removed from the sampling line. In the riser section of the
TUHH test rig it was sufficient to operate with ceramic filter probes, which separated the particulate phase from the gas phase already within the combustion chamber. The high gas velocities around 4–5 m/s and the high concentrations of solids of a wide size distribution kept the ceramic filters free from any deposition (Fig. 3-4).



Fig. 3-5: Gas suction probe as used at CTH

CTH solved the problem by a small filter tip and cooled probes. In order to avoid condensation of water during suction of combustion gases, the center tube of the probe is electrically heated by a low-voltage high-current transformer, Fig. 3-5. The ceramic filter is kept clean by the coarse bed material as long as the measurement is performed upstream of the hot cyclone. Measurements after the hot cyclone leads to continuous build up of a filter cake, containing fine char particles, which gradually increases the CO concentration. By keeping the measurement time below 10 minutes, back-flushing the filter with pressurized air and repeating the measurement, reliable CO concentrations could be recorded. In addition, gas velocities in riser and afterburner did not vary as much as in the small plant at TUHH

3.5 Limitations of the test facility of TUHH

Much stronger transient variations were observed for SO₂ than for other emissions. As soon as the O₂ concentration is stable, char hold-up is stable, and therefore NO_x/N₂O do not change anymore. However, SO₂ is still far away from steady state, since it relies more on a steady state composition of the bed material. A comparison of solids inventory of the plant and the ash input shows that the bed material attains steady state composition only after several hours. The exchange of the total inventory of the TUHH riser, about 6 kg corresponding to a pressure drop of 7500 Pa, needs more than nine hours with an ash flux of approximately 0.65 kg/h. Taking into account that there is a considerable amount of bed material in the siphon and that the fluidized bed behaves like a stirred tank for the solid phase, this time is most likely several times longer. Consequently, the composition of the bed material does not attain steady state, and the SO₂ results measured at TUHH are not reliable.

3.6 Reliability of FTIR measurements in flue gas analyzing

Comparison of conventional gas analysis and FTIR leads to the conclusion that reliable routines for online-evaluation of the FTIR spectra exist for measurement of flue gas concentrations, Fig. 3-6. However, during in-furnace measurements of high pollutant concentrations no commercial routines are available. Here, the spectra must be evaluated manually off-line. Experience from previous projects exists regarding the use of various evaluation methods [5,6]. The present set-up also permits a comparison between the two detector systems. Species not found with the MCT detector, for example, should hardly lead to any measurable concentration with the DTGS detector either. In this way the risk of producing ghost species in the evaluation methods is minimized. Still, evaluation methods for the in-furnace measurements are time-consuming and could not be fully utilized for the spectra collected.



Fig. 3-6: Comparison of flue gas emissions measured by means of the FT-IR analyzer and the conventional non-dispersive gas analyzers. FTIR spectra were evaluated on-line by Bomem software package MB 9100.

3.7 Fouling and corrosion

The higher content of alkali metals, namely potassium and sodium, in the ash of municipal sewage sludge may cause enhanced fouling of the heat exchanging surfaces may occur. The alkali oxides K_2O and Na_2O are known to have comparatively low melting points. In addition, alkali oxides or salts can react with silicon compounds of the bed material to form eutectic mixtures such as $Na_2O \cdot 2SiO_2$ or $K_2O \cdot 4SiO_2$ the melting temperatures of which are lower than for the individual compounds. $K_2O \cdot 4SiO_2$, for example, has a melting point of 764 °C [7]. The ash of sewage sludge, however, also contains compounds with high melting points such as Al_2O_3 and SiO_2 , and the melting point of the total ash

 Tab. 3-1:
 Alkali indices and energy related chlorine contents of selected fuels

	alkali index, g/MJ	energy related chlorine content, g/MJ
coal II	0.005	2.23
wood II	0.003	0.50
dried sludge A	0.113	2.59

is around 1100 to 1200°C [8], which is only slightly below the typical melting point for the ash of bituminous coal with approximately 1300 °C [7]. In order to quantify the potential risks originating from alkali oxide concentrations, an alkali index has been introduced which relates the mass flow of alkali metal oxides (K₂O and Na₂O) produced with the ash to the energy input of the fuel [9]. Threshold values of 0.17 g alkali/MJ, below which fouling is unlikely, and 0.34 g alkali/MJ, above which fouling is virtually likely to occur, can be found in the literature [9]. As shown in Tab. 3-1 even the alkali index of the investigated dried sludge A is below the first threshold value. Severe fouling is therefore being unlikely. However, it should be noticed that ash melting points measured in the laboratory or indices calculated from the ash composition are not sufficient to predict the ash behavior with certainty in a technical plant, since additional factors may play a role.

Elevated hydrogen chloride concentration in the flue gas may contribute to high temperature corrosion during co-combustion. HCl could react in an oxygen lean atmosphere with the iron of the heat exchanger surfaces in a complex way to form $FeCl_2$ [8]. The hydrogen chloride concentration is related to the chlorine input with the waste fuel and hence to the energy content of the fuel, since the heating value of the fuel determines the input flow. As can be seen from Tab. 3-1 the energy related chlorine content of the dried sewage sludge A is not significantly higher than the value for coal. Thus, in the case investigated enhanced high temperature corrosion due to high HCl concentrations is probably negligible. Since the chlorine content of municipal sewage sludges can fluctuate strongly, one has to be careful if other sludges than the investigated one are to be co-combusted.

In accordance with this theoretical consideration based on the fuel and ash composition only, there were no signs of deposition and corrosion during tests carried out. However, the tests were too short to draw fundamental conclusions, and the investigations were not primarily focused on deposits or corrosion.

4 **RESULTS AND DISCUSSION**

4.1 Emission results

In the following, first the influence of air supply will be accounted for, then, at constant air supply, the influence of waste fraction is shown. Finally, a number of different results are presented: influence of wet sludge on emissions, local concentrations of the pollutant species, influence of bed material, mercury emission etc.

4.1.1 Influence of air staging

As an initial step for investigation of co-incineration, the combustion characteristics of the pure fuels were examined first in the TUHH unit, and then a comparison is made with results from the CTH boiler. Fig. 4-1 compares emissions of CO, NO and N₂O during combustion of pure coal with results of pure sludge incineration as a function of the combustor air ratio. The exhaust gas oxygen concentration is given as an indicator of steady state conditions and shows 3.6 to 3.9 % oxygen, corresponding to $\lambda_{total} = 1.20$ to 1.23.

In the case of coal the most significant effect of adding secondary air after the cyclone is seen in the NO plot: lowering $\lambda_{primary}$ decreases NO significantly below the value obtained during no-staging ($\lambda_{primary} = 1.2$). At $\lambda_{primary}$ of 1.05 the NO concentration is about 80 mg NO₂/m³, 75 % lower than the no-staging case. Also compared to normal staging the alternative staging method reduces NO from 125 to 80 mg NO₂/m³, i.e. by 35 %. This reduction of NO is accompanied by rising CO emission, which can be explained by higher char hold-up in the combustor at lower primary air. More char in the riser leads to more char in the cyclone, providing a potential source for freshly produced CO. Carbon monoxide also increases at normal staging. For coal, the alternative staging technique very efficiently lowers the N₂O emission. While the emission under normal staging conditions is 200 mg N₂O/m³, it falls to 60 mg/m³ at a primary excess-air ratio of 1.05 and secondary air injection after the cyclone. A tendency of increasing SO₂ emissions with decreasing $\lambda_{primary}$ was noted.

The dried pure sludge behaved in a different way. The NO concentrations are one order of magnitude higher than in the coal case due to the high nitrogen content of the fuel (note the logarithmic scale in Fig. 4-1. If all fuel-N of the sludge were converted to nitric oxide, a concentration of 8130 ppm NO (16660 mg NO₂/m³) would be obtained, whereas a complete fuel-N conversion to NO in the case of coal would result in an emission of 1180 ppm NO (2420 mg NO₂/m³) only. Although staging lowers the NO emissions during pure sludge combustion, it is less effective than for coal. When burning sewage sludge virtually no CO emissions were detected, which indicates a low char hold-up in the combustor and a complete volatile burn-out. Even alternative staging could not reduce the N₂O emissions for mono-combustion of dried sludge. On the contrary, if the air supply to the riser is reduced too much, advanced staging tends to increase the nitrous oxide emissions. However, in all cases the N₂O emissions from dried sludge was comparatively low.



Fig. 4-1 Influence of combustor air ratio on CO and NO emissions. TUHH test facility. Pure fuels. (♦ coal II, O dried sewage sludge A). N means normal staging.



Fig. 4-2: Influence of combustor air ratio on CO and NO emissions. TUHH test facility. Coal II/dried sludge A mixtures. (◆ coal II, □ 15 % dried sludge A, ▲ 25 % dried sludge A, ▼ 50 % dried sludge A). N means normal staging.

Since the emission characteristics of coal and sludge are very different, the question arises how the mixtures of the fuels would behave. In Fig. 4-2 emissions for three coal/dried sludge mixtures, 15 %, 25 % and 50 % of the total heat release, are shown along with pure coal. The CO emissions for all mixtures increase with decreasing $\lambda_{primary}$, similar to the behavior of coal. However, the slope of increase becomes smaller for a higher ratio of dried sludge. Staging lowers more effectively the NO emissions in the case of co-combustion than during mono-combustion of dried sludge, but for higher sludge contents the reduction effect becomes smaller and the emission tends to increase. The N₂O emission is similar to that from coal for all mixtures, i.e. the sensitivity of the N₂O emissions versus air staging is ruled by the coal combustion.

