Oxygen/Steam Gasification of Wood

L. K. Mudge
D. G. Ham
S. L. Weber
D. H. Mitchell

April 1980

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Pacific Northwest Laboratory
Richland, Washington 99352

* Currently employed by:
Southern Union Refinery
Hobbs, NM 88260

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OXYGEN/STEAM GASIFICATION OF WOOD

INTRODUCTION

Pacific Northwest Laboratory (PNL), operated by Battelle, is conducting studies sponsored by the Department of Energy (DOE) on the catalytic gasification of biomass (wood). Catalytic gasification offers the potential advantage of improved gas yields over conventional gasification systems. Due to a lack of data from conventional wood-fed systems, however, direct comparison of the catalytic system with conventional technologies has been difficult. The purpose of the work reported here was to develop basic reference data for oxygen-blown biomass gasification in fixed bed gasifiers.

The need for such reference data arises from the fact that such information has not previously been available in the open literature. An early technique for synthesis gas production used air gasification of wood to yield a producer gas for reduction of iron oxide in the generation of hydrogen by the steam-iron process.⁽¹⁾ The hydrogen was then used in the manufacture of nitrogen based fertilizer. This technique, however, was very inefficient and costly. Recent studies^(2,3) referred to the use of pure oxygen for the direct production of a synthesis gas, but definitive data on yields from the process were lacking. Recent review articles^(4,5) on biomass gasification also did not report data on synthesis gas yields from oxygen/steam gasification of wood.

This study was conducted to obtain reference data on synthesis gas yields from fixed bed gasification of wood with counterflow oxygen/steam mixtures. These conditions are similar to those in the Purox system,⁽²⁾ for which gas yields have been reported using municipal solid waste as a feedstock. Pure oxygen, as employed by the Purox process, could not be used in this study since the resulting high temperatures would cause ash slagging. The gasifier used in these studies cannot operate in an ash slagging mode. We operated over the range of 4 to 1/2 for weight ratios of oxygen-to-steam.

A summary of the results obtained in this work is given immediately below. Major conclusions and recommendations for future work are also itemized. An in-depth report of the experimental apparatus, procedures, and results are then presented and discussed.

SUMMARY OF RESULTS

Yields from fixed-bed gasification of stoker fuel, pressed planar shavings, are presented for air/steam operation and oxygen/steam operation at four different compositions. The wood carbon distributes between gas products and liquid products in the respective ranges of 75 to 80 wt% and 20 to 25 wt%.

The cold gas efficiencies, defined as the combustible energy in the gas divided by the heat of combustion of the wood, were 52%, 61%, 59%, and 52% for operation with oxygen-to-steam weight ratios of 1/2, 1, 2, and 4 respectively. The other energy inputs to the system were less than the energy output in tar products so that the above efficiencies are about the same as the overall efficiencies.

Concentrations of the gas at various heights in the fixed bed are presented. Conclusions about the height of the various zones can be inferred. Temperature profiles are also presented.

Stoker fuel was used instead of wood chips because its bulk density was greater by a factor of 4. Limitations on the maximum volumetric input rate were such that only 50 lb/hr of wood chips could be added. Since gasification rates exceeded this amount, stoker fuel was used. Rates for stoker fuel gasification are presented for the different conditions.

The results indicate that the maximum weight fraction of methanol from dry wood that could be obtained from the synthesis gas is 0.45 at an oxygen-to-steam weight ratio of 1 with reforming of the methane in the gas, or a yield of 135 gallons of methanol per ton of dry wood. The maximum methane yield could be obtained at the same oxygen-to-steam weight ratio of 1 and is 8600 scf/dry ton ($270 \text{ nm}^3/\text{dry MT}$).

CONCLUSIONS

These conclusions are based on the results of this study:

- Maximum yields of synthesis gas for methanol or methane production were obtained with an oxygen-to-steam ratio of 1 and were calculated to be sufficient for production of 0.45 kg of methanol per kg dry wood (135 gal/dry ton) or 270 nm^3 of methane per dry MT (8600 scf/dry ton).

