Biomass Reburning - Modeling/Engineering Studies

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ABSTRACT

This project is designed to develop engineering and modeling tools for a family of NO_x control technologies utilizing biomass as a reburning fuel. The forth reporting period (July 1 - September 30) included ongoing kinetic modeling of the reburning process while firing biomass.

Modeling of biomass reburning concentrated on description of biomass performance at different reburning heat inputs. Reburning fuel was assumed to undergo rapid breakdown to produce various gaseous products. Modeling shows that the efficiency of biomass is affected by its composition. The kinetic model agrees with experimental data for a wide range of initial conditions and thus can be used for process optimization.

Experimental data on biomass reburning are included in Appendix 2. These data were obtained during the reporting period in the scope of a coordinated program funded by the U.S. Department of Agriculture.

No patentable subject matter is disclosed in the report.

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1.0 Introduction

This report describes ongoing efforts to model biomass reburning. This work is a continuation of efforts undertaken at the beginning of the project and described in Quarterly Reports No. 2 and 3 (*Zamansky et al., 1998a,b*). Current work concentrates on description of biomass performance at different reburning heat inputs.

Significant efforts were undertaken to improve the model of natural gas reburning. This work was performed within the scope of the project sponsored by the U.S. Department of Energy under Contract No. DE-AC22-95PC95251 (Project Manager Thomas J. Feeley). Results of these efforts were presented by *Zamansky et al. 1998e*. The kinetic chemical mechanism of the gas reburning process is a cornerstone of the biomass reburning model. Progress in the description of natural gas reburning provided foundation for the development of the biomass reburning model.

Experimental data used for comparison with modeling were obtained within the framework of the Phase II SBIR project “Advanced Biomass Reburning for High Efficiency NO_x Control” sponsored by the U.S. Department of Agriculture. Appendix 2 contains description of the experimental data on advanced biomass reburning.

2.0 Modeling of Biomass Reburning

The model includes 447 reactions of 65 chemical species (*Glarborg et al. 1998*). Since the adopted kinetic mechanism extends to only C_1 and C_2 hydrocarbons, biomass was represented by a mixture of decomposition products, thus assuming rapid breakdown of the fuel. The fuel oxygen was assumed to form CO in the substoichiometric environment. The remaining hydrocarbon component was represented by a mixture of C_2H_6 , C_2H_4 and C_2H_2 . Fuel-N was represented as HCN. The composition of decomposition products corresponds to the biomass ultimate analysis.

The EER chemical kinetic code ODF, for “One Dimensional Flame” (*Kau and Tyson 1987*) was employed to model experimental data. ODF treats a system as a series of one-dimensional reactors. Each reactor may be perfectly mixed (well-stirred) or unmixed (plug flow). Each ODF reactor may be assigned a variety of thermodynamic characteristics, including adiabatic, isothermal, or specified profiles of temperature or heat flux, and/or pressure. Process streams may be added over any interval of the plug flow reactor, with arbitrary mixing profiles along the reactor length. The flexibility in model setup allows for many different chemical processes to be simulated under a wide variety of mixing conditions.

The reburning process was treated as series of four reactors. Each reactor described one of the physical and chemical processes occurring in a boiler: addition of the reburning fuel, NO_x reduction as a result of the reaction with the reburning fuel, addition of overfire air (OFA), and oxidation of partially oxidized products.

The following features of the mixing process were addressed in the modeling:

- Injected gases are available for the reaction over certain period of time (mixing time) rather than instantaneously.
- Injection of reburning fuel results in mixture stratification such that mixture composition in the mixing area is not uniform.

For the reburning fuel jet, the mixing time was adopted to be 200 ms for all biomass fuels. This time is considerably longer than mixing time for natural gas injection (100 ms) estimated using a model of single jet in cross flow. This increase in mixing time was introduced to allow to take into account longer heating and decomposition times of biomass particles. For the overfire air jet, the mixing time was assumed to be 100 ms.