One can conclude from Fig. 4-2 that the emissions cannot be predicted as an average of the pure fuel emissions, but for small mixture fractions of sludge the emissions follow the trends of pure coal combustion. The very different emission behaviors of the dried sludge and the coal are most probably related to their different volatile and nitrogen contents. In order to examine separately the influence of volatiles and fuel nitrogen, blends of wood and dried sludge (high volatile, high nitrogen content) and of coal and wood (high volatile, low nitrogen content) were co-combusted and the results are shown in the following.

Fig. 4-3 shows the results of co-combustion with wood as base fuel and dried sewage sludge as additional fuel. Air staging has hardly any effect on the emissions from



Fig. 4-3: Influence of combustor air ratio on CO and nitrogen oxide emissions. TUHH test facility. Wood II/dried sewage sludge A mixtures. (**O** wood II, ■ 15 % dried sludge A, **V** 50 % dried sludge A). N means normal staging.

combustion of pure wood, regardless of whether the secondary air is injected into the riser in the form of normal staging or after the cyclone in the form of advanced staging. The NO emission is fairly constant at about 125 mg NO_2/m^3 . This value is high compared to the small input of nitrogen with the wood. The maximum concentration of NO, if all fuel-N in wood would be converted to NO, is only 180 ppm NO (370 mg NO_2/m^3), that is, one third of the fuel-N is converted to nitric oxide. As a contrast, the N_2O emission remains at a very low level of about 10 mg NO_2/m^3 for the whole range of air ratios.

Addition of sludge to wood had virtually no impact on the CO emission, which is well below 20 mg/m³. This is understandable, since both fuels have high-volatile content. The volatiles can be assumed to burn completely, and the additional production of CO from char combustion is insignificant. A direct consequence of the increased nitrogen feed with the sludge is the rise in the level of both nitric and nitrous oxide emissions. As in the case of pure wood, air staging does not affect the NO or the N₂O emissions strongly. Only a minor decrease with reduction of primary air ratio down to a value of $\lambda_{primary} = 1.08$ is observed, but for further reduction of $\lambda_{primary}$ both emissions increase again. Obviously, the NO_x reduction potential of high volatile fuels is much lower than that of fuels with high fixed carbon content and consequently higher char concentration in the furnace.

During co-combustion of wood and coal, the increased fraction of wood lowers the nitrogen input to the combustor. Consequently, nitric and nitrous oxide emissions are



Fig. 4-4:Influence of combustor air ratio on CO and nitrogen oxide emissions. TUHH test facility. Coal II/wood II mixtures. (♦ coal II, □ 15 % wood II, ∇ 50 % wood II,
wood II). N means normal staging.

expected to decrease with increasing wood fraction. This was confirmed for no-staging, $\lambda_{primary} = 1.2$ (Fig. 4-4). However, when air staging is applied to fuel mixtures containing coal, the nitrogen reduction ability of the char produced compensates for the additional N-fuel input by coal, and the NO emission is reduced to the same level as in the case of mono-combustion of wood. The CO emission follows very much the behavior observed during coal combustion.



Fig. 4-5: Influence of combustor air ratio on CO, SO₂ and nitrogen oxide emissions. CTH boiler. Pure fuels: coal I (◆),wood I (chips, ⊗), wood II (pellets, ●) and paper sludge (*). Mixture: Wood II/dried sewage sludge A, 60.8/39.2 % (▲). N means normal staging.

Fig. 4-5 shows the influence on emissions of combustor air ratio in the CTH boiler. A number of tests with pure fuels (coal, wood, paper fiber sludge) and one wood/ sludge mixture can be compared with each other and with TUHH data from Fig. 4-1 and Fig. 4-3. In addition to the TUHH data in previous figures, sulfur emission was also recorded. Note that the scales start at an air ratio of 0.9 in the CTH case. The general conclusion from the comparison of the influence of the air supply for the emission performance of the two plants is that they perform in a quite similar way. The greatest difference is that the N₂O reduction versus air ratio is more powerful in the CTH plant. A few comments can be made regarding the minor peculiarities in the curves: The rise in the CO emission for wood chips at low air ratio depends on insufficient reaction in the afterburner because of too low a temperature. This particular test was done before the cold tube walls were insulated to raise the gas temperature. The rise in SO₂ for wood pellets does not come from the pellets, but from a minor fraction of CaSO₄ remaining in

the bed material from the previous test with coal. SO_2 is released from $CaSO_4$ when exposed to severe reducing conditions, such as at low air ratio. The fiber sludge is a biogenous fuel. It is therefore reasonable that the NO versus $\lambda_{primary}$ relationship is the same as for wood. However, the CO emission does not fit into this picture. The reason why the CO emission from the fiber sludge is 200 mg/m³ for single stage combustion is that the fiber sludge combusted contained a large fraction of fines that were elutriated to the afterburner chamber when supplying all combustion air to the bottom of the combustion chamber at single stage combustion. Since the combustor could only be operated during one day with fiber sludge, more experiments are needed to clarify the situation. The SO₂ emission in the fiber sludge case is high because the sulfur content of the fiber sludge is equivalent to that of the coal, but the coal was burned in the presence of limestone that binds the SO₂, whereas there was no addition of limestone together with fiber sludge.

While CO emissions can be regarded as an indicator of the completeness of volatile burn-out, the loss of ignition (LOI) of the fly ash gives an indication of the char hold-up of the riser: the more char that is withheld in the furnace, the higher the LOI of the fly ash. Fig. 4-6 shows the LOI obtained from the ash collected in the cyclone of the flue gas sampling line, versus the staging technique applied, i.e. single staging, normal staging and advanced staging. Since the fly ash was not regularly sampled from the hot bag filter of the test facility at TUHH, the ash from the gas-sampling cyclone was analyzed in all cases. The figures presented may differ from the bag filter ash, but they are expected to give the correct tendencies. In general, the lowest LOI values are obtained for single stage combustion, regardless of the fuel or fuel mixture burned. The LOI increased for normal staging and was highest for advanced staging. If the LOI of the fly ash is an indicator of the char content in the riser, these data support the assumption of improved NO reduction at elevated char concentrations. A drawback of high LOI values is a corresponding loss of combustion efficiency. However, this loss of efficiency also depends on the absolute flux of fly ash, and this was not measured at the TUHH test facility.



Fig. 4-6: Influence of applied staging technique on loss of ignition of fly ash; TUHH test facility. Left: coal/dried sludge mixtures; right: wood/dried sludge mixtures (◆ base fuel, ■ 15 % dried sewage sludge, ∇ 50 % dried sewage sludge, ● 100 % dried sewage sludge).

4.1.2 Influence of fuel composition

The data in this section express the influence of the energy fraction of the additional fuel recorded for an operation condition with $\lambda_{primary} = 1.05$ and total air ratio 1.2.

In Fig. 4-7 emission of nitrogen oxide and conversion of fuel-N for co-combustion of coal or wood with dried sewage sludge are compared. Due to its high nitrogen content, the addition of dried sewage sludge results in a significant increase of the NO emissions in both cases. However, the nitrogen conversion in terms of kilogram fuel nitrogen converted to kilogram nitrogen in NO, shows a reverse pattern. With coal as a base fuel, up to an energy input of 50 % sludge there is still enough char and carbon monoxide present in the riser for a strong reduction of nitric oxide. This is reflected in the comparatively



Fig. 4-7: Nitrogen oxide emissions and conversion ratios, CO emissions and LOI values of fly ash of co-combustion of coal II/dried sludge A and wood II/dried sludge A under advanced staging conditions; λ_{primary}=1.05, λ_{total}=1.2, secondary air injection after cyclone (TUHH: ◆ base fuel coal, ● base fuel wood; CTH: ◆ base fuel coal, ● base fuel wood; CTH: ◆ base fuel coal, ● base fuel wood.

high CO emission and the high LOI values (Fig. 4-7). The fuel-N input increases with the sludge supply, while the reduction conditions are simultaneously good. Only for pure sludge combustion the NO conversion increases again, since less char is available, but the reduction is still considerable, 96 %, which possibly indicates an effect of the volatile nitrogen compounds for NO reduction.

The argument for the favorable NO reduction conditions caused by char does not hold any more when wood is the base fuel. Both wood and sludge have a high volatile content, and only small amounts of char are present in the combustor as indicated by the low LOI values of the fly ash. Indeed, mono-combustion of wood yields, relatively seen, high fuel-N conversion to NO, above 10 %, but sludge addition lowers the NO conversion again. Wood as a high-volatile and low-nitrogen fuel and dried sewage sludge as a high-volatile and a high-nitrogen fuel have different combustion properties with respect to nitric oxide emissions. Possibly, further influencing factors arise due to bed ash composition and from different volatile and/or nitrogen release mechanisms. In all cases of co-combustion of dried sewage sludge, the effective conversion of fuel-N to NO was below 4 %, i.e. 96 % up to 99 % of the fuel nitrogen was not converted to NO. Due to the high nitrogen content of the dried sludge, a small change in conversion leads to a great change in the emissions, and consequently prediction of the NO emission becomes uncertain.

The favorably low emission of N_2O depends on the alternative staging and on the insignificant emissions of nitrous oxide during mono-combustion of wood, regardless of the staging method applied. During co-combustion with sludge the emission rises slightly, but the values are still low, both with coal and with wood as base fuels. The conversion of fuel nitrogen to N_2O is also only a few percent. Again, it is verified in Fig. 4-7 that the same emission trends were observed in the two combustors. Only minor differences occurred.