- The fraction of wood carbon that is in the liquid products is high, about 20%. The large amount of tars and water soluble organics is the reason for the low yields of methanol and methane synthesis gases.
- An oxygen requirement of 0.25 lb/lb dry wood was obtained with a weight ratio of 1 for oxygen-to-steam in the blast. Oxygen requirements were higher for other oxygen-to-steam ratios. The maximum was 0.31 lb oxygen/lb dry wood for an oxygen-to-steam weight ratio of 4 in the blast.
- Although no attempt was made to find the maximum wood gasification rate, a rate of 120 lb dry wood/hr ft² was obtained during the testing.
- Concentration profiles in the gasifier show that gasification reactions between the char residue and steam and CO₂ occur exclusively within 18 in. of the grate.
- Concentration profiles show that with air/steam gasification about 60% of the energy in the product gas is generated in the gasification zone and 40% is generated in the pyrolysis zone near the top of the bed of solids.

RECOMMENDATIONS

The results of this study apply primarily to the use of pressed planar shavings. These were used because of their high bulk density. Results will vary slightly with other biomass species. The following oxygen-steam gasification studies are needed for definitive design data:

- Operation with different biomass species and moisture content.
- Operation to determine maximum throughput.
- Operation to determine optimum oxygen-to-steam ratios in the blast.
- Operation to find methods for reducing yield of tars and water-soluble organics.

DESCRIPTION OF EXPERIMENTAL EQUIPMENT

A flow diagram of the equipment used in this study is shown in Figure 1. Figure 2 shows a schematic of the gasifier. The fixed bed gasifier consists of a 7 ft (210 cm) section of 20 in. (50 cm), Schedule 40 carbon steel pipe. The gasifier has a 10 in. (25 cm) opening for addition of feed and a 3 in. (76 mm) opening for the off-gas line. The gasifier is lined with 1 in. (25 mm)-thick KAOWOOL^(a) insulation plus a 3 in. (76 mm) layer of KAOTAB CS^(a) castable refractory. The refractory is 95% alumina, rated for service at 1800°C. The effective inside diameter of the gasifier is about 11 in. (28 cm). The operating height extends 5 ft (150 cm) above the rotary grate for ash discharge. The gasification reactants enter below the grate through traced carbon steel lines.

Thermocouples are provided to record the temperature in the ash zone below the grate, in the product gas line into and out of the scrubber at 8 in. (20 cm) height intervals above the grate along one wall of the gasifier. All temperatures are recorded every 15 min during the test on a Fluke Model 2240A^(b) data logger.

The distributor/compactor in the gasifier is used during the test for determination of the solids level only. It is not used to distribute and compact the bed of solids.

The product gases are scrubbed for removal of condensables in an 8 ft (240 cm) high, 8 in. (20 cm) diameter column packed to a 4 ft (120 cm) height with 1 in. (25 mm) Pall rings. Water containing the scrubbed condensables is recirculated through the scrubber from a 103 gal (390) tank at the base of the column. A 55 gal (210) overflow tank is provided to collect the liquids scrubbed from the product gas. Condensed liquids are weighed and analyzed for total carbon. The oil fraction is analyzed for C, H, N, O, and S.

The product gases are sampled and analyzed with in-line gas chromatographs for H₂, N₂, O₂, CO, CO₂, CH₄, C₂H₄, C₂H₆, C₃H₆, C₃H₈, H₂S, and COS. After sampling and flow measurement, the gases are flared and vented.

(a) Trademark of Babcock and Wilcox
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P.O. Box 43210
Mountlake Terrace, WA 98043

FIGURE 1. Fixed Bed Process Flow Schematic

FIGURE 2. Schematic of Gasifier

Solids are added to the feed hopper which has a capacity of 5.7 ft³ (0.16 m³). The hopper is closed and purged with an inert gas. A 10 in. (25 cm) knife gate valve isolates the bin from the gasifier. The gate valve is opened to drop the solids into a 9 in. (23 cm) diameter auger with a 9 in. (23 cm) pitch that conveys the solids into the top of the gasifier. A volume of about 1 ft³ is added each time, which corresponds to a bed height fluctuation of about 1 ft in the shaft of the gasifier.