Mixture stratification in the reburning zone, or existence of local fuel rich zones in which concentration of fuel is higher than average, is an important factor that affects efficiency of the reburning process. It is believed (*Luan et al. 1996, Miller et al. 1998*) that this factor is mainly responsible for high levels of NO reduction (40-50%) under slightly fuel rich and even fuel lean reburning conditions observed in experiments (*Kokkinos 1992, Folsom et al. 1991*). The mixing process in the reburning zone was described in modeling by addition of flue gas to the stream of natural gas over mixing time (so-called inverse mixing). Inverse addition of reagents partially addresses the issue of mixture stratification in the mixing area: at first moments of mixing the reaction occurs in a fuel-rich environment and then progresses gradually to the final composition determined by the amount of the injected fuel. This approach was found (*Luan et al. 1996, Glarborg et al. 1998, Zamansky et al. 1998c*) to be most effective in description of the mixing process in one dimensional approximation. Additional fuel stratification in modeling was introduced by assuming that within the reburning zone there is one zone with 25% larger and one zone with 25% smaller concentration of fuel than average. NO reduction was calculated as average of that for these two zones. This approach showed to be effective in description of natural gas reburning (*Zamansky et al. 1998e*).

The current biomass reburning model takes into account ultimate analysis of biomass composition. It includes not only balance of C, H and O, but fuel-N and partially ash analysis as well. Concentration of N in all tested biomass fuels ($0.7\text{--}1.2 \times 10^{-2} \%$) is less than that usually found in coals ($2\text{--}3 \times 10^{-2} \%$). However, this amount of fuel-N can contribute to NO production at large heat inputs of the reburning fuel. It was assumed that fuel-N was present in the form of HCN.

Ash analysis shows that biomass contains many elements (Na, Fe, K, P, S and others) that can affect the reburning process. Chemical interaction of these elements and their compounds with C-H-O-N species at high temperatures is not well defined. Therefore, the current model only took into account the presence of Na in ash. Reactions of Na with components of flue gas have been studied (*Perry and Miller 1996, Zamansky et al. 1998c,d*) in connection with reduction of NO and N_2O emissions in SNCR and reburning processes. It was shown (*Zamansky et al. 1998d*) that Na_2CO_3 additives can improve the efficiency of the reburning process if coinjected with NH_3 . Since the amount of Na found in ash of furniture waste and walnut shells is significant, the presence of Na can affect the performance of these fuels. Reactions of Na species (*Zamansky et al. 1998c*) and thermodynamic data for these species are shown in Appendix 1. These reactions were added to the reaction mechanism used to describe biomass reburning.

The important question is in what form Na is present in biomass. The mineralogical composition of biomass fuel is generally complex and difficult to determine quantitatively. It was found (*Hald 1994*) that in straw most alkali metals are present in a water soluble form (in the form of NaCl or ionically linked to the surface) and only small amounts are in water insoluble form (mostly silicates). However, distribution between forms of Na may be different for different biomass fuels. Since $\text{Na}_2\text{O} \cdot \text{Al}_2\text{O}_3 \cdot (\text{SiO}_2)_2$, found in biomass ash, is insoluble and stable at temperatures relevant to the reburning process (*Hald 1994*), it was assumed in the current model that most sodium found in ash is present in the form of silicate. The amount of sodium present in the form of silicate was determined by the amount of silicate ($\text{Al}_2\text{O}_3 \cdot (\text{SiO}_2)_2$) found in biomass ash and was calculated using biomass ash analysis (Table 2-1, Appendix 2). The rest of sodium was assumed to be present in the form of NaOH. These considerations result in the following amount of NaOH in different biomass fuels:

Furniture waste	1400 ppm
Walnut shells	600 ppm
Willow wood	0

Since chemistry of NaOH decomposition is relatively well defined (Zamansky *et al.* 1998c), it can be easily incorporated into the kinetic model.