Sulfur is added by sludge and coal but not by wood. The addition is such that SO_2 has to be removed to comply with the emission regulations. In fluidized bed combustion this can be achieved by addition of limestone to the bed to absorb the SO_2 . Almost complete absorbtion can be attained, but then large quantities of limestone are needed. Generally, it is satisfactory to reach about 90 % absorbtion by an addition of 2 to 3 times the



Fig. 4-8: Lime efficiency versus sludge fraction for coal (♦) and wood (**O**) as base fuels in the CTH boiler.

stoichiometric amount of limestone, and this is so also in the present case. The Ca/S molar ratio was 2.3 and another 0.3 can be added to this number to account for calcium in the sludge ash. However, the sulfur content of the sludge varied and the exact molar ratio of Ca/S could only be determined after the tests. To eliminate the resulting variations in Ca/S, the efficiency of sulfur capture is instead expressed in the form of lime efficiency, that is, the fraction of the lime that was utilized for sulfur capture to attain an acceptable emission (below or close to the emission limit). This is plotted in Fig. 4-8, which shows that the lime efficiency was between 40 and 30 %, high for coal as a base fuel and lower for wood. The efficiency did not vary very much with sludge content at an approximately constant Ca/S. The differences between coal and wood as base fuels can be compensated for in practice by adding more or less limestone, but a more important conclusion is that in the case of coal, desulfurization with limestone is carried out also in the absence of sludge addition, and the sulfur content of the sludge requires only a marginal rise in the feed rate of limestone. If wood is base fuel, on the other hand, sulfur removal is needed only because of the sludge.



^{Fig. 4-9: Effect of staging under co-combustion conditions with coal II(left) and wood II(right) as base fuels (TUHH combustor, NO concentrations based on 6 vol-% O₂, λ_{total}=1.2, air staging with λ_{primary}=1.05 and secondary air addition after cyclone, ▲ single staging, ■ advanced staging).}

Fig. 4-9 illustrates the impact of staging on coal/sludge and wood/sludge mixtures. Two cases are plotted in each graph: no-staging and advanced staging, the latter with a primary excess air ratio of 1.05 in the combustion chamber. In both cases the total excess air ratio was kept at 1.2.

Staging has a great effect on combustion of coal, but with increasing addition of sludge, the difference between no-staging and staging becomes small. In the case of coal, reduction of the primary air addition leads to a char- and CO-rich environment in the primary combustion zone. NO is reduced there, either directly by char, or by reaction with CO

catalyzed on the char surface. The smaller the char concentration becomes during sludge addition, the lesser the effect of this reduction, and hence of staging.

In accordance with this hypothesis, there is no effect of staging on the NO emissions during co-combustion of wood with sludge, since both fuels are high-volatile and therefore generate only small quantities of char. During both normal and advanced staging increased sludge supply results in an exponential increase of the NO emission.



Fig. 4-10: Nitrogen oxide emissions and conversion ratios, CO emissions and LOI values of fly ash of co-combustion of coal II/wood II under advanced staging conditions; $\lambda_{primary}=1.05$, $\lambda_{total}=1.2$, secondary air injection after cyclone, TUHH combustor.

To verify the role of char in the NO reduction process, similar combustion tests under advanced staging conditions were performed with mixtures of coal and wood. The results of these tests are summarized in Fig. 4-10 and Fig. 4-11. As reported in earlier studies, carbon monoxide and nitrous oxide [10] as well as the LOI of the fly ash are expected to



Fig. 4-11: Effect of staging under co-combustion conditions of coal II with wood II (TUHH combustor, NO concentrations based on 6 vol-% O₂, λ_{excess}=1.2, air staging with λ_{primary}=1.05 and secondary air addition after cyclone, ▲ single staging, ■ advanced staging)

decrease linearly with increasing wood content of the fuel mixture, because these emission parameters are close to zero for mono-combustion of wood, but much higher for combustion of coal.

The NO emission depends on the fraction of wood in a complex way. The emissions from combustion of pure wood and coal are in the same range despite the large differences in fuel nitrogen content. Consequently, the conversion of fuel nitrogen to NO is much higher for wood than for coal. Starting from pure wood combustion and reducing the amount of wood in the feed mixture increases the fuel nitrogen input to the reactor, and thus the NO emission rises. However, with further coal supply the char content in the riser increases too, and the NO reduction reactions supersede the formation of NO caused by the higher N-fuel supply. Therefore, after passing through a maximum, the NO emission declines with further increase of coal fraction to concentrations around 100 mg NO₂/m³. The conversion ratio increases monotonously for increasing amount of wood. The emissions of N₂O and CO follow the same pattern as has been described before: they go from high to low values when the wood fraction increases. These relationships have been further elucidated in tests in the CTH boiler [10].

Fig. 4-11 presents staging and no-staging for wood/coal mixtures similar to the results in Fig. 4-9 for coal/sludge and wood/sludge. In Fig. 4-9 the emissions in the no-staging case were significantly influenced by the nitrogen content of the sludge, and the emissions did not fall as in the case of wood, Fig. 4-11. The difference between the two cases in Fig. 4-9 and Fig. 4-11 is found in the volatile nitrogen content, since most of the other conditions were similar.

4.1.3 Emission behavior of mechanically de-watered sewage sludge

In order to investigate whether the emission behavior of blends of dried sludge under cocombustion conditions is comparable with other kinds of waste, co-incineration tests with mechanically de-watered sewage sludge were performed. Since not all waste-water treatment plants have the opportunity to dry the sewage sludge, wet sludge is of great practical importance. Co-combustion of only mechanically de-watered sludge saves the effort for additional sludge drying on the expense of a lower mixing ratio, since otherwise the combustion temperature tends to be too low. Wet and dry sewage sludges are known to have different emission behaviors when mono-combusted [11], and the question arose if these different emission manners are of relevance in the case of co-combustion.

Co-combustion tests with coal and wet sludge, having a water content of approximately 76 % (sludge B, Tab. 2-2), were conducted in the TUHH combustor only, since it was not possible to feed such a paste fuel into the Chalmers boiler at the time of the experimental investigations. In the TUHH combustor the sludge was fed via an eccentric screw pump through a tube with an inner diameter of 16 mm into the bottom zone of the combustor at 0.4 m above the distributor. As in the previous case the energy fraction of the sludge was computed on the basis of the heating values of the raw sludge and coal. No corrections according to legal requirements as highlighted in Section 4.2 were made in this present section. The amount of wet sludge was limited to an energy fraction of 15 % corresponding to a mass fraction of about 74 % in the TUHH plant. Above this limit it was not possible to maintain a combustion temperature of 850°C.



Fig. 4-12: Influence of combustor air ratio on CO and nitrogen oxide emissions. TUHH test facility. Coal II/wet sludge B mixtures (◆ coal II, ■ 8 %wet sludge B, ▼ 10 % wet sludge B, ▲ 12 % wet sludge B). N means normal staging.

In Fig. 4-12 the influence of the combustor air ratio $\lambda_{primary}$ on the CO and nitrogen oxide emissions is shown for normal and advanced staging. The CO emissions rose from the minimum of around 60 mg/m³ in the no-staging case to 130 mg/m³ with $\lambda_{primary}$ at 1.0 with no clear influence of the sludge content. Since high-volatile sewage sludge is supposed not to contribute much to the CO emissions under the chosen combustion conditions, and the energy fraction of sludge is comparatively small, the CO emission pattern mostly depends on the coal. The NO emission is strongly related to the wet sludge fraction under no-staging conditions. Although the nitrogen input to the combustor increases with the fraction of wet sewage sludge, the NO emission decreases in the case of non-staged combustion. This effect was not observed for co-combustion of dried sewage sludge. Apparently, some components in the wet sewage sludge affect the formation or destruction of nitric oxides. On the other hand, during air staging, the NO reduction potential of char, mostly originating from the base fuel coal, evens out the differences between the different fuel blends and leads to a reduced nitric oxide emission of ca. 100 mg/m³ at $\lambda_{primary} = 1.05$. As in the tests with dried sludge, the only staging method, which effectively reduces the nitrous oxide emissions, is the advanced staging. However, if wet sludge is co-combusted, care must be taken not to reduce the primary air supply too much, since for $\lambda_{primary}$ below 1.08 a significant increase of N₂O was observed.



Fig. 4-13: Nitrogen oxide emissions and conversion ratios for co-combustion of coal II/dried sludge A and coal II/wet sludge B under advanced staging conditions, TUHH test facility, λ_{primary}=1.05, λ_{total}=1.2, secondary air injection after cyclone (◆ co-fuel dried sludge, ∇ co-fuel wet sludge).

In general, the emissions of the fuel blends investigated are dominated by the characteristics of coal. This is clearly seen in the co-combustion tests with dried sludge, where especially low amounts of co-fuel did not significantly change the emission characteristics of the base-fuel. However, distinct differences in the nitrogen oxide emission behaviors of wet and dried sewage sludge were also found in some cases.

In Fig. 4-13 the dependence of NO and N₂O on the fraction of mechanically de-watered sludge is compared to that of dried sludge for advanced staging. The NO emissions during staged combustion are quite similar for both co-fuels, i.e. there was only a slight increase of NO emission, despite a strong increase of fuel nitrogen input with a larger sludge fraction, and this resulted in smaller NO conversion. In contrast, the N₂O emission and the corresponding conversion rise with wet sludge as co-fuel, whereas dried sludge as co-fuel resulted in decreased N₂O conversion, if the waste fraction in the fuel blend is high. This opposite trend can be explained by the combustion behavior of pure wet and pure dried sludge [11].



