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ENGINEERING ANALYSIS OF BIOMASS GASIFIER PRODUCT
GAS CLEANING TECHNOLOGY

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ABSTRACT

For biomass gasification to make a significant contribution to the energy picture in the next decade, emphasis must be placed on the generation of clean, pollutant-free gas products. This report attempts to quantify levels of particulates, tars, oils, and various other pollutants generated by biomass gasifiers of all types. End uses for biomass gases and appropriate gas cleaning technologies are examined. Complete systems analysis is used to predict the performance of various gasifier/gas cleanup/end use combinations. Further research needs are identified.

SUMMARY

An interpretive literature search was done to determine particulate and tar emissions from various biomass gasifiers, identify particulate and tar limits for specific end uses for the gas, and evaluate different gas cleaning options which meet the specifications for different end uses.

The literature search revealed a profound lack of quantitative information in this area. In particular there is very little information on the properties of tars and particulates from biomass gasification and there is almost no documentation on the effectiveness of gas cleaning systems for biomass derived gas, particularly for tar removal.

In light of the lack of data available on gas cleaning with biomass gasification systems the results reported here are based heavily on information from other systems, primarily coal gasification and biomass combustion. Numerous citations on particle removal, but only limited information on tar removal were found. As a result the report addresses particulate removal in more detail. Tars are, in most instances, ultimately removed as liquid droplets so much of the data on particulate removal is applicable to tar removal. However, the properties of tars such as their viscosity and stickiness must be considered.

A variety of units were encountered during the course of this study. We attempted to keep the units consistent for each subject area but to do so for the entire report we felt was not appropriate. Appendix A is a nomograph that can be used to convert different sets of units used in the text.

Gasifiers that have been used with biomass are fixed bed updraft, fixed bed downdraft, fluid bed, and entrained bed units. Fixed bed updraft units typically produce large quantities of tars and oils ($10\text{--}100\text{ g/m}^3$). Fixed bed

downdraft and fluid bed units generally have lower tar emissions in the range of 0.05-0.5 g/m³.

Particulate emissions vary from low particulate loadings (0.1-1.0 g/m³) for an updraft gasifier to 10-100 g/m³ or greater for a fluid or entrained bed unit.

The type of gasifier also influences the physical characteristics of both tars and particulates. High boiling, condensed aromatic tars are generated from fluid beds and downdraft units. Lower boiling, highly oxygenated wood oils are generated from updraft and entrained flow pyrolysis units. Fixed bed gasifiers emit small quantities of very fine particulates (mostly ash) entrained in the gas stream. Fluid beds on the other hand release large amounts of very coarse particulates which are primarily char and ash. Data on particulate and tar production rates and characteristics is limited. Further study which addresses quantitative pollutant levels for each type of gasifier is needed.

End uses for biomass gases include use as a fuel for industrial process boilers, dryers or kilns; as a fuel for diesel and spark ignition engines and gas turbines; and as a synthesis gas for methanol, methane, hydrocarbons and ammonia. Each end use has specific requirements for the cleanliness of the gas. For example, particulates in burner flue gases must meet environmental standards which vary depending on the size and location of the facility. Many studies have been done with internal combustion engines. Particulates or tars above 0.05 g/m³ are shown to cause excessive engine wear or gum formation on the valves. Additional research is needed to define limits for gas turbines and synthesis gas applications.

Gas cleanup technologies that are most applicable for biomass gasification are cyclones, wet scrubbers, various filters (including baghouses) and

electrostatic precipitators. Data on gas cleaning from operating biomass gasifiers has not been extensively reported.

Based on data for coal systems and limited biomass data, particulate removal systems appear adequate for most biomass gasifier applications. However, tar removal presents problems which heretofore have not been studied in any detail. Further research is needed to confirm the applicability and efficiency of various gas cleanup methods coupled with different gasifier types.

Table 1 summarizes the results of this study. Areas where available information is adequate for selection and design of gas treatment systems are identified. Areas where sufficient information is not available and additional research and development are necessary are also listed in Table 1. This should serve as a basis for further efforts in gas cleaning for biomass gasification.

TABLE 1. Summary of Gas Cleaning Technology Research Needs for Biomass Gasifiers

Areas Where Available Information is Adequate for Selection and Design of Gas Treatment Systems

- Particulate production rates
- Particulate and tar limits for internal combustion engines
- Particulate limits for direct-fired equipment (boilers, dryers, kilns, etc.)
- Tar composition

Areas Where Additional Research and Development are Necessary

- Tar production rates for all types of gasifiers -- effects of operating conditions, type of biomass, moisture content, etc.
- Composition and size distribution of particulates
- Particulate and tar limits for gas turbines and synthesis gas applications
- Efficiency of tar removal devices
- Effect of conditions (temperature, pressure, tar-loading) on tar deposition in transfer pipes, burners, and other downstream equipment
- Volatility of ash components

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INTRODUCTION

Biomass gasification has the potential to make a significant contribution to the future energy supply in the United States and is already finding commercial applications primarily in the forest products industry. One area that has not been studied in detail is gas cleaning, primarily for particulate and tar removal. Particulates can cause plugging and erosion of downstream equipment and may also be an environmental problem. Plugging of downstream equipment is the primary problem associated with tars. Once they are separated from the gas, tars may be an environmental hazard and present a disposal problem.