RESULTS AND DISCUSSION

Operations were conducted on two separate days and were initiated with an air/steam blast for heatup. Temperature profiles during heatup with air and steam and during testing with different oxygen/steam blasts are given in Figures 3 and 4 for the two days of testing. Each test with oxygen and steam lasted about 4 hours as can be seen from Figures 3 and 4. Tests 1 through 4 were conducted on the first day of operation; tests 5 through 7, on the second day. The ultimate analysis of the stoker fuel is given in Table 1. The bulk density of the stoker fuel was 35 lb/ft³, and the heating value on a dry basis was 8850 Btu (21 MJ/kg).

Conditions and results for the different gasification tests are summarized in Table 2. Feed rates in Tests 1, 2, and 4 are rather uncertain since startup and filling of the reactor was effected during these tests. The average stoker fuel addition rate was difficult to ascertain during startup conditions. The objective of the study was to operate with oxygen and steam; therefore, no attempt was made to obtain steady state conditions with air or air and steam.

TABLE 1. Stoker Fuel Analysis

<u>Element</u>	<u>wt%</u>
C	46.9
H	6.0
O	41.3
N	0
Ash	0.5
Moisture	5.3

FIGURE 3. Temperature Profiles in Tests 1 Through 4

FIGURE 4. Temperature Profiles in Tests 5 Through 7

TABLE 2. Summary of Stoker Fuel Gasification Tests

Item	Test Number						
	1	2	3	4	5	6	7
Feed Rate, lb/hr	38	41	63	37	48	69	80
dry lb/hr	36	39	60	35	45	65	76
dry lb/hr ft ²	57	61	94	55	71	100	120
Pressure, atm	1	1	1	1	1	1	1
Residence Time, hr	2.9	2.7	1.8	3.0	2.3	1.6	1.4
Oxygen Feed Rate, lb/hr	--	--	15	10	--	20	20
Steam Feed Rate, lb/hr	--	5	15	20	5	5	10
Air Feed Rate, lb/hr	50	50	--	--	45	--	--
Oxygen/steam, lb/lb	--	2.3	1	1/2	2.1	4	2
Oxygen/dry Wood, lb/lb	--	--	0.25	0.29	--	0.31	0.26
Steam/dry Wood, lb/lb	--	0.13	0.25	0.57	0.11	0.077	0.13
Air/dry Wood, lb/lb	1.4	1.3	--	--	0.99	--	--
Product Gas Flow							
Rate, dry scfm	--	20	20	11	--	15	20
scf/lb dry	--	31	20	18	--	14	16
Btu Output, Btu gas/ lb dry wood	--	4690	5490	4430	--	3980	4690
Carbon Recovery							
Gas Products, %	--	70	78	73	--	73	72
Liquid Products, %	--	22	23	24	--	16	20
Gas (Composition (dry)							
Vol %							
H ₂	6.3	7.0	30.6	37.2	10.0	21.3	25.9
CO	27.5	30.5	37.0	22.9	27.9	49.5	49.1
CH ₄	2.3	2.7	4.9	4.5	3.2	5.8	5.1
C _n H _m	0.2	0.2	0.3	0.2	0.2	0.4	0.3
O ₂	1.4	0.6	0.6	0.9	0.6	0.4	0.4
N ₂	55.5	53.0	0.4	0.7	50.9	0.1	0.1
CO ₂	6.7	6.0	26.2	33.6	7.1	22.5	19.1
Mole Ratio:							
H/C	0.622	0.651	1.197	1.520	0.875	0.861	0.996
O/C	1.107	1.072	1.300	1.466	1.089	1.201	1.180

Carbon balances are 100% accountable only for Test 3. Carbon accountabilities in Tests 2, 4, 6 and 7 are respectively 92%, 97%, 89%, and 92%. The carbon is more than likely lost by product gas leakage and should, therefore, be assigned to the gas products. Yield of carbon to liquid products increases slightly with the steam content in the oxygen/steam blast. The ratio of aqueous phase carbon to organic phase carbon varied from 3 to 4 and increased with steam content of the blast. Residence time in the reactor had no discernable influence on carbon yields to liquid products.