Fig. 4-14: Effect of staging under co-combustion conditions of coal II/dried sludge A (left) and coal II/wet sludge B (right) mixtures (TUHH combustor, NO concentrations based on 6 vol-% O_2 , λ_{total} =1.2, air staging with $\lambda_{primary}$ =1.05 and secondary air addition after cyclone, \blacktriangle single staging, advanced staging)

Fig. 4-13 suggests that the NO emissions of wet and dried sewage sludge are similar under co-combustion conditions. However, the effect of staging is quite different for the two fuels as can be seen in Fig. 4-14. Two cases are plotted in each graph: no-staging and staging. In the latter case the primary excess-air ratio was 1.05 in the combustion chamber and the secondary air was injected after the cyclone. In both cases the total excess air ratio was kept at 1.2. In the no-staging case the pure coal leads to a fairly high emission of about 330 mg NO₂/m³. Addition of sludge causes a significant difference between the dried and the wet sludge: dried sludge results in a roughly unchanged NO emission, whereas wet sludge produces a drastic decrease of the NO in the no-staging case. The same effect has been noted before [11]. The behavior may be tentatively explained by the de-noxing action of species in the wet sludge, mainly ammonia. Staging is seen to have a great effect on combustion of coal, but with

increasing addition of the biogenous fuel the difference between no-staging and staging becomes smaller, since the char concentration in the boiler is reduced, resulting in less destruction of NO.

As a concluding remark one has to admit that it is uncertain to transfer emission behaviors of one fuel to another. Even in the case of fuels that have similar composition and origin and differ only in water content, predictions are uncertain. This problem is diminished in the case of co-combustion: If the fraction of co-fuel is small the emission behavior of the mixture follows the pattern of the base fuel.

4.1.4 Local gaseous concentrations

Gas concentrations along the gas path through the riser, cyclone and afterburner show the progress of combustion and the transformation of the pollutants. Such data, measured on the centerline of the combustors are presented below for the case of $\lambda_{primary} = 1.05$ and $\lambda_{total} = 1.2$.

The oxygen concentration, Fig. 4-15, falls almost instantaneously to zero in the lower part of the riser. As the quantity of primary air was stoichiometric, this behavior reveals that almost all fuel burns in the bottom bed. The rise of oxygen concentration downstream of the cyclone shows the effect of secondary air injection. In the CTH boiler the oxygen concentration was uneven between 5 and 10 m, most likely because of mixing effects, although the air was evenly supplied to the bottom air distributor with the exception of some air introduced from the front with the fuel.

It is remarkable how the different fuel mixtures have behaved in a similar way in all cases: the progress of combustion has been similar, except for a minor difference in the case of pure wood where combustion (as has often been observed) was less intensive in the bottom bed and the oxygen concentration was higher.

The concentrations of CO and CH_x (Fig. 4-16) give an indication of the progress of combustion as well as of the release of volatiles. As expected, the data for wood are higher than those of coal, and also the impact of sludge is clearly seen, especially for CH_x . The concentrations in the combustion chamber are quite high, the scales on the axes are in percent for both CO and CH_x , but the emissions are low as seen from the data in previous diagrams, even practically zero for CH_x . In the small test plant at TUHH the peak concentrations for unburned hydrocarbons were lower than in the large-scale facility and the decrease over height is less pronounced. However, the final emissions are virtually the same.



Fig. 4-15: Gas concentrations of oxygen on the centerline of the combustion chamber of the CTH and TUHH during co-combustion of dried sewage sludge A with wood II and coal II as base fuel.



Fig. 4-16: Concentrations of carbon monoxide (above) and total hydrocarbons (below) on the center-line of the combustion chamber of the CTH and TUHH during co-combustion of dried sewage sludge A with wood II and coal II as base fuel. Symbols with a cross indicate measurements with the FTIR where $C_{org} = C_{CH4} + 2 \cdot C_{C2H4}$.



Fig. 4-17: Concentrations of ammonia (above) and HCN (below) on the centerline of the combustion chamber of the CTH and TUHH during cocombustion of dried sewage sludge A with wood II and coal II as base fuel.



Fig. 4-18: Concentrations of nitric oxide (above) and nitrous oxide (below) on the center-line of the combustion chamber of the CTH and TUHH during co-combustion of dried sewage sludge A with wood II and coal II as base fuel.



Fig. 4-19: Concentrations of sulfur dioxide on the center-line of the combustion chamber of the CTH and TUHH during co-combustion of dried sewage sludge A with wood II and coal II as base fuel.

As seen in Fig. 4-17 the concentrations of ammonia are more important than those of hydrogen cyanide. The concentration of hydrogen cyanide is about one order of magnitude lower than that of ammonia. There is a substantial contribution from the sludge, as expected, considering the high fuel-nitrogen content.

Fig. 4-18 shows both the concentrations of NO and N_2O . Note the differences in scale in the two sets of figures. The large scale for NO makes the exit concentrations (shown in other sections in this report) disappear, at least for the pure fuels. There is a strong influence of the sludge with its high nitrogen content, and the NO concentration is extremely high in the locations where combustion takes place, in the bottom bed. As the gas moves upwards, NO is reduced and the concentration falls.

In analogy with the concentration of HCN (Fig. 4-17), the principal precursor of N_2O , the concentration of N_2O is low. As measured several times before, the N_2O concentration increases on the way of the gas up in the riser of the CTH boiler, but for some reason this process is somewhat different in the TUHH combustor. Furthermore, it is seen in both combustors that additional N_2O is formed by the secondary air injection in the afterburner despite the fact (Fig. 4-17) that HCN is zero there. It is not clear how this N_2O was formed in the afterburner. Ammonia is the only gaseous precursor present.

The sulfur dioxide concentrations recorded in the furnace of the CTH boiler and shown in Fig. 4-19 indicate a good sulfur capture at the lower part of the combustor if limestone is added. In the case of wood no new limestone was added and a peak concentration recorded at the bottom of the boiler could have some connection with release and recapture of SO₂ from old limestone unintentionally remaining in the bed from previous tests. With increasing height, and especially across the cyclone, the SO₂ concentrations increase again. There are several reasons for such a behavior, such as conversion of H₂S, but this matter was not further investigated and no definite explanation can be given. The limestone particles tend to remain in the bottom part, and thus, SO₂ produced in the upper part could not be captured again.

4.1.5 The importance of the cyclone as a burn-out system

One goal of the present work is to investigate the role of the cyclone and the afterburner chamber as a burn-out zone downstream of the combustion chamber. The comparison presented in Tab. 4-1 relates the sizes of the two experimental units to a large industrial CFB combustor. One of the two boilers operated by Stadtwerke Flensburg in Flensburg/Germany was chosen as an example. The analysis of the design and operating data shows that the average gas residence time in the Flensburg combustion chamber is about 3.77 s, whereas the residence time in the burn-out zone, i.e. in the cyclone, is only slightly smaller. The gas residence time in the CTH combustion chamber is 2.2 s and in the cyclone 0.9 s. If the residence time in the exit duct of the cyclone is added a total residence time in the burn-out zone of 1.6 s is obtained, which results in a ratio $(\tau_c + \tau_{ac})/\tau_{cc}$ of 74 % compared to 84 % in the Flensburg unit. The gas residence time in the cyclone time in the cyclone compared to 84 % in the Flensburg unit. The gas residence time in the combustion chamber. In order to obtain conditions that are comparable to those of an industrial unit, the separate after-burner chamber with a diameter of 0.3 m and a total length of 4.5 m

was added to the TUHH unit (cf. Chapter 2.1). This made the gas residence time in hot conditions downstream of the riser roughly identical to that in the Flensburg cyclone.

	TUHH	CTH	Flensburg
volume of combustion chamber, m ³	0.127	31.4	588
volume of cyclone including the			
entry duct, m ³	0.024	12.4	492
volume of after-burner chamber, m ³	0.133	10.7	-
gas residence time in combustion			
chamber, τ_{cc} , s	2.58	2.2	3.77
gas residence time in cyclone, τ_c , s	0.49	0.9	3.15
gas residence time in after-burner			
chamber, τ_{ac} , s	2.70	0.8	-
gas residence time in burn-out zone,			
$\tau_c + \tau_{ac}, s$	3.19	1.6	3.15
ratio of gas residence times			
τ_c/τ_{cc}	0.19	0.4	0.84
$(\tau_{c+}\tau_{ac})/\tau_{cc}$	1.24	0.7	0.84

Tab. 4-1: Gas residence times in CFB units of different scales

Fig. 4-20 and Fig. 4-21 show cases similar to those in Fig. 4-16 and Fig. 4-18 but related to gas residence time instead of height. The purpose is to show the progress of reactions, especially in the cyclone and in the afterburner.

Fig. 4-20 compares the effect of air staging in the TUHH combustor during introduction of secondary air after the cyclone for three coal/dried sludge mixtures. Until the addition of secondary air, the level of CO is high regardless of fuel mixture, but then it rapidly drops two orders of magnitude inside the afterburner. Similar to CO, the nitrous oxide emission is not clearly dependent on the fuel blend. In the cyclone it is strongly reduced, but after secondary air injection the concentration increases again to values even higher than at the top of the riser section. Since no HCN is measured at gas residence times larger than 2 s, there is only some ammonia that could have produced N₂O after that time, but ammonia is not a very important precursor. The NO concentration is already strongly reduced in the combustion chamber itself, and further significant reduction takes place both in the cyclone and in the afterburner. The relative reduction increases with the sludge content in the fuel. The role of the afterburner is obviously not only to oxidize CO, but a significant decrease of NO takes place here, too. The combination of cyclone and afterburner in the small pilot plant simulates the large-scale cyclone in an industrial power plant. Consequently, the cyclone of a CFB boiler may be used as a burn-out system to lower the gaseous emissions, and this is particularly true for co-combustion with biogenous fuels.



Fig. 4-20: Pollutant reduction in cyclone and afterburner during advanced staging. TUHH unit. Coal II and dried sewage sludge A. (♦ coal II, ● 15 % dried sludge A, ▼ 50 % dried sludge A).