The objective of this study was to evaluate gas cleaning technology, primarily for particulate and tar removal, as it applies to biomass gasification, and identify potential problem areas and gaps in the technology that could impede development and utilization of biomass gasification.

Selection of gas cleanup equipment for biomass gasification depends primarily on two factors: 1) the type of gasifier used and 2) the intended end use of the gas. The type of gasifier, and to some extent, the feedstock, will determine the concentration of particulates and tars in the gas. The end use will define the particulate and tar concentrations which can be tolerated. This relationship is shown in Figure 1.

Phase I of this study reviewed the state-of-the art of gas cleaning technology as it applies to biomass gasification. The results of Phase I which are presented here have been divided into three chapters:

- Chapter 1 Biomass Gasification Gas Contaminants
- Chapter 2 End Uses for Biomass Gas
- Chapter 3 Gas Cleanup Technology

FIGURE 1. Gas Cleanup Technology for Biomass Gasification

Phase I was based on an extensive literature survey. This survey revealed a profound lack on data on gas cleaning systems for biomass gasification, particularly for tar removal. Information on gas cleaning systems for coal gasification and biomass combustion were also studied. This provided a sizeable base of information on particulate removal, but still little on tar removal. Because of this the results of Phase I focus most heavily on particulate removal.

Specific gas cleaning methods were applied to various gasifiers in Phase II. The primary objective was to identify gas cleaning methods which could meet specifications for the projected end uses of biomass gas. Gaps in the technology were identified and recommendations for additional research were made. This material is included in Chapter 4, Evaluations and Recommendations.

1.0 BIOMASS GASIFICATION GAS CONTAMINANTS

When carbonaceous materials are gasified or combusted, char, ash, tar, and gases are produced. The relative fractions of these four products will vary and depend primarily on the type of gasifier, gasifier operating conditions, and the feedstock.

Char/ash particles and higher hydrocarbon vapors or droplets are considered contaminants in a gas stream. Gasification processes either seek to minimize char and tar production or to maximize their removal from the gas stream. Other contaminants or pollutants which may be present in biomass gas include olefins, hydrogen sulfide, sulfur dioxide, carbonyl sulfide, nitrogen oxides, and trace metals. This chapter identifies the concentrations and properties of contaminants produced in various types of biomass gasifiers with emphasis primarily on particulates and tars.

In the multitude of references cited in this report, researchers have given pollutant concentrations in a wide variety of units. We have attempted to leave the citation in its original form to avoid any interpretational errors. For the readers assistance Appendix A is a nomograph for conversion of contaminant loadings from one set of units to another.

1.1 GASIFIER TYPES

Over the years many different types of gasifiers have been constructed for experimental and commercial use. However, most fall into three distinct categories. The three main types are: fixed bed, fluid bed, and entrained flow, referring to the motion of the solids in the gasifier. Each type of gasifier has different characteristics which result in significantly different yields of particulates and tars.

1.1.1 Fixed Bed Updraft

"Fixed bed" is a broad term describing a gasifier in which the fuel is fed onto a relatively stationary inventory of biomass already present in the reactor. The fixed bed category encompasses updraft, downdraft, and cross-draft units. Each type has different operating characteristics and emissions. The term "fixed" refers to the condition at the extremes of the bed, which do not vary at steady state. Most updraft biomass gasifiers are operated at atmospheric pressure but commercial coal gasifiers operate up to 30 atm.

Updraft units (Figure 2) exhibit a countercurrent flow of fuel and gas. Solid fuel is fed from the top by lock hoppers or feeders. The bed of fuel is supported by a grate at the bottom of the reactor. The fuel flows down through the drying zone, pyrolysis zone, reduction zone, and combustion zone. Ash and unreacted fuel (char) exit through the grate at the bottom. Reactant gases (air, oxygen, steam) are introduced into the reactor through the grate. The hot gases from the combustion zone provide energy for the endothermic processes in the upper zones and exit at the top of the gasifier saturated with pyrolysis oils and water. Exit temperatures range from 50-150°C. Approximately 20-25 percent of the carbon in wood is recovered as liquid products. The condensed liquids usually are in two phases: an aqueous phase containing highly oxygenated water soluble organics (pyroligneous acids) and a separate tar phase (Mudge, et al., 1980). Because of the low velocity of the gases in the reactor and the filtering effect of the bed, the product gas contains little particulate matter. Detailed descriptions of fixed-bed, updraft biomass gasifiers are given by Baker (1984), Fritz (1978), Miller (1983), Mudge and Rohrmann (1978), Reed (1979), and Oliver (1982).