The carbon yields in the aqueous scrub solution were determined by sampling the scrub solution at the end of each test and analyzing for TOC. Total tars were determined at the end of Tests 4 and 7. Yields of wood carbon to the aqueous scrub solution and to tars are summarized in Table 3. The TOC of the scrub solution ranged from 50 to 86 g/. The tars contained some water, which was very difficult to remove, and averaged about 70.7% C, 7.8% H, 21.5% O, and 0% N with a heating value of 12,000 Btu/lb. The % of wood energy in the tars is 6.3 for Tests 2, 3, and 4 and 3.3 for Tests 6 and 7.

Energy yields in the gas phase normalized for 100% carbon accountability by adjustment of the product gas flow are given in Table 4. The maximum possible yields of methane and methanol from the oxygen/steam gasification tests are given in Table 5. The yields of methanol in Table 5 assume use of the methane in the product gas, which would require reforming of the methane.

TABLE 3. Carbon in Liquid Products

Test No.	Oxygen/Steam In Blast	% of Wood Carbon	
		In Scrub Solution	In Tars
2	2.3 (O ₂ in air)	15	6.5
3	1	17	6.5
4	0.5	18	6.5
6	4	13	3.4
7	2	17	3.4

TABLE 4. Normalized Gas Phase Energy Yields

Test No.	Oxygen/Steam, lb/lb	Btu/lb dry wood	Btu/Btu wood
3	1	5420	0.61
4	1/2	4610	0.52
6	4	4580	0.52
7	2	5210	0.59

TABLE 5. Methane and Methanol Yields from Oxygen/Steam Gasification of Wood in a Fixed Bed

Test No.	Oxygen/Steam, lb/lb	Methane Yield			Methanol Yield		
		scf/dry ton	MMB/dry MT	Btu/Btu Wood	gal/dry ton	lb/dry lb	Btu/Btu wood
3	1	8600	270	0.51	136	0.45	0.52
4	1/2	7300	230	0.43	115	0.38	0.44
6	4	7300	230	0.43	115	0.38	0.44
7	2	8300	260	0.49	130	0.43	0.50

Analysis of the hydrogen to carbon ratios in the product gases allows calculation of the fraction of steam in the blast that reacts to form CO and H₂. The following relationship is used to determine the H in the product gas from H₂O decomposition:

$$h_s = (R_s - R_o) A F X_c$$

where h_s is the lb moles per hour of H from steam decomposition, R_s is the ratio of H to C in the product gas with steam in the blast, R_o is the ratio of H to C in the product gas with no steam in the blast, A is the correction factor for 100% carbon recovery, F is the product gas flow rate in lb moles/hr, and X_c is the fraction of wood C in the product gas. From Test 1, R_o is 0.622. Results of the steam decomposition calculations are given in Table 6. Operation with above 70% steam decomposition is desirable.

Concentrations at different levels in the fixed bed were determined by sampling at the thermocouple locations (see Figure 1). Results for air/steam operation in Test 2 are given in Table 7. The percent energy contribution to the gases is determined by the gas heating value and nitrogen concentration,