Not only in the case of advanced staging, but also during normal staging, cyclone and afterburner contribute to emission reduction. In Fig. 4-21 gas profile measurements for advanced and normal staging are compared. The differences in gas flow are small and the gas residence time depends only slightly on staging method. The arrow marks the location of the secondary air injection in the advanced staging case. During normal staging, secondary air is injected at a height of 3.05 m above the air distributor. Residence times below 3 s belong to the furnace and concentrations recorded at a residence time of about 3.7 s correspond to the measurement port right after the cyclone. The other points are taken in the afterburner.



Fig. 4-21: Influence of cyclone and afterburner during advanced and normal staging. TUHH unit. Coal II and 15 % dried sewage sludge A. (□ normal staging, ○ advanced staging).

The CO concentrations at the top of the riser in the case of normal staging are one order of magnitude lower than in the advanced staging case. However, they are still almost 1000 ppm, and only further reduction in the cyclone allows keeping the legal limits. For both staging techniques, CO values below 100 ppm are obtained after approximately 2.5 s in the afterburner. In both cases the relative NO reduction from the top of the riser to the exit of the cyclone is very large, roughly 100 ppm. In the afterburner, however, the NO reduction slows down for normal staging, whereas the NO concentration gradient remains steep for advanced staging. Finally, after approximately 2.5 s in the afterburner,

the NO emissions of both staging techniques are close together, and, except for some scattering, they do not change significantly anymore.

Thus, the optimum total gas residence time concerning CO and NO emissions is between 6 and 7 seconds. The emissions reported previously for the TUHH plant were withdrawn from a port in the after-burner corresponding to 6.8 s total gas residence time.

The concentration patterns of nitrous oxide differ greatly for the two investigated staging techniques. While in the case of normal staging N_2O is further reduced in the afterburner, the nitrous oxide concentration rises after secondary air injection during advanced staging. However, the final N_2O emission for normal staging is twice as high as for advanced staging.

4.1.6 Influence of bed material

Sewage sludge may contain considerable quantities of metals that can serve as catalysts for the gaseous emissions. For instance the present sludge contains 74 gram of iron per kg ash in the form of oxide or iron converted into oxide in the bed. This can be compared with the corresponding quantity of 8 g/kg ash for the coal used. The effect is observed visually, since the ashes are colored red. Iron oxide serves as a catalyst for oxidation of ammonia (released from the fuel) to NO. The possible catalytic effect cannot be isolated in the co-combustion tests carried out, but catalytic effects can have been present. The catalytic effect has been identified in a special test, where iron oxide powder (Fe₃O₄ with an average size of 20 μ m) was introduced into the cyclone of the CTH boiler



Fig. 4-22 Addition of iron oxide (Fe₃O₄) to the CTH combustion chamber during combustion of coal II under normal operating conditions. Between t=1.42 and t=2.30 the average iron oxide addition is 485 kg/h which is equal to 2.8 kmol/h.

during combustion of coal, Fig. 4-22. The figure shows the weight of the feed hopper during addition of iron oxide at a constant feed rate of 485 kg/h or 2.8 kmol Fe/h between the hours 1.42 and 2.30. The feed rate was 4.8 times higher than the iron supply by the sludge. The effect was seen as an immediate rise of the NO concentration from 100 ppm to 300 ppm. When the supply of iron oxide was stopped, the NO emission gradually returned to its original level as the iron disappeared. There are no data to relate this experience to the present sludge results, but the existence of catalytically active species in the sludge may affect the emission of nitrogen oxides especially.



Fig. 4-23 Relative increase of input flow to the boiler of ash components as function of sludge supply with wood (Figs. a,b) or coal (Figs. c,d) as base fuels. The vertical scales are different for enriched (Figs. a,c) and for less enriched (Figs. b.d) elements.

4.1.7 The fate of trace elements

A number of trace elements have been selected for comparison with the regulations of the European Commission for emissions of trace elements related to co-combustion of sludge with conventional fuels. These trace elements are Cd, Tl, Hg, Sb, As, Pb, Cr, Co,

Cu, Mn, Ni and V. The differences in the feed rate of trace elements when substituting coal or wood with sludge are particularly important in the present tests. A comparison of this supply of trace elements with that of the sludge, accounting for the concentrations according to Tab. 2-2 and the energy contents of coal, wood and sludge, is found in Fig. 4-23. For these particular fuels, the replacement of coal with sludge leads to an enrichment of Hg, Cu, Cr, Cd, Sb and As in the input ash flow, whereas Mn, Ni, Pb, Co, V and Tl are not significantly enriched compared to pure coal. The replacement of wood with sludge leads to higher enrichment of trace elements than for coal as base fuel because of the low ash content in wood and the lower levels of most of the trace elements in the ash. Cd and Mn are exceptions. For these species the relative increase of the input flow is higher using coal as base fuel than for wood, Fig. 4-23.

First, mass balances are given in order to illustrate the degree of accuracy of the measurements, and then material flows and species concentrations are presented.

The total mass balance of solid material over a fluidized bed combustor can be expressed as:

mass flow of fuel ash and lime = mass flow of bottom ash and fly ash

Here conversions of the lime and unburned char in the exit flows are taken into account and no additional feeding of silica sand was done during the test period. The species balances are based on total mass flows and measured species concentrations. These balances can be expressed as:

mass flow of species X = mass flow of total solids * concentration of X

Only small amounts (from a mass balance point of view) of species are found in the gas phase, so the closure of these balances depends on the total solids mass balance and on the measurements of species concentration.



Fig. 4-24: Ash recovery fraction (out/in*100) for wood (left) and coal (right) as base fuel.

The total mass balances shown in Fig. 4-24 verify that the closure has been fairly good. Only in the case of pure wood the balance is not closed in a satisfactory way. This is caused by the very small ash flows that could not be recorded accurately. In order not to bias the species balances due to deviations in the total mass, the total mass balances are assumed to be closed to 100 % when calculating the species balances presented in Fig. 4-25.



Fig. 4-25: Recovery fractions (out/in*100) for Hg, Pb, Cd, Cr, Cu, Ni, Mn, Co, As, Sb, V and Tl. The recovery fraction for Hg, 0 % sludge and coal as base fuel is 544 %. The recovery fractions for Tl with coal as base fuel are 1500-1700 %.

The species balances show deviations from perfect closure, but despite this they should be considered satisfactory with a few exceptions. In the case of pure wood also the species balance suffered from the low ash flows mentioned above. Also in the case of pure coal more Hg, Cd, Cr, Cu and As were found in the exit flows than what was added with the fuel. The reason is most probably that this test was performed after the test with 46 % sludge, and 24 hours of operation between the tests was not sufficient to attain steady state regarding the concentrations of these species in the bed. Finally, for Tl the analysis

of the concentration of in the fuel is probably too low. However, as a whole the closure of the mass and species balances should be considered as satisfactory.

The solids flows leaving the unit (now expressed in actual kg/h) show a situation that is typical for a CFB: about 70 % leaves as fly ash, split into secondary cyclone and bag filter flows, and only 30 % is bottom ash, Fig. 4-26. The increase in solids flow is clearly seen. (The very small ash flows for pure wood are seen in the diagram. In fact, because of the very small flows, the impact of make-up bed material reflects itself as a higher bottom ash flow (74 %) compared to the fly ash flow (26 %). This is partly due to the addition and removal of bed material to avoid accumulation of alkali compounds that could increase the risk for bed agglomeration.)



Fig. 4-26: The solids flows through the boiler for the two test series with wood (left) and coal (right) as base fuels.

The flow of species can be represented in the same way as the total solids flow, but a plot of the species concentrations may give some more information. This is seen in Fig. 4-27 to Fig. 4-31, whose vertical scales are different, adapted to the maximum concentration of each species. However, even in this form the interpretation of the data is not straightforward. A few comments are necessary.

In general the concentrations are lower in the bed than in the fly ash. This depends in most cases on the dilution of the bed by the make-up bed material, silica sand. Most of the ash, but not the sand, is elutriated. Only in the case of volatile ash species, such as Hg (Fig. 4-27) the low concentration in the bed (in this case zero) depends on the gaseous state of the material. Condensation has taken place to a large extent at 150°C (in the secondary cyclone and bag filter). The reason why the concentration is higher in the bag filter may be that the size of the ash particles is smaller there, hence providing more surface for condensation. Similar registrations in the TUHH unit resulted in lower mercury concentrations in the ashes at 500°C, so the condensation temperature is important.



Fig. 4-27: Concentration of the enriched species Hg in the exit streams for test series with wood (left) and coal (right) as base fuels. Comparison of tests with various amounts of sludge.

The enrichment and the amount of ash in the base fuel are other important factors that influence the representation of the species concentration (cf. Fig. 4-23). Because of these factors the Hg concentration is higher for wood as base fuel than for coal, Fig. 4-27, and the same trend is seen for the other enriched species in Fig. 4-28, whereas there is a smaller impact for less enriched species such as Pb, Mn and Co (Fig. 4-29 and Fig. 4-30).

An important property of the CFB is the homogeneity of the circulating bed. This has been clearly verified by the fact (not shown in the diagrams) that the concentrations of species measured in the bed were identical to those measured in the recirculating bed material returning from the cyclone.



Fig. 4-28: Concentration of the enriched species Cu, Cr and Cd in the exit streams for two test series with wood (left) and coal (right) as base fuels. Comparison between tests with various amount of sludge.



Fig. 4-29: Concentration of the species As, Sb and Pb in the exit streams for two test series with wood (left) and coal (right) as base fuels. Comparison between tests with various amount of sludge.



Fig. 4-30: Concentration of the species Ni, Mn and Co in the exit streams for two test series with wood (left) and coal (right) as base fuels. Comparison between tests with various amount of sludge.


Fig. 4-31: Concentration of the species V and Tl in the exit streams for two test series with wood (left) and coal (right) as base fuels. Comparison between tests with various amount of sludge.