FIGURE 2. Schematic of a Fixed Bed Updraft Gasifier

1.1.2 Fixed Bed Downdraft

Production of pyrolysis oils is largely eliminated in downdraft gasifiers (Figure 3). As in updraft units solid fuel is fed from the top. However air, which is used in most downdraft units, is introduced concurrently into the combustion zone through a distributor as shown in Figure 3. Pyrolysis oils and moisture from pyrolysis and drying are drawn down through the high temperature reduction and combustion zones where they undergo thermal cracking. The product gases exit near the bottom of the reactor at 300 to 550°C. Ash and char leave through a grate at the bottom of the reactor. As with updrafts, most operate at atmospheric pressure.

FIGURE 3. Schematic of a Fixed Bed Downdraft Gasifier

Low gas velocities result in low particulate loadings in the same range as updraft. Downdraft gasifiers exhibit very low tar yields which are dependent on the combustion zone temperature.

A uniform combustion area is critical for proper operation of this type of gasifier and scale up of the air distribution system is difficult. As a result downdraft gasifiers are usually small in diameter compared to the other types of gasifiers.

Kaupp and Goss (1981) provide a detailed treatise of small downdraft biomass gasifiers. Additional information can be obtained from Fritz (1978), Groeneveld (1983), Hodam (1978), and Reed (1983).

1.1.3 Fixed Bed Crossdraft

Crossdraft gasifiers (Figure 4) exhibit many of the operating characteristics of downdraft units. Tars and particulates are both quite low and gasifier heat efficiency is higher than a downdraft.

Air or air/steam mixtures are introduced in the side of the gasifier near the bottom. Producer gases are drawn off the opposite side at temperatures near those of downdraft units (300-550°C). Tars and oils are drawn through the reduction zone and cracked to lighter components. Some particulates (mostly ash) are entrained in the product gases. More detailed information on crossdraft gasifiers can be obtained from Kaupp and Goss (1981) and Miller (1983).

FIGURE 4. Schematic of a Crossdraft Gasifier

1.1.4 Fluid Bed

In a fluid bed gasifier (Figure 5) the incoming and evolved gases maintain the reactor bed in a turbulent fluid-like state much like a boiling liquid. The result is an expanded reactor bed of char particles and, in most biomass gasifiers, an inert solid such as sand. Because biomass is less dense and has less ash and fixed carbon than coal the inert solid is used to maintain proper fluidization (prevent bridging and channeling) and to provide additional heat capacity in the bed.

No distinct zones exist in a fluid bed gasifier as near isothermal operation is maintained with good fluidization. The product gas contains some tars and oils depending on the bed temperature (600-900°C), and does have a fairly large loading of particulates (ash and char). Operating pressures to

FIGURE 5. Schematic of a Fluidized Bed Gasifier

20 atm are common in experimental units. Depending on the design, ash and char may be removed from the top of the reactor with the product gases, from the bottom of the reactor, from the top of the bed, or a combination of the three. Datin (1981), Flanigan (1983), Feldman (1983), Goldbach (1983), Miller (1983), Mudge (1983), Murphy (1984), and Oliver (1982) provide more details on design and operation of fluid-bed, biomass gasifiers.

1.1.5 Entrained Flow

In an entrained flow (or transport) gasifier (Figure 6), finely sized fuel particles are entrained in the feed gas (usually oxygen and steam) prior to entry into the reactor. Gasification takes place with the feed particles suspended in the gas phase. The product gas, ash and char leave the top of the reactor. Limited data indicate particulate and tar loadings similar to

FIGURE 6. Schematic of an Entrained Bed Gasifier

or greater than those from fluidized bed units. Most entrained flow units are low temperature short residence-time pyrolysis units which operate about 500°C at atmospheric pressure. A cyclonic, entrained-flow, air blown gasifier feeding sawdust operated at 800-1,000°C (Cousins and Robinson 1985).

1.2 TARS AND OTHER CONDENSIBLE ORGANICS

Tar is a generic term for the higher boiling (>150°C) constituents of biomass gas which are formed during the pyrolysis reactions. Depending on the degree of cracking, the tar may range from light, oxygenated hydrocarbons to heavy, polyaromatic hydrocarbons (PAH).