TABLE 6. Steam Decomposition

Test No.	Oxygen/Steam, lb/lb	% H ₂ O Decomposition
3	1	74
4	1/2	45
6	4	93
7	2	88

TABLE 7. Concentration Profiles for Air/Steam Operation

Item	Sample Location					
	1 (top)	2	3	4	5	6 (Bottom)
Dry Gas Composition, Vol %						
H ₂	6.6	5.0	2.8	1.4	1.2	0.5
CO ₂	5.2	1.3	0.4	0.1	0.1	0.1
C ₂ H ₄	0.1	--	--	--	--	--
C ₂ H ₆	0.2	--	--	--	--	--
O ₂ , Ar	0.9	1.1	0.9	1	1	1
N ₂	52.9	59.8	63.1	63.6	64.1	64.4
CH ₄	3.3	1.0	0.1	--	--	--
CO	30.8	31.8	32.6	33.9	33.6	34.0
Vol % H ₂ O, wet basis						
	26.1	2.25	<2	<2	<2	<2
Heating Value, Btu/scf (dry)						
	160	118	113	113	112	111
% Energy Contribution to gases ^(a)						
	34.8	6.0	0.5	0.9	0.8	57

(a) Energy from this zone as a percent of total energy in gas.

since the total nitrogen flow is constant. It appears that about 40% of the energy in the gas is from the pyrolysis zone whereas 60% comes from gasification of the char residue in the bottom 16 in. of the gasifier.

The data in Table 7 indicate that very little hydrogen is generated in the gasification zone. The steam flow was supposed to be 5 lb/hr, which would give

only 7% steam decomposition; yet no moisture is in the gases from the gasification zone. Apparently, the expected steam flow was not attained in Test 2.

As expected, the entire yield of methane is from the top pyrolysis zone. Surprisingly, however, almost all of the CO₂ is also from the pyrolysis zone. This is not true for oxygen/steam operation as seen in Table 8.

TABLE 8. Concentration Profiles for Oxygen/Steam Gasification

Oxygen/Steam, lb/lb	Sample Position	Volume % Dry Basis									Btu/ scf dry
		H ₂	CO ₂	C ₂ H ₄	C ₂ H ₆	O ₂	N ₂	CH ₄	CO	H ₂ O ^(a)	
4 (Test 6)	1 (top)	17.32	16.3	0.2	0.27	0.41	16.3 ^(b)	5.4	43.8	53.46	258 (308) ^(c)
	2	18.66	16.15	0.25	0.27	0.3	9.58 ^(b)	5.35	48.93	26.59	279 (308) ^(c)
	3	21.60	15.07	0.12	0.12	0.1	0.4	4.32	58.27	24.01	304
	4	23.01	14.83	0.18	0.1	0.2	0.2	4.19	57.27	12.62	304
	5	21.45	14.47	0.1	0.01	0.2	0.2	2.20	61.37		290
	6 (bottom)	14.13	20.69	0	0	0.2	2.22	0.2	62.56	5.08	248
2 (Test 7)	1	26.04	16.45	0.11	0.19	--	1.10	5.84	50.26	58.40	309
	2	26.43	14.63	0.11	0.2	--	0.94	5.29	52.39	30.36	311
	3	30.02	13.21	--	--	--	0.45	4.00	52.33	8.53	304
	4	29.97	14.39	--	--	.15	0.40	2.90	52.19	10.77	293
	5	26.07	16.29	--	--	--	0.28	1.02	56.33	7.14	275
	6	24.13	20.77	--	--	0.1	3.57	1.45	49.95	15.01	252
1/2 (Test 4)	1	34.6	35.2	0.05	0.04	0.2	0.12	4.9	24.8	61.98	242
	2	39.9	33.6	--	--	0.1	0.1	3.5	22.8	13.11	236
	3	39.2	35.6	--	--	0.1	0.1	2.9	22.2	22.24	226
	4	40.4	33.6	--	--	0.1	0.1	2.5	23.3	24.58	230
	5	39.9	34.1	--	--	0.1	0.1	2.1	23.7	24.98	225
	6	35.5	39.1	--	--	--	--	1.1	24.3	20.17	203
1 (Test 3)	1	28.1	26.3	0.16	0.21	0.8	0.3	5.8	38.3	57.86	278
	2	31.9	25.6	0.02	0.04	0.2	0.6	4.0	37.6	10.77	264
	3	32.5	25.7	--	--	0.2	0.5	2.7	38.4	6.45	255
	4	31.9	26.2	--	--	--	0.5	2.3	39.1	7.61	251
	5	32.9	25.9	--	--	--	0.5	2.2	38.4	6.36	251
	6	28.4	39.8	--	--	0.2	0.5	1.2	29.9	4.10	199

(a) Wet basis.