The heavy metals are mostly found in the ashes as illustrated by previous balances and diagrams. However, a minor part leaves with the flue gas in the form of emissions measured after the bag filter. These emissions are shown in Tab. 4-2 and Fig. 4-32. It can be concluded that the emissions from the CTH boiler, including the Hg emission, are usually below the emission limits set by the European Union [12] even at a high sludge fraction and independent of the base fuel.

	wood 1	wood 2	wood 3	coal 1	coal 2	coal 3
energy from sludge, %	0	15.5	52.2	0	12.8	46.3
emissions,						
µg/Nm ³ @ 6% O ₂ , dry						
Hg	0.06	0.35	0.30	0.12	0.13	0.23
Pb	1.23	1.17	0.55	4.08	1.50	0.68
Cd	0.04	0.10	0.03	0.10	0.17	0.04
Cr	2.48	1.39	1.36	1.62	1.51	1.42
Cu	4.17	6.86	3.86	20.58	5.87	2.91
Ni	2.17	16.89	19.26	6.00	21.28	7.57
Mn	5.12	2.78	7.60	11.95	4.76	5.40
Со	2.07	0.15	0.11	0.08	0.25	0.11
As	5.43	8.10	7.55	5.29	4.01	5.88
Sb	0.78	0.63	0.58	0.55	0.58	0.70
V	14.50	6.90	5.20	9.10	10.80	12.30
Ti	0.80	0.60	0.60	0.50	0.50	0.70

Tab. 4-2: Emissions of heavy metals



Fig. 4-32: Emissions of heavy metals in relation to emission limits proposed by the European Commission for co-combustion of sludge with wood (left) and bituminous coal (right).

The mechanisms for removal of Hg and Cd (the most volatile heavy metals) in the fluidized bed application need to be further investigated. It is important to find out the role for removal of Hg and Cd of various types of particles in the CFB, including those of the textile filters. Finally, it should be emphasized that the gas concentration of Hg has been measured by two independent methods. Both methods gave the same results. One of the methods is the SEMTECH on-line analyser. This analyser shows the instantaneous Hg concentrations. Fig. 4-33 gives an example of a one-hour recording of the flue gas concentration. The periods measuring elemental and gaseous Hg are indicated in the figures. The range of Hg is on the lower limit of calibration, and no difference

between the forms of Hg is detected. Apart from this, the concentration of Hg fluctuates between zero and 8 mg/Nm³ both gradually and as spikes. One could interpret this as an effect of variation of the feed of Hg by the sludge, but this needs further investigation.



Fig. 4-33: Concentration of Hg measured with the SEMTECH analyzer. Hg^{tot} = gaseous Hg concentration; Hg^o = elemental Hg concentration. Fuel: 46 % sludge with bituminous coal as base fuel.

The conclusion of this study on heavy metals is that although the flows of heavy metals increase with addition of sludge they are mostly found in the ashes, and the gaseous emissions are below the limits of the EU regulations. This is the case also for the most volatile species Hg and Cd. The reasons for this favourable capture of Hg and Cd should be investigated further.

4.2 Legal requirements for co-combustion

Knowledge of the combustion process makes it possible to better understand the influence of the operating conditions on the emissions. However, although the emissions can be influenced, finally the legal requirements determine whether and to what extent cocombustion is feasible. Therefore it is important to compare the present results with emission regulations.

4.2.1 Mixing rules for emission limit values in co-combustion

If co-combustion is applied in an existing power plant the emission limits may have to be revised. The new limits may differ from those previously applied for the plant. According to the new EU Directive on the incineration of waste (2000/76/EC, [12]) the

resulting emission limits for co-incineration are a weighted average of the limits for burning the pure base fuel, e.g. coal, and for incinerating the pure waste, e.g. sewage sludge. The weighting factor needed to determine the mixed emission limits is the fraction of flue gas produced during mono-combustion of either the base fuel or the waste. Thus, the emission limit (EL) for each relevant pollutant i in the exhaust gas, resulting from co-incineration of waste, is calculated as follows:

$$EL_{i,mix} = \frac{V_w \cdot EL_{i,w} + V_{bf} \cdot EL_{i,bf}}{V_w + V_{bf}}$$
(3)

where

- V_w : exhaust gas volume resulting from the incineration of waste only and standardized at the conditions given by the EU Directive, m³/h
- V_{bf} : exhaust gas volume resulting from the combustion of the authorized fuels normally used in the plant (wastes excluded) determined on the basis of oxygen contents at which the emissions must be standardized as laid down in European or national regulations, m³/h
- $EL_{i,w}$: emission limit for pollutant *i* given for plants intended to incinerate waste only, mg/m³ at standardized conditions given by the EU Directive
- $EL_{i,bf}$ emission limit for pollutant *i* given for plants for certain industrial sectors (e.g. for power plants), mg/m³ at standardized conditions for the relevant industrial sector as indicated in Annex II of the EU Directive

The standard conditions of the EU Directive [12] for combustion of waste are given in Article 11, No. 8 (a) as temperature 273 K, pressure 101.3 kPa, 11 % oxygen and dry gas. The same conditions hold for combustion plants with the exception of an oxygen off gas concentration of 6 % mentioned in Annex II.2 of the same directive.

In Germany the standardized conditions for waste incineration plants are largely identical to the European regulation. However, the reference oxygen concentration for fluidized bed combustors burning coal is set to 7 %, dry gas, by the 13th Federal Emission Protection Law (13. BImSchV, [13]).

Due to the different standard oxygen concentrations in the exhaust gas, related either to waste incinerators or to power plants, a mixed reference oxygen concentration has to be determined in the case of co-combustion:

$$C_{O_2,mix} = \frac{V_w \cdot C_{O_2,w} + V_{bf} \cdot C_{O_2,bf}}{V_w + V_{bf}}$$
(4)

where $C_{O2,w}$ and $C_{O2,bf}$ are standard oxygen concentrations in the off gas of waste incineration plants and power plants, respectively. The gas concentrations of the pollutants actually measured at the stack of the power plant at a distinct air ratio have to be recalculated to this new mixed reference oxygen concentration in order to compare with the legal emission limits:

$$C_{i@O_2,mix} = \frac{21 - C_{O_2,mix}}{21 - C_{O_2,meas}} \cdot C_{i,meas}$$
(5)

The legal limits are kept as long as $C_{i@O_2,mix} \leq EL_{i,mix}$ holds.

4.2.2 Range of application of the mixing rules

Whether the mixing rules are applicable to a distinct case of co-incineration or not depends on type and amount of waste to be co-incinerated. If a certain amount of waste is exceeded, the power plant may be classified as a waste incineration plant and, therefore, all its off-gas becomes subject to the more severe emission limits for waste incineration.

While the weighting factor for the mixed emission limits is the exhaust gas volume of each fuel, the range of application of the mixing rules is related to the lower calorific value H_u of each fuel. The contribution of waste – including its auxiliary fuel to sustain the waste incineration – to the total resulting heat release may be defined as:

$$\xi_{w} = \frac{\left(\dot{m}_{w} \cdot H_{u,w} + \dot{m}_{aux} \cdot H_{u,aux}\right)}{\left(\dot{m}_{w} \cdot H_{u,w} + \dot{m}_{aux} \cdot H_{u,aux}\right) + \dot{m}_{bf} \cdot H_{u,bf}}$$
(6)

where

 \dot{m}_W : feed rate of waste, kg/h

- \dot{m}_{aux} : feed rate of auxiliary fuel to sustain waste combustion under mono-combustion conditions, kg/h
- \dot{m}_{bf} : feed rate of base fuel, kg/h
- H_u : lower calorific values of waste, auxiliary fuel and base fuel, respectively, at raw conditions, MJ/kg

For each value of ξ_w it is possible to calculate a unique emission limit $EL_{i,mix}$, below which the actual measured pollutant concentration must be kept.

It is not fully clarified in legislation, when a waste can be regarded as capable of sustaining its combustion or when an auxiliary fuel is needed. The definition of self-sustaining combustion of a specific waste becomes important when the contribution of the waste to the resulting heat release ξ_w is to be computed. For a low calorific value $H_{u,w}$ the waste contribution ξ_w becomes low. From the viewpoint of the power plant operators, a low contribution of the waste to the heat release is desirable, since in this case the mixed emission limit $EL_{i,mix}$ approaches the less severe values applicable to power plants. This would lead to the contradictory effect that co-combustion of low-grade wastes allows the application of high emission limits.



Fig. 4-34: Sketch of an adiabatic combustion chamber. 9 temperature, m mass flow; index FG - flue gas, a - ash.

A reasonable way of defining self-sustaining combustion is to set the combustion temperature to a fixed value, namely the value attained for adiabatic combustion for a given set of operating conditions, e.g. in the case of fluidized bed combustion the temperature could be fixed to 850 °C. Since no heat losses are allowed to occur in an adiabatic combustion chamber as sketched in Fig. 4-34 the incoming enthalpy flux h' must equal the leaving flux h'':

$$h'(\mathcal{G}_{air},\lambda,\xi_w) = h''(\mathcal{G}_{ad},\lambda,\xi_w) \tag{7}$$

Here, h' includes the heat generated by combustion which is described be the heating value H_u . Setting the base temperature \mathcal{G}_o equal to the temperature of the entering fuel \mathcal{G}_{fuel} , the incoming enthalpy flux h' can be computed as

$$h'(\mathcal{G}_{air},\lambda,\xi_w) = H_u(\xi_w) + \lambda l_{\min}(\xi_w) [h_{air}(\mathcal{G}_{air}) - h_{air}(\mathcal{G}_o)]$$
(8)

where l_{min} is the air ratio at stoichiometric combustion and \mathcal{G}_{air} is the temperature of the pre-heated air. The out-going enthalpy flux h'' depends on the composition of the flue gas at stoichiometric combustion, denoted by an asterisk, and on the excess air ratio λ :

$$h''(\mathcal{G}_{ad},\lambda,\xi_{w}) = \mu_{v}^{*}(\xi_{w}) \Big[h_{v}^{*}(\xi_{w},\mathcal{G}_{ad}) - h_{v}^{*}(\xi_{w},\mathcal{G}_{o}) \Big] + (1-\lambda)l_{\min}(\xi_{w}) \Big[h_{air}(\mathcal{G}_{ad}) - h_{air}(\mathcal{G}_{o}) \Big] + a(\xi_{w}) \Big[h_{a}(\xi_{w},\mathcal{G}_{ad}) - h_{a}(\xi_{w},\mathcal{G}_{o}) \Big]$$

$$(9)$$

where μ_v stands for the specific mass of flue gas produced per mass of fuel burned and *a* represents ash. If both the entering and the leaving enthalpy fluxes are known, the adiabatic temperature can be calculated according to eq. (7), as illustrated in Fig. 4-35.