Figures 1, 2 and 3 show comparison of modeling predictions with experimental data for different reburning fuels. The model quantitatively agrees with experiments within scatter of experimental data for walnut shells and willow wood and qualitatively agrees with experimental data on furniture waste reburning. High efficiency of furniture waste at low heat inputs of the reburning fuel is well predicted, even though maximum efficiency in the modeling is smaller than that in experiments. A decrease in efficiency at large heat inputs is also predicted.

Figure 1. Performance of furniture waste reburning. Symbols are experiments, line represents calculations.

Modeling shows that there are several factors that determine the efficiency of biomass as the reburning fuel. At low heat inputs, fuels with high concentration of minerals (furniture waste and walnut shells) show NO reduction efficiencies similar or better than that of natural gas. At high heat inputs, fuels with high fuel-N content (furniture waste) show degradation in the reburning efficiency. Willow wood has high nitrogen and low mineral content and as a result is less effective than natural gas at all heat inputs of the reburning fuel.

The fact that the model agrees with experimental data suggests that contribution of heterogeneous reactions to NO reduction is not very significant and NO reduction is mainly determined by reactions in the gas phase.

Figure 2. Performance of walnut shells reburning. Symbols are experiments, line represents calculations.

Figure 3. Performance of willow wood reburning. Symbols are experiments, line represents calculations.

In summary, developed kinetic model of basic biomass reburning agrees with experimental data for a wide range of initial conditions and thus correctly represents main features of the biomass reburning process.

3.0 Future Work

Future kinetic modeling will concentrate on description of advanced biomass reburning. This modeling will be done using current mechanism since it already includes reactions of Na containing species.

During the next quarter, FETC R&D personnel will work on Task #2 CFD Modeling, Task #4 Biomass Preparation Economics, and Task #5 Evaluation of Slagging and Fouling.

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Appendix 1

Thermodynamic Data for Na Species (in NASA format)

NAO2	D=37.2NA	1O	2	0	G	300.000	2000.000	1000.00	1
.24373729D+01	.11708054D-01	-.12465450D-04	.60394798D-08	-.10877028D-11					2
-.68349080D+04	.15175355D+02	.24373729D+01	.11708054D-01	-.12465450D-04					3
.60394798D-08	-.10877028D-11	-.68349080D+04	.15175355D+02						4
NAOH	J12/70NA	1O	1H	100	OG	300.000	2000.000	1000.00	1
.45711116D+01	.61346093D-02	-.76237353D-05	.43706135D-08	-.89064713D-12					2
-.25359026D+05	-.95321963D-01	.45711116D+01	.61346093D-02	-.76237353D-05					3
.43706135D-08	-.89064713D-12	-.25359026D+05	-.95321963D-01						4
NA	L 4/93NA	100	000	000	OG	300.000	2000.000	1000.00	1
.25010442D+01	.00000000D+00	.00000000D+00	.00000000D+00	.00000000D+00					2
.12157060D+05	.42385793D+01	.25010442D+01	.00000000D+00	.00000000D+00					3
.00000000D+00	.00000000D+00	.12157060D+05	.42385793D+01						4
NAO	J12/67NA	1O	100	000	OG	300.000	2000.000	1000.00	1
.36192660D+01	.29441938D-02	-.35206654D-05	.18827273D-08	-.36198896D-12					2
.88821327D+04	.62033018D+01	.36192660D+01	.29441938D-02	-.35206654D-05					3
.18827273D-08	-.36198896D-12	.88821327D+04	.62033018D+01						4

Reactions of Na Species (in Chemkin format, $k = AT^n \exp(-E/RT)$ / cm; mol; s; K)