(b) Apparent air inleakage to sample.

(c) Nitrogen-free value.

For oxygen/steam operation the CO_2 concentration is highest in the lowest zone for every test. The CO_2 concentration then decreases to the value in the adjacent zone and remains relatively constant in the other 5 zones. Apparently, all reaction between CO_2 and C occurs in the lower two zones only; this would also be expected from the temperature profiles shown in Figures 3 and 4.

Unlike the case for air/steam operation, we cannot determine the relative flow rates in the different zones from the concentration of a species (N_2) with a constant flow rate. Some observations can be made by analysis of the relative mole ratios of H, C and O in the dry product gas from each zone. These values along with a hypothetical water decomposition based on these values are presented in Table 9.

The moles of water per mole of O_2 are calculated from the H/O ratio in the dry product gas by the relation:

$$\frac{\text{H}_2\text{O}}{\text{O}_2} = \frac{2 (\text{H/O})}{2 - (\text{H/O})}$$

The water decomposition is then calculated as 100 times this calculated ratio divided by the mole ratio of water-to-oxygen in the blast (32 divided by 18 times the oxygen-to-steam weight ratio). The calculation of water decomposition is valid only if carbon alone is being gasified. This condition is probably met only in the lower zones in the gasifier as the data confirm. In the lower zones, the % water decomposition agrees well with the values in Table 6 calculated from overall yields and shown in parentheses in Table 9.

The contribution of hydrogen to the dry product gas from the pyrolysis zone is seen much more strongly in Tests 6 and 7 with low steam content in the blast than in Tests 3 and 4 with high steam contents in the blast. It is apparent from the data in Tables 7, 8 and 9 that gasification reactions occur only in the extreme bottom two zones while pyrolysis occurs throughout the remaining zones except for air/steam operation where most of the pyrolysis occurs in the top two zones.

TABLE 9. Analysis of Yields from Gasifier Zones

Oxygen/Steam, lb/lb	Sample Position	Molar Ratios in Dry Gas				% H ₂ O Decomposition
		C/H	C/O	H/O	H ₂ O/O ₂	
4 (Test 6)	1 (top)	1.13	0.86	0.76	1.23	277
	2	1.17	0.87	0.74	1.16	263
	3	1.27	0.88	0.69	1.05	236
	4	1.20	0.88	0.73	1.15	259
	5	1.50	0.86	0.57	0.80	180
	6 (bottom)	2.87	0.80	0.28	0.33	74 (93) ^(a)
2 (Test 7)	1	0.95	0.88	0.93	01.74	196
	2	0.96	0.89	0.93	1.74	196
	3	0.91	0.88	0.97	1.88	211
	4	0.97	0.85	0.88	1.57	176
	5	1.31	0.83	0.63	0.92	103
	6	1.33	0.79	0.59	0.84	94 (88) ^(a)
1/2 (Test 4)	1	0.73	0.68	0.93	1.74	49
	2	0.64	0.66	1.03	2.12	60
	3	0.67	0.65	0.97	1.88	53
	4	0.65	0.65	1	2.00	56
	5	0.68	0.65	0.96	1.85	52
	6	0.84	0.63	0.75	1.20	34 (45) ^(a)
1 (Test 3)	1	0.88	0.77	0.88	1.56	88
	2	0.84	0.76	0.90	1.65	93
	3	0.88	0.74	0.84	1.45	82
	4	0.93	0.74	0.80	1.32	74
	5	0.89	0.74	0.83	1.42	80
	6	1.15	0.64	0.56	0.77	43 (74) ^(a)

(a) Values from Table 6 calculated from overall yields.

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