Fig. 4-35: Determination of the adiabatic combustion temperature. Mono-combustion of wet sewage sludge (coal used as auxiliary fuel, coal fraction in mixture: 3.9 wt.-%).

For a desired combustion temperature represented by \mathcal{G}_{ad} and for a given excess air ratio λ , the value of ξ_w can be computed. If ξ_w and the lower heating values of the fuels and the waste are known, the mass flow of auxiliary fuel can be determined by eq. (4). This can be illustrated by an example. If mechanically de-watered sewage sludge with a lower heating value of 1.5 MJ/kg is mono-combusted with preheated air (\mathcal{G}_{air} =400 °C) at an excess air ratio of λ =1.2, the resulting adiabatic temperature is about 650°C. Thus, this sludge is not a self-sustaining combustible. However, co-combustion with bituminous coal as the base fuel with a comparatively high heating value of 23.5 MJ/kg is feasible. The contribution of sewage sludge to the total resulting heat release has to be corrected by the amount of coal needed as auxiliary fuel to sustain sludge combustion (compare "energy fraction" and ξ_w in Tab. 4-3). Thus, the mass flow of the base fuel is lowered by the amount of coal needed as auxiliary fuel.

$\frac{mass\ fraction,\ wt-\%}{\frac{\dot{m}_{w}}{\dot{m}_{w}+\dot{m}_{bf}}}$	$\frac{energy fraction, \%}{\dot{m}_{w} \cdot H_{u,w}}$ $\frac{\dot{m}_{w} \cdot H_{u,w}}{\dot{m}_{w} \cdot H_{u,w} + \dot{m}_{bf} \cdot H_{u,bf}}$	$\frac{\xi_{w}, \%}{\left(\dot{m}_{w} \cdot H_{u,w} + \dot{m}_{aux} \cdot H_{u,aux}\right)}$ $\frac{\left(\dot{m}_{w} \cdot H_{u,w} + \dot{m}_{aux} \cdot H_{u,aux}\right) + \dot{m}_{bf} \cdot H_{u,bf}}{\left(\dot{m}_{w} \cdot H_{u,w} + \dot{m}_{aux} \cdot H_{u,aux}\right) + \dot{m}_{bf} \cdot H_{u,bf}}$
58.4	8	13.1
64.2	10	16.4
68.6	12	19.7

Tab. 4-3: Relations of mass supply and energy supply of waste in co-combustion

This discussion seems to be rather theoretical. However, legislation allows a range of variables to be defined individually by the power plant operators, which can decide whether the power plant becomes a waste incineration plant or not with a significant impact on the emission regulations. In Fig. 4-36 the influences of temperature of preheated air and excess-air ratio are shown. Obviously, not only the fuel type, but also the operating conditions in terms of air preheating or excess air ratio determine which legal regulations have to be applied for an individual power plant.



Fig. 4-36: Influence of the temperature of preheated air (left) and of the excess air ratio (right) on the fraction of waste in the total resulting heat input. Co-combustion of mechanically de-watered sewage sludge with bituminous coal. Mass fraction of sludge in the fuel mixture: 64.2 wt.-%; combustion temperature: 850 °C.

The most important quantities influencing ξ_w are the heating values of the fuels in raw conditions. At first sight these values seem to be fixed. However, H_u strongly depends on the condition of the fuel when it is being fed into the combustor. Of special importance is the water content. For instance, drying of the mechanically de-watered sludge from 76 % water content to 10 % increases its heating value by one order of magnitude. Thus, dried sewage sludge will be classified as waste capable sustaining its combustion.

4.2.3 Differences and similarities of local and European legislation

In the near future the differences in the emission regulations for power plants and waste incinerators in the countries of the European Union will be evened out. The new EU Directive on the incineration of waste will accelerate this adjustment process.

At present, the emission limits for power plants in Germany are determined by the 13th Federal Emission Protection Law (13. BImSchV, [13]), while for waste incineration plants the 17th Federal Emission Protection Law holds (17. BImSchV, [14]).

In the European Union a European Parliament and Council Directive on Incineration of Waste [12] is established, which is strongly influenced by the regulations of the 17. BImSchV. The emission limits given in the EU Directive depend on the capacity of the power plant.

				German legislation		
	EU Directive			13. BImSchV	17. BImSchV	
	combustion plants		waste	combustion	waste incineration	
			incineration	plants		
	solid fuels	biomass				
CO,	local		50	250	50	
mg/m ³	directives					
NO _x ,	300	300	200	800	200	
mg						
NO_2/m^3						
SO _x ,	525	200	50	400	50	
mg SO_2/m^3						
Hg,	0.05	0.05	0.05	-	0.03	
mg/m ³						
ref. O ₂ ,	6	6	11	7	11	
vol-%						

Tab. 4-4:Emission limits for CO, NO, SO2 and mercury. All concentrations refer to dry
flue gas at standard conditions. Given are the limits for the daily mean values.

Tab. 4-4 presents a comparison between the local emission limits in Germany and the European legislation for a plant of 200 MW_{th} . In Sweden there are no fixed values, emission limits are subject to judgement from case to case, but in general the levels are similar to those in the table.

For waste incineration the directive of the EU, as well as the 17. BimschV, specify identical pollutant concentration limits except for mercury, where the German directive is slightly more severe. However, for combustion plants the EU Directive contains NO_x

emission limits, which are much lower than the corresponding German ones. This is because the EU Directive has been published in 2000 and could therefore consider more recent developments in emission reduction. Regarding the sulfur dioxide limits, the EU Directive distinguishes between solid fuels, such as coal, and biomass. The SO₂ limits for biomass combustion are much lower than those for fossil fuels. The EU Directive does not give CO limits for combustion plants explicitly but refers to local directives. Below, the values of the 13. BImSchV are adopted.

The mixing rules, defined above and applied to co-combustion of the bituminous coal and dried sewage sludge described in Chapter 2.3, result in sliding emission limits, as shown in Fig. 4-37.



Fig. 4-37: Sliding emissions and reference oxygen content for co-combustion of dried sewage sludge with coal. Comparison of EU and German directives.

In the German regulations there are two additional restrictions. If the energy supply of waste (ξ_w) is below 10 %, emissions limits have to be kept as if the energy fraction of the waste were exactly 10 %. If the waste energy contribution exceeds 25 %, the power plant is considered a waste incineration plant and the limits of the 17. BImSchV for pure waste incineration have to be applied.

A diagram, such as presented in Fig. 4-37 is unique for each co-combustion case, since the amount of flue gas depends on the composition of the fuel, and the amount of flue gas influences the mixed emission value. In addition, different heating values change the energy fraction ξ_w supplied by the waste.

4.3 Comparison of legal requirements and measured emissions

The measured emission data from the two plants are compared with the regulations for coal/dried sludge in Fig. 4-38, wood/dried sludge in Fig. 4-39 and coal/wet sludge in Fig. 4-40. Since the reference oxygen concentration of the EU Directive deviates from the German regulation (Fig. 4-37) the recalculated emission values which are based on the actual reference O_2 concentration are also differing. However, the deviations are generally small.

The reasonable similarity in emission results from the two plants, seen in Fig. 4-38 and Fig. 4-39, should be pointed out. Despite the large difference in the size of the two plants, the same tendencies for the emissions were recorded.

In general, the EU Directive is less severe than the German regulation, but in both sets of regulations and in all cases investigated a content of sludge energy of less than 25 % satisfies the regulations. This is a very satisfactory conclusion, since in practical applications of co-combustion probably only minor quantities of additional fuel are of interest. The only exception is the sulfur dioxide emissions during co-combustion with wood II. The peculiar discrimination of SO₂ emissions from biofuel (see Tab. 4-4) makes it more difficult to obey the EU Directive in the case of SO₂ emission during co-combustion with wood as base fuel (Fig. 4-39). As has been pointed out above, this depends on the sulfur content in the sludge, which requires the application of some sulfur removal arrangement. Lime was added in connection to the tests shown in Fig. 4-39 but the lime efficiency was lower than for coal (Fig. 4-8).

Although there is no regulation for N_2O emissions, the benefits of low nitrous oxide emissions should be appreciated from an environmental point of view. Only with advanced staging the N_2O emission remains on a fairly low level, except for wet sludge/coal.



Fig. 4-38: Emissions measured under co-combustion conditions of coal II with dried sewage sludge A and compared with legal limits; EU limits for a 200 MW_{th} plant; concentrations per m³ at standard conditions based on sliding O₂ concentration according to EU Directive (above, open symbols) and German legislation (below, solid symbols); λ_{primary}=1.05; secondary air addition after cyclone).