1.2.1 Production Rates and Concentrations

Each type of gasifier has different reaction conditions and consequently different tar compositions and production rates. An updraft fixed bed gasifier generates a high yield of tar (~ 20 wt% of the carbon in wood feed is converted to liquids) due to the low temperature at which drying and pyrolysis take place in the gasifier. The drying and pyrolysis zones in an updraft gasifier will range from 80 to 200°C. At these temperatures the tars are, for the most part, condensed droplets entrained in the gas phase. Reported tar yields range from 10-100 g/m³ of gas (Kaupp and Goss 1981; Baker 1984; Fritz 1978; Dravo Corp. 1976). Downdraft gasifiers pass the pyrolysis gases through the combustion zone which thermally cracks and oxidizes much of the tar at temperatures near 1200-1600°C. Reported tar yields from downdraft units range from 50-500 mg/m³ (Kaupp and Goss 1981; Groenevald 1983; Reed 1983; Kumar 1984).

Fluidized (or entrained) beds when operating properly are nearly isothermal. Biomass feed is pyrolyzed either submerged in the bubbling bed or entrained in a hot gas stream. High temperature (600-900°C) and good gas-

solid contacting results in some cracking of pyrolysis oils. The final concentration of tars is a function of the reaction temperature, pressure, and residence time in the bed and is typically in the range of 2-10 g/m³. The product gases from downdraft, fluidized bed, and entrained bed gasifiers are hot and any tars that are still present will be mostly in the vapor phase. Typical tar production rates for each type of gasifier are summarized in Figure 7.

1.2.2 Properties of Biomass Tars

A recently completed study of biomass gasification/pyrolysis condensates at PNL (Elliott 1985) concludes that there is no typical tar composition which can be adequately used to represent all thermally produced biomass tars. The tar composition, as well as the amount, is dependent on the operating conditions, principally a time/temperature thermal severity-type function. The properties of the tar therefore appear to vary on a continuum from "primary" oxygenated pyrolysis tar collected after a short residence

FIGURE 7. Typical Tar Concentrations from Various Gasifiers

time at low temperatures of around 500°C to highly aromatic, deoxygenated tar which is produced at short residence time at high temperatures of around 900°C. This continuum of tar cracking/condensation appears to argue against any purely pyrolytic mechanisms for complete conversion of biomass to a char-free and tar-free gas. The intermediate products of such a progression would be some grade of tar with the end product being a finite amount of coke or graphite.

This study suggests that a spectrum of tar condensates can be recovered from entrained-flow or fluidized-bed reactors processing wood over a temperature range from 450°-950°C. In these short residence time reactors the extent of progression along a path of thermal decomposition is dependent on the upper temperature limit achieved in the reactor. These reactions are very fast and require only short residence times on the order of fractions of seconds. Whether there is a smooth continuum or a sharp demarcation between the oxygenated and deoxygenated products at some intermediate temperature around 650°C is, as yet, unknown.

The location of the organic condensates from the fixed bed gasifiers within this tar property continuum is most affected by specific process configurations. A wide range of condensate properties have been measured. In theory the organic vapors from the downdraft gasifier should be completely combusted; practically speaking such complete combustion is not obtained. The extent and type of organic contamination in the downdraft condensate is mainly a function of the efficiency of the combustion zone and the extent of channeling in the bed.

In the case of updraft operation the condensates appear to be very similar to the well-known pyrolysis condensate which is recovered from batch carbonization of wood. The condensate has two phases with a heavy organic

phase containing the typical components found in "settled tar" from charcoal manufacture. The lighter aqueous phase is highly acidic and carries a large amount (20 to 25 percent by weight) of dissolved organic material much like "pyroligneous acid."

The chemical compositional changes in the tars suggest a general process of deoxygenation and dehydrogenation. These changes are noted by comparison of elemental analyses and volatile component identification as well as several forms of spectrometry. A general pathway of tar chemical functional degradation is represented below. This pathway is meant to represent the nature of the tar composition as a function of thermal processing and can be viewed as a spectrum of increasing temperature from low temperature (450°C) on the left to high temperature (950°C) on the right.

Table 2 summarizes the properties of tars from different gasifiers as analyzed by Elliott (1985). Wood was the feedstock for all of these gasifiers. The properties of tars from fluidized bed or entrained flow gasifiers varied widely depending on the temperature of operation. The properties of downdraft tars were also varying indicating the mode of operation has a significant effect on tars from these units. The analysis of the fixed-bed tar from the Rome, Georgia gasifier is similar to a previous analysis of fixed-bed tar from PNL's fixed bed gasifier (Baker 1984).

TABLE 2. Properties of Biomass Tars

	Fluidized Bed/ Entrained Flow		Updraft		Downdraft	
	480°C	880°C	tar	aqueous	tar	aqueous
Carbon, %	52.7*	84.0*	70.9*	11.3	67.2*-0**	7.0-0.5
Hydrogen, %	6.2	5.7	7.2	--	5.8-0	--
Oxygen, %	40.5	8.7	21.7	--	25.0-0	--
Ash, %	0.6	1.6	0.2	0.1	2.0-0	1.4-0.02
pH	--	--	--	2.1	--	3.0-5.3
viscosity, cps	220-1300 @40°C	9800-26,800 @51°C	410 @40°C	--	7500-41,000 @78°C	--
moisture, wt%	16	20-28	8	--	12-13	--
density, g/ml	1.26-1.28	1.14-1.16	1.13	--	1.16	--

* elemental and ash analyses for tars are reported on a dry basis

** downdraft gasifiers may or may not produce a separate tar phase depending on the specific operation

Table 3 shows some of the major organic components of the two general types of biomass tars, oxygenated and deoxygenated. Table 2 also gives typical trace metal analysis. Iron and silica are the most abundant followed by calcium, potassium, and zinc.

TABLE 3. Components of Biomass Tars (Elliott 1985)

Major Organic Components	
Low Temperature Oxygenated Tars	High Temperature Deoxygenated Tars
phenol	acenaphthalene
methylphenols	naphthalene
cresols	methylnaphthalenes
methoxyphenols	fluorene
dimethoxyphenols	phenanthrene
levoglucosan	fluoranthrene
	acephenanthrylene
	benzanthracenes

Trace Metals (ppm)

Fe	200 - 7850
Si	<250 - 4670
Ca	<20 - 3100
K	<30 - 1060
Zn	0 - 1950

1.3 PARTICULATES

The concentration, size, and composition of particulates in gas from biomass gasifiers vary considerably. The variations are primarily a function of the type of gasifier and the feedstock.

1.3.1 Production Rates and Concentrations

The concentration of particulate matter in the product gas from a biomass gasifier depends on numerous factors including the type of gasifier, feedstock, and gasifier operating conditions. Operating conditions that influence particulate emissions include gas velocity in the bed, temperature, moisture content of the blast, and the rate of gasification. The concentration of particulates from various gasifiers is summarized in Figure 8.

FIGURE 8. Typical Particulate Concentrations from Various Gasifiers

Fixed bed gasifiers, both updraft and downdraft, employ large (2.5-5 cm, 1-2 in) chunks of feedstock that are not entrained by the gas flow; nevertheless, the product gas does contain a significant quantity of particulates, 100-1000 mg/m³ (Finnie 1979, Hoenig and Cole 1981, Kaupp and Goss 1981, Katz 1983, Jacko 1983, Richey 1985, Levelton 1980). The lower values were typically measured after the gas had been combusted in a burner. The higher values are measured directly from the gasifier. The values for downdraft gasifiers are somewhat higher than updraft (Kaupp and Goss 1981) possibly because the higher temperature of the product gas results in higher gas velocities exiting the gasifier.

Gas velocities in fluid-bed and entrained-flow gasifiers are sufficiently high to entrain considerable particulates from the fine feedstock to these gasifiers. Typical particulate concentrations for fluid bed gasifiers range from 10-456 g/m³ (Datin 1981, Datin 1983, Murphy 1984, Moreno and Goss 1983, Oliver 1982, Mudge 1983). The higher values are for high ash feedstocks (cotton gin trash and cattle manure) or gasifiers where conversion is less than 100% and char is a significant byproduct. For air blown gasifiers with a low ash wood feedstock the particulate concentration ranged from 10-30 g/m³.

1.3.2 Particle Size

The assumed mechanism for particle elution from a gasifier is entrainment in the gas stream. The size of particulates carried out with the product gas will depend primarily on gas velocity exiting the gasifier. Only a limited amount of data on the particle size distribution from biomass gasifiers has been published as shown in Table 4. A particle size distribution for a fixed bed gasifier could not be found. Jacko (1984) tested a fixed-bed gasifier and indicated the particle diameters were small similar to the downdraft data

TABLE 4. Particle Size Distribution From Various Biomass Gasifiers

<u>Particle Size Microns</u>	<u>Downdraft (Jacko 1985)</u>	<u>Downdraft (SERI 1979)</u>	<u>Fluid Bed (Datin 1981)</u>
250		74	
100		50	
50		30	99
30		18	97
20	99+	11	87
10	97		38
5	96		17
2	95		2
1	89		
0.5	81		

reported in Table 3. The particulates from the downdraft unit were quite small (89% less than 1 micron). It is surprising to see the large particle size distribution reported for Imbert downdraft vehicular gasifiers. This is probably due to the hot gas exiting through a small annular space which results in a high gas velocity. Other downdraft units (Hoenig and Cole, 1981) show more than 99% of the particles being less than 3 microns. Particulates from a fluid bed were larger than those from the fixed bed units, primarily between 5 and 30 microns.

1.3.3 Composition

Particulates from the different gasifier types are comprised of the inorganic constituents in the feedstock, any unconverted feedstock, and dirt. In many systems the particulates in the gas stream are collected as a byproduct fuel, generally referred to as char because they contain a large percentage of carbon. Char is often a byproduct from fluid-bed and entrained-flow gasifiers. Carbon content of solids from fixed-bed gasifiers is very low.

Published data on the composition of wood and agricultural residue ash (Table 5) show considerable variation in concentrations of major constituents

(Ca, K, Si, Na, and Al). The usual main component in ash is CaO, which accounts for 10 to 60 wt% of the ash and is typically present at about 40 wt%. Concentration of K₂O can be 10 wt% or greater. Ash from agricultural residues consists primarily of silica and alkali. Feedstocks which have a large percentage of dirt or other inorganics can raise the silica content of any ash considerably.

Particulate composition from the different gasifier types is not reported. With fluid and entrained bed gasifiers entrainment of solid particles from the bed is the likely mechanism for particle elution from a gasifier. Particulate entrainment, therefore, increases with increased energy output, and the inorganic content of the feedstock.

In a fixed bed gasifier alkali and possibly other more volatile components of the ash could be volatilized in the combustion zone of the gasifier, condensed or solidified into small particles in the cool zones, and entrained in the gas stream. Analysis of entrained particles would determine if such a mechanism does occur. Corrosion products from the gasifier and outlet piping may also be present in the product gas as particulates.

TABLE 5. Typical Ash Compositions for Biomass Feedstocks
(Kaupp and Goss 1981)

<u>wt %</u>	<u>Wood</u>	<u>Agricultural Residues</u> <u>(wheat straw, corn stover, rice straw)</u>
Al ₂ O ₃	1-10	0-2
CaO	10-60	2-14
Fe ₂ O ₃	0.5 - 4	0-2
K ₂ O	2-41	10-26
MgO	1-17	2-3
Na ₂ O	1-20	2-13
SiO ₂	0-2	18-78

1.4 OTHER CONTAMINANTS

1.4.1 Sulfur Compounds

Biomass feedstocks in general have a very low sulfur content. Table 6 indicates typical sulfur levels in some biomass materials. The fate of the sulfur in gasification depends on the temperature and the amount of water present. Low steam rates and temperatures favor the formation of COS and CS₂. At steam fractions greater than 20 mole % nearly all of the sulfur compounds are converted to H₂S. Higher temperatures also favor H₂S formation.

TABLE 6. Sulfur Content of Biomass Fuels*

<u>Biomass</u>	<u>% sulfur, dry weight basis</u>
Alfalfa Seed Straw	0.3
Almond Shells	less than 0.02
Bagasse	0.03-0.12
Barley Straw	0.14
Coffee Hulls	0.2
Corn Cobs	0.001-0.007
Corn Fodder	0.15
Corn Stalks	0.05
Oat Straw	0.23
Cotton Gin Trash	0.26-0.31
Flax Straw, Pelleted	less than 0.01
Furfural Residue	0.4
Olive Pits	0.02
Manure	0.4-0.6
Peach Pits	0.04
Peanut Husk	0.1
Peat (Finnish)	0.05-0.2
Peat, General	1.5-2.0
Rice Hulls	0.16
Rice Straw	0.10
Walnut Shells	0.03-.09
Wheat Straw	0.17
Wood, Chipped	0.08
Wood, General	0.02
Wood, Pine Bark	0.1
Wood, Green Fir	0.06
Wood, Kiln Dried	1.0
Wood, Air Dried	0.08

*Adapted from Kaupp and Goss (1981)

Sulfur concentrations in gas streams are generally low. As a comparison, crossdraft gasifiers fueled with anthracite (0.5% S) typically run about 1 g S/m^3 (Kaupp and Goss, 1981). Baker and Brown (1984) measured 80-240 ppm H₂S ($0.2\text{-}0.5 \text{ g s/m}^3$) in the gas from a continuous laboratory gasifier with bagasse as the feedstock and 20-30 ppm with wood. No sulfur was detected by Hodam & Williams (1978) from their gasifier/boiler combination. Murphy (1982) measured 4-6 ppm SO_x ($\sim 0.01 \text{ g s/m}^3$) from a fluid bed gasifier/boiler combination. Both of these units were wood fueled.

1.4.2 Nitrogen Compounds

Fixed nitrogen in biomass feedstocks (Table 7) is converted during gasification to gaseous nitrogen compounds, primarily ammonia (NH₃) and hydrogen cyanide (HCN). Most researchers agree that nitrogen in the fuel is the main contributor to these compounds and that little or no gaseous N₂ is converted to NH₃ or HCN.

TABLE 7. Nitrogen Content of Biomass Fuels*

<u>Biomass Fuels</u>	<u>% nitrogen dry weight basis</u>
Bagasse	0.2-0.3
Barley, Straw	0.59
Corn Cobs	0.16-0.56
Corn Fodder	0.94
Cotton Gin Trash	1.34-2.09
Corn, Stalks	1.28
Flax Straw, Pelleted	1.1
Manure	2.5-3.1
Oat Straw	0.66
Olive Pits	0.36
Peach Pits	1.74
Peat	0.5-3.0
Prune Pits	0.32
Rice Hulls, Pelleted	0.57
Safflower, Straw	0.62
Walnut Shells	0.26-0.4
Wood (General)	0.009-2.0

* Adapted from Kaupp and Goss (1981)

After gasification the specific use of the gas will also create additional nitrogen compounds. Any NH_3 or HCN will be converted to NO_x by high temperature burners, boilers, or combustion turbines. If the gasifier is air blown then some elemental nitrogen (N_2) may also be converted to NO_x in the combustion zone or in high temperature downstream equipment.

Airborne NO_x releases from biomass facilities have been reported in the range of 100 ppm. Hodam & Williams (1978) refer to a downdraft gasifier/boiler combination as having 130 ppm NO_x emissions. From a cross-flow gasifier/burner $0.15 \text{ lb}/10^6 \text{ Btu}$ ($\sim 300 \text{ ppm}$) can be expected (Miller 1983). Fluid bed units coupled with boilers may show lower NO_x levels. Murphy (1984) found 58 ppm NO_x from a 54 MM Btu fluid bed gasifier/boiler combination. Battelle-Columbus uses an air-blown char combustor to supply heat to a circulating bed of solid material. They (Feldman et al. 1983) found $0.25 \text{ lb NO}_x/10^6 \text{ Btu}$ ($\sim 500 \text{ ppm}$) was emitted from this combustor.

All of the available data suggests that NO_x emissions from biomass facilities will be at or below those required for New Source Performance Standards (NSPS), $0.6 \text{ lb}/10^6 \text{ Btu}$ ($\sim 1200 \text{ ppm}$).

1.4.3 Olefins

Most gasification or pyrolysis reactions generate some olefinic hydrocarbons. Ethylene and propylene are the most prevalent. When the gas is used as a fuel olefins contribute to the heating value of the gas; however, if the gas is to be used as a synthesis gas, olefins may be catalyst poisons. They polymerize on the catalysts' surfaces and plug the pores. This restricts the number of active sites available for the desired reactions causing decreased or significantly altered product yields.

Olefin production depends on the type of gasifier, the temperature and pressure, the gas residence time, the amount of H_2O present, and/or the

presence of a catalyst. Typical olefin concentrations are tabulated below in Table 8. The highest concentrations are found in short-residence time pyrolysis reactors.

TABLE 8. Olefin Production in Biomass Gasifiers

	<u>Fixed Bed Updraft</u>	<u>Fixed Bed Downdraft</u>	<u>Fluid Bed</u>		<u>Entrained Flow</u>
			<u>non-catal.</u>	<u>catalytic</u>	
C ₂ H ₄	1-2.4%	0.2-0.4%	0.8-2.9%	0%	1-14%
C ₃ H ₆ ⁺			0.9-2.8%	0%	
Reference	Overend (1979)	Johannson (1979)	Flanigan, et al. (1983)	Mudge et al. (1983)	Diebold & Scahill, (1983)

1.4.4 Condensate

Cooling and scrubbing the raw gas from biomass gasification produces a condensate or wastewater stream. Elliott (1985) analyzed condensate samples from nine experimental gasifiers. The pH of the water ranged from 2.1 for a fixed-bed updraft to 3.0-5.3 for downdrafts, and 5.9-8.7 for fluid bed gasifiers. The total organic carbon dissolved in the water was very high for the fixed bed updraft (110,000 ppm), somewhat lower for downdrafts (5,000-71,000 ppm) and quite low for fluidized beds (0-400 ppm).

1.5 PRESSURE EFFECTS

The data on emissions presented in this chapter are based primarily on gasifier operation at atmospheric pressure. Advanced gasifiers may operate at elevated pressures. This may have a significant effect on emissions.

Calculations show much higher methane and higher hydrocarbons concentrations at equilibrium for higher pressures (Mudge 1983). This might lead to higher tar concentrations as well, although this has not been substantiated.

Particulates concentrations from fluid beds appear to be greater for higher pressures. Zenz and Othmer (1960) make reference to work by May and Russell in Gohr (1956) which examined entrainment as a function of pressure and gas velocity with cracking catalysts. They found for a given gas velocity, entrainment at 200 psig could be over 10 times that at atmospheric pressure. Density and viscosity of entraining gases plays an important role in entrainment. Increasing the pressure increases both the density and viscosity of gases and the resulting drag forces on the particle. Little is known about fixed bed particulates vs. pressure. Most fixed bed gasifiers are operated near atmospheric pressure.

Thermodynamics and reaction equilibria (partial pressures) will also effect the particular species and relative concentrations of sulfur compounds, nitrogen compounds, and olefins, but experimental data involving biomass does not appear to exist.

1.6 SUMMARY AND CONCLUSIONS

Fixed-bed updraft units exhibit the highest tar and oil emissions but the lowest particulate emissions, while fluid beds are highest in particulate emissions but lower in tar and oil loadings. Table 9 summarizes available information on contaminants from biomass gasification systems.

Particulate characteristics are not well known. Various gasifiers exhibit different particle size distributions and compositions. Particulates can be mostly char in fluid beds, but mostly ash in fixed bed units. Wood

ash has a relatively large amount of potassium and sodium which may volatilize into the gas stream. Further work is needed to evaluate alkali transport phenomenon.

TABLE 9. Summary of Emissions from Biomass Gasifiers

	<u>Fixed Bed Updraft</u>	<u>Fixed Bed Downdraft</u>	<u>Fixed Bed Crossdraft</u>	<u>Fluid Bed</u>	<u>Entrained Bed</u>
Product Gas Temperature, °C	50-150	300-550	300-550	600-900	800-1,000
Pressure, atm	1	1	1	1-20	1
Particulates					
Loading, g/m ³	0.1-1.0	0.1-10	--	10-500	--
Particle size Distribution, microns	--	< 2	--	1-50	--
Tar Loading, g/m ³	10-100	.05-0.5	--	2-10	--
SO _x ^(a) , ppm	--	0	--	4-8	--
NO _x ^(a) , ppm	--	130	300	58	--
Condensate (wastewater)					
pH	2.1	3.0-5.3	--	5.9-8.7	--
TOC, ppm	110,000	5,000-71,000	--	0-400	--

a) based on combustion of the raw gases

Tars and oils produced during biomass gasification are also not well characterized. Of particular interest is the dependence of chemical constituents on the reaction conditions. Other areas of interest include tar deposition in transport lines, catalytic destruction of tars, and possible recovery/recycling methods.

Not enough is known about the basic gasification processes to predict pollutant concentrations reliably. Before gas cleanup systems can be adequately designed the anticipated loadings must be known as accurately as possible.

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2.0 END USES FOR BIOMASS GAS

The extent of gas cleanup required will depend on the intended end use of the gas. Low-Btu gas (producer gas) from biomass has been used to fire industrial process burners including direct fired equipment such as dryers and kilns and indirect fired equipment such as boilers and oil heaters. Low-Btu gas has also been used as a fuel for internal combustion reciprocating engines, both gasoline and diesel, and is being considered for use in gas turbine engines. Medium-Btu gas can be substituted for low-Btu gas in these applications. In addition it can be used as a synthesis gas for making fuels such as methanol, methane (SNG), and liquid hydrocarbon fuels such as gasoline. This section discusses the gas quality, primarily the tar and particulate levels, that are required for each of these end uses.

2.1 BURNERS (BOILERS/KILNS)

The most common use of gas from biomass to date is as a fuel for industrial process burners. Typical applications include direct fired process equipment such as dryers and kilns and indirect fired equipment such as boilers and oil heaters. In these applications only minimal gas cleanup is used. With fixed bed gasifiers the gas is usually piped directly to the burner. With a fluid bed gasifier a hot cyclone is generally used to reduce particulates. The gas can also be cooled and scrubbed prior to burning but this results in a significant loss of efficiency.

There is a wide variety of burners available for burning gas and/or oil. Gas burners are generally of two types. In premix burners air and fuel gas are mixed ahead of the burner ports. Non-premixing burners use a nozzle,

diffuser or spider to mix the gas and air at the burner tile. Most of the burners used for biomass have been the non-premixing type.

Low-Btu gas requires 6-10 times the volume of fuel delivered to the burner and less air than natural gas for the same capacity. For this reason a special burner is required in most cases. One option would be the use of a refractory lined combustor in place of a conventional burner. The primary advantages of this method are minimal pilot fuel usage, less wear and deposition on the boiler tubes, and reduced emissions. Very little work has been done in this area and consequently there are no publications in the open literature as yet.

Boiler and burner manufacturers generally recommend a scroll-type burner for low-Btu gas because it permits passage of large volumes of gas and air with minimal pressure drop (Schwieger 1979). Scroll burners have a history of success on waste gases, such as blast-furnace gas, which has a heating value of about 80 Btu/scf. A modification commonly used for hot dirty gas is the inclusion of self-cleaning steam jets (McGowan and Jape, 1981). Medium-Btu gas can usually be burned in normal natural gas burners. Many references on using medium-Btu gas from coal are available. Little information on using medium-Btu gas from biomass as a fuel gas was found.

Table 10 lists some problems encountered when burning fuel gas from biomass. Derating and other problems associated with the heating value of the gas are not included.

Tars and particulates in the fuel gas contribute to plugging, primarily in the transfer line to the burner. In some applications a standard natural gas burner has been used by reducing the size of the air ports and derating the burner. In these cases plugging of the burner itself becomes more of a problem with the