Reaction	A	n	E
NA+N2O=NAO+N2	1.69E+14	0.00	3159.
NAO+H2O=NAOH+OH	1.32E+13	0.00	0.
NAO+O=NA+O2	2.23E+14	0.00	0.
NAO+NO=NA+NO2	9.04E+13	0.00	0.
NAO+H2=NAOH+H	1.25E+13	0.00	0.
NA+O2+M=NAO2+M	1.74E+21	-1.30	0.
H2O/5/ CO2/3/ CO/2/ H2/2/			
NA+OH+M=NAOH+M	1.82E+21	-1.00	0.
NAO+OH=NAOH+O	2.00E+13	0.00	0.
NAO+HO2=NAOH+O2	5.00E+13	0.00	0.
NAO+H2=NA+H2O	3.13E+12	0.00	0.
NAO+CO=NA+CO2	1.00E+14	0.00	0.
H+NAO2=HO2+NA	2.00E+14	0.00	0.
NAO+H=NA+OH	2.00E+14	0.00	0.
NAO+OH=NA+HO2	3.00E+13	0.00	0.
NA+HO2=NAOH+O	1.00E+14	0.00	0.
NAO2+H=NAO+OH	5.00E+13	0.00	0.
NAO+HO2=NAO2+OH	5.00E+13	0.00	0.
NAO2+H=NAOH+O	1.00E+14	0.00	0.
NAO2+CO=NAO+CO2	1.00E+14	0.00	0.
NAO2+O=NAO+O2	1.00E+14	0.00	0.
NAO+NH3=NAOH+NH2	1.00E+13	0.00	0.
NAOH+H=NA+H2O	1.07E+13	0.00	1967.
NAO2+OH=NAOH+O2	8.00E+13	0.00	0.

Appendix 2

Experimental Data on Biomass Reburning

Experimental data reported here are obtained within the framework of the Phase II SBIR project “Advanced Biomass Reburning for High Efficiency NO_x Control” sponsored by the U.S. Department of Agriculture.

The pilot scale tests were conducted in EER's 300 kW (1×10^6 Btu/hr) Boiler Simulator Facility (BSF) described in detail in previous reports. In the current reporting period test work focused on advanced reburning technologies, involving the coupling of biomass reburning with injection of nitrogen agents and chemical promoters. Biomass test fuels included waste furniture pellets and Willow wood.

2.1 Test Methods and Conditions

Three AR technologies tested were: AR-Lean, AR-Rich, and reburning + SNCR. With AR-Lean the nitrogen agent is co-injected along with the secondary air. With AR-Rich the nitrogen agent is injected into the fuel-rich reburn zone. With reburning + SNCR the nitrogen agent is injected downstream of the OFA. Test variables included additive injection temperature, N-agent to NO stoichiometric ratio (NSR), and sodium promoter concentration. A high-sulfur bituminous Illinois coal was primarily used as the main fuel, with a few tests conducted with natural gas for comparison.

Two N-agents were tested, including ammonia and urea. Each N-agent was formulated as an aqueous solution and injected by means of a twin fluid nozzle. For the AR-Lean tests the overfire air itself served as the atomization medium. Sodium was added in the form of Na₂CO₃, and was dissolved into the N-agent solution. For all tests the reburn fuel was injected at 1700 K. Reburn heat input was 10%, corresponding to a reburn zone stoichiometry of 0.99. This ratio was shown to be optimum in previous test work. Primary zone stoichiometry was 1.10 and final burnout zone stoichiometry was 1.15.

The test work focused on the biomass fuels believed to be most promising for commercial application, i.e. waste furniture pellets and Willow wood. Table 2.1 presents analytical data for each of the solid fuels. Each biomass fuel was prepared by pulverizing it in a hammer mill. The size distribution of the furniture pellets was such that 48% passed through a 200 mesh sieve. The Willow wood was more fibrous and gave larger size particles, with 23% passing through 200 mesh.

2.2 AR-Lean Test Results

AR-Lean tests were conducted with both furniture pellets and Willow wood. Test variables included the injection temperature of the N-agent + OFA, NSR, and concentration of sodium promoter. Figure 2-1 shows AR-Lean results for furniture pellets as a function of injection temperature with Illinois coal as the main fuel. Urea was the N-agent, and was added at NSR = 0.75. Reburning alone provided approximately 47% NO reduction. With urea, performance improved with decreasing injection temperature. The highest NO reduction achieved was 68% at an injection temperature of 1280 K.

Sodium promoter tests were performed during the furniture pellets AR-Lean tests with both natural gas and Illinois coal as main fuels. Sodium was added at concentrations corresponding to 10, 30, and 50 ppm in the flue gas. As shown in Figure 2-2, as the sodium concentration was increased, NO reduction increased significantly during natural gas firing, but only minimally during coal firing. It is possible that coal constituents such as ash and sulfur may have reacted with and partially deactivated the sodium promoter.

During natural gas firing, sodium significantly reduced emissions of N_2O . Figure 2-3 shows N_2O emissions as a function of injection temperature for different sodium levels. With no sodium, N_2O increased with decreasing injection temperature. For injection at 1310 K, N_2O emissions decreased from 14 ppm with no sodium to 2 ppm at 30 ppm sodium.

Figure 2-4 shows AR-Lean results for Willow wood as a function of injection temperature with Illinois coal as the main fuel. Urea was the N-agent, and was added at $NSR = 1.0$. Reburning alone provided 45% to 50% NO reduction. With urea, performance improved with decreasing injection temperature. The highest NO reduction achieved was 76% at an injection temperature of 1280 K.

Figure 2-5 shows Willow wood AR-Lean performance as a function of NSR with urea and aqueous ammonia N-agents. An injection temperature was 1310 K was selected to minimize potential byproduct emissions of N_2O and NH_3 while providing high level NO control. NO reduction increased with increasing NSR and was generally similar for the two N-agents. The highest NO reduction achieved was 78% with aqueous NH_3 at an NSR of 1.4.

2.3 AR-Rich Test Results

AR-Rich tests were then conducted in which the N-agent was injected into the fuel rich reburn zone. Both furniture pellets and Willow wood were tested. Test variables included additive injection temperature, NSR, and sodium promoter concentration. Figure 2-6 shows results of furniture pellets AR-rich tests. Urea was injected at 1390 and 1510 K, and OFA was injected at 1310 K. Performance was slightly better at the lower injection temperature. Maximum NO reduction achieved was 65%.

AR-Rich performance was characterized as a function of NSR with both furniture pellets and Willow wood. Urea injection temperature was 1370 K. As shown in Figure 2-7, performance was generally similar for the two biomass fuels. Maximum NO reduction was 68% for Willow wood AR-Rich at an NSR of 1.6.

AR-Rich tests were performed at sodium promoter concentrations of 10, 30, and 60 ppm. While impacts upon NO reduction were small, sodium did show the ability to control N_2O emissions. Figure 2-8 shows N_2O emissions as a function of NSR. At an NSR of 1.0, N_2O emissions decreased from 14 ppm with no sodium to 6 ppm with 30 ppm or more sodium.

2.4 Reburn + SNCR Tests

Tests were then conducted in which the N-agent was added downstream of the OFA. Test variables included reburn fuel type (furniture pellets and Willow wood), NSR, and N-agent injection temperature. Figure 2-9 shows Willow wood reburn + SNCR test results as a function of urea injection temperature. Reburn heat input was 10%, and OFA was injected at 1450 K. Reburning alone gave about 40% NO reduction. With urea injection, the optimum injection temperature was found to be 1330 K, at which condition NO reduction was 77%.

Reburn + SNCR performance was characterized as a function of NSR with both furniture pellets and Willow wood. Urea injection temperature was 1310 K. As shown in Figure 2-10, with both reburn fuels NO reduction increased with increasing NSR. Better performance was achieved with Willow wood than with furniture pellets. Maximum NO reduction was 83% for Willow wood at an NSR of 1.45. This is the highest NO reduction obtained for any of the AR test conditions.

In general, despite the fact that the Willow wood had a larger particle size distribution, it provided somewhat higher NO control than did furniture pellets at each of the AR test conditions. The Willow wood contains slightly higher volatiles and more alkaline components than the furniture pellets. It was also observed that while sodium

promoter was somewhat effective with natural gas as the main fuel, it had minimal effect with Illinois coal as the main fuel. This is attributed to two effects. First, the relatively high inherent alkaline concentrations in the biomass fuels minimize the benefits of adding additional sodium. Second, the sulfur and fly ash generated by the coal might partially react with and deactivate the sodium. Sodium did, however, prove to be effective at controlling N₂O emissions.

Table 2.1 - Test fuel analytical properties.

Parameter	Units	Illinois Coal	Ohio Coal	Furniture Waste	Walnut Shells	Willow Wood
<u>Proximate analysis</u>						
Ash	% dry	9.60	10.66	1.31	1.93	1.60
Volatiles	% dry	41.15	40.83	79.06	72.02	82.29
Fixed Carbon	% dry	49.25	48.51	19.63	26.05	16.11
Moisture	% as fired	6.00	8.68	6.37	12.43	1.97
<u>Ultimate analysis</u>						
Carbon	% dry	66.48	70.14	53.91	51.00	50.48
Hydrogen	% dry	4.12	4.66	6.07	5.72	5.98
Nitrogen	% dry	1.30	1.32	0.56	0.32	0.53
Sulfur	% dry	3.11	2.49	0.03	0.00	0.04
Oxygen (difference)	% dry	9.60	10.73	38.12	41.03	41.37
Ash	% dry	15.39	10.66	1.31	1.93	1.60
Chlorine	% dry	0.005	0.010	0.190	0.130	0.040
Higher Heating Value	Btu/lb dry	11,210	12,296	8,824	8,624	8,539
<u>Ash analysis</u>						
SiO ₂	% dry	60.54	55.96	13.86	34.98	2.65
Al ₂ O ₃	% dry	18.08	23.99	3.29	6.09	0.41
TiO ₂	% dry	0.87	1.31	6.72	0.21	0.50
Fe ₂ O ₃	% dry	9.45	9.92	8.25	10.71	2.42
CaO	% dry	2.56	2.43	24.10	23.25	41.80
MgO	% dry	0.69	0.73	3.00	3.16	4.80
K ₂ O	% dry	2.55	3.22	7.50	11.15	14.00
Na ₂ O	% dry	0.69	0.66	11.16	6.64	0.24
SO ₃	% dry	2.98	0.67	6.73	1.59	1.80
P ₂ O ₅	% dry	0.26	0.24	2.20	1.92	9.50
SrO	% dry	0.03	0.08	0.13	0.02	0.34
BaO	% dry	0.10	0.24	3.83	0.08	0.06
Mn ₃ O ₄	% dry	0.03	0.00	1.75	0.10	0.18
Undetermined	% dry	1.17	0.55	7.48	0.10	21.30

Figure 2-1. Furniture Pellets AR-Lean Results with Illinois Coal Primary.

Figure 2-2. Furniture Pellets Promoted AR-Lean Results.

Figure 2-3. Furniture Pellets AR-Lean Results with Natural Gas Primary.

Figure 2-4. Willow Wood AR-Lean Results with Illinois Coal Primary.

Fig. 2-5 Willow Wood AR-Lean Performance vs. NSR with Illinois Coal Primary

Figure 2-6. Furniture Pellets AR-Rich Results with Illinois Coal Primary.

Figure 2-7. AR-Rich Results as a Function of NSR with Illinois Coal Primary.

Figure 2-8. Willow Wood AR-Rich N_2O Emissions with Illinois Coal Primary.

Figure 2-9. Willow Wood Reburn+SNCR Results with Illinois Coal Primary.

Figure 2-10. Biomass Reburn+SNCR Results with Illinois Coal Primary.