Fig. 4-39: Emissions measured under co-combustion conditions of wood II with dried sewage sludge A and compared with legal limits; EU limits for a 200 MW_{th} plant; concentrations per m³ at standard conditions based on sliding O₂ concentration according to EU directive (open symbols) and German legislation (solid symbols); $\lambda_{primary}$ =1.05; secondary air addition after cyclone.

The emission limits for co-combustion of coal/wet sludge along with the measured emissions are depicted in Fig. 4-40. The decrease of the emission limits is not strictly linear, but slightly over-proportional with increasing wet sludge supply. This is due to great differences in the heating values (H_u) and stoichiometric air requirements (L_{st}) of coal and wet sludge. To derive the mixed emission limits the emissions for mono-combustion of the added fuel and the base fuel have to be weighted with the flue gas amount produced by each fuel (eq. (3)). The flue gas volumes should be computed on a dry basis for a specific reference oxygen concentration. The energy fraction of the co-fuel is related to its heating value while the amount of flue gas on a dry basis depends on both the stoichiometric air requirement and the excess air ratio defined by the oxygen off gas concentration. The emission limit graphs in the figures above would follow a linear relationship only if the flue gas volume fraction produced and the energy fraction is altered.

This, however, is only the case if the relative difference of H_u and L_{st} between waste and base fuel is similar and if a unique O₂ off gas concentration is applicable. If, for instance, both the heating value and the stoichiometric air requirement of the co-fuel are half of the value of the base fuel and a single O₂ concentration holds for both fuels, the energy fraction and the flue gas volume fraction on a dry basis would show a linear dependency. Since naturally L_{st} and H_u are linked for a given fuel, one could generally assume a linear relationship provided that the difference of the O₂ reference concentrations is small. This is the case in Fig. 4-38 and Fig. 4-39. However, in the case of wet sludge/coal combustion the heating values differ by a factor of 16 whereas the values of L_{st} differ by a factor of 9 only, and thus a deviation from a linear relationship is obtained in Fig. 4-40.



Fig. 4-40: Emissions measured under co-combustion conditions of coal II with wet sewage sludge A at TUHH and compared with legal limits; EU limits for a 200 MW_{th} plant; concentrations per m³ at standard conditions based on sliding O₂ concentration according to EU directive (open symbols) and German legislation (solid symbols); $\lambda_{primary}$ =1.05; secondary air addition after cyclone.

5 CONCLUSIONS AND RECOMMENDATIONS

Tests in two quite different CFB plants have been carried out with similar results in the two plants. The same fuels were used in the two plants: dried sewage sludge as additional fuel to either coal or wood as base fuel. Some tests were also made with wet sewage sludge and paper fiber sludge.

Dried sewage sludge can be handled together with the base fuels without any technical problems. The co-combustion worked well, employing the conventional fuel feed system, and only small differences from the over-all performance of the base fuels was observed, despite fractions of added fuel of up to 50 % energy. This presumes that the ash handling system of the plant can handle the increased ash flow resulting from the sludge. Moreover, the fly ash from sludge combustion is stickier than ash from coal or wood.

With coal and wood as base fuels German and EU emission limits were not exceeded for energy fractions of additional fuel of less than 25 %.

The emission behavior of the base fuel plays a dominant role, particularly for energy fractions of additional fuel of less than 25 %.

Sewage sludge contains large quantities of nitrogen, and high emissions of NO may occur, especially during mono-combustion. However, the reduction in a CFB combustor is also high, particularly in the presence of char, and emissions are only moderate for energy fractions of less than 25 %; the conversion of fuel nitrogen to NO was only a few percent. Also the conversion to N₂O was relatively small. In the combustion chamber the influence of the nitrogen content is clearly seen in the form of higher concentrations of NO and N₂O at higher sludge fractions.

Advanced staging has been shown to effectively reduce emissions of NO and N_2O without seriously affecting sulfur capture by limestone during combustion of coal in CFB. In contrast to the case of coal it was found that air staging in CFB is not important for the emissions from fuels with a high volatile content, such as wood or sludge. The reason can be that char plays a dominant role for reduction of NO as well as for N_2O in the combustion chamber, and the char content in the bed becomes small during combustion of fuels with a small content of fixed carbon. The oxygen concentration in the bed affects the char concentration, but with a small char concentration in the bed a change in the char concentration does not play a great role for NO reduction. So, in conclusion, for sludge combustion in CFB with a fuel that yields a small amount of char, such as wood, as a base fuel, the arrangement of the air supply is only of minor importance for NO_x reduction.

What is the difference between wood and coal as base fuel? The comparison can be linked to the emission regulations. Both base fuels perform well, but minor differences can be identified: 1) Reduction of NO is in general better with coal than with wood. 2) CO emissions are lower with wood. 3) N_2O emissions are low with wood compared to coal, but the emissions in the two cases become more equal when the fraction of sludge increases 4) The weak point with wood is that sulfur capture with limestone is slightly less efficient than with coal as base fuel. The two fuels have their advantages and

disadvantages relative each other, but the general conclusion is that both are on the whole equally good as base fuels.

A few tests were made with mechanically dewatered sludge, wet sludge. On the whole, wet sludge performed similar to dried sludge with respect to NO emission, but the N_2O emissions were slightly higher. In detail, some minor differences in behavior were found during staging: wet sludge appears to be more sensitive to the staging conditions than dry sludge, In addition, there is a difference in energy content of the two types of sludge, of course.

Due to mixing limitations, high volatile fuels often give rise to high concentrations of unburned gases in the upper regions of a CFB riser. This was the case even in the narrow TUHH combustor, also during combustion of coal, and of course the effect is present in the wider CTH boiler. When the gases enter the cyclone, mixing is improved and the gaseous combustion is enhanced. Burn-out is achieved if the gas residence time is sufficient. It was shown that, after extension of the combustion space with the afterburner combustion chamber, the present plants attained gas residence times that were in the same order as those in a large commercial CFB boiler. The afterburner was also utilized for late addition of secondary air, applying the advanced staging method. For CO (representing combustible volatiles) there was a strong reduction of concentration downstream of the main combustion chamber. This was noted also for NO, but at least for coal with addition of sludge, unexpectedly, the N₂O concentration rose in the afterburner. Ammonia or residual char are not known to be primary precursors of N₂O, but they are the only candidates for N₂O formation remaining in and after the cyclone. This needs to be further investigated. In general, however, the conclusion is that a satisfactory pollutant reduction and burn-out was achieved after a gas residence time of 6 to 7 seconds from the fuel inlet.

The progress of combustion is quite similar in the two research plants, especially as the air addition to the combustion chamber takes place in the bottom and secondary air is only added downstream of the riser, where the mixing is quite intensive (because of the intense swirl created by the cyclone). Therefore, there were no "macroscopic" mixing differences as those observed in the case of normal staging, when the mixing is almost instantaneous in the narrow TUHH reactor but only gradual in the CTH boiler. Most oxygen is consumed in the bottom of the combustion chambers and consequently most combustion takes place there. Although there were minor differences in the progress of combustion in the two combustion chambers, these were evened out in the cyclones and the resulting emissions did not differ much. There was only a very small influence of the sludge addition on the progress of combustion at the same time as a clear effect was seen on the concentrations of nitrogen oxides.

Sewage sludge contains sulfur, to a large extent arising from conversion of sulfurcontaining proteins, but some may remain from the sludge treatment process. The sulfur content of the presently investigated sewage sludge was twice as high as that of the coal used (Tab.2). Sulfur cleaning is necessary. In case of co-combustion with coal in circulating fluidized bed this does not cause any problem because the plant is most likely equipped for sulfur removal by limestone, and the additional sulfur from the sludge is just a marginal increase in the sulfur supply, which is handled with a likewise marginal increase in limestone feed. In a wood fired boiler, on the other hand, there is no equipment for sulfur removal and the sulfur has to be removed from the sludge prior to combustion or by installation of sulfur cleaning equipment. Sulfur capture with limestone is the classical method in fluidized bed combustion. This method can be used also with wood/sludge, but it was found to be slightly less efficient in this case than with coal.

The ash content of sewage sludge is about 45 % of dry matter. This ash contains more trace elements than what is normally found in coal and, of course, much more than is found in wood with its very small content of ash. Emission of heavy metals is one important issue, but the metals may play a role also inside the combustion chamber in the form of catalysts to influence NOx emissions. For instance, the ash of the present sludge contained 74 g/kg ash of iron (to be compared with that of coal, 8 g/kg ash). By dosage of iron powder to the bed, the effect of iron was demonstrated and showed itself as an increase of the NO emission. Such effects could have influenced the results of the investigation, but it was not possible to separate catalytically produced NO from "normal" NO.

Most of the trace elements, such as heavy metals, introduced with the fuels, were found in the ashes. The very small emission of heavy metals recorded after the bag filter was below the emission limits established by the European Commission.

Some measurements were made to study mercury emissions. The principal observations can be summarized as follows:

- The gaseous mercury emissions were higher for advanced staging than with the other staging arrangements. An explanation is not yet found.
- There was almost no mercury in the bed ash, but more and more was found in the fly ash as the temperature falls along the gas path. The TUHH data were measured at 500°C. At normal filter temperatures (150°C) the gaseous emissions are even smaller, and it should be emphasized again, the Hg emission, measured by two independent methods in the CTH boiler, was lower than existing emission limits.
- The more carbon in the fly ash the more mercury is attached to the ash.

Wastes with low heating value (such as, for instance, wet sewage sludge) need an auxiliary fuel to be burned. It is not evident how to treat such an auxiliary fuel in a cocombustion situation with respect to emission regulations. Both the definition of energy content and the adjustment of air ratio and air preheat influence the interpretation of the regulations and may decide if the plant will be classified as a waste incineration plant or a power plant, which has a significant impact on the emission regulations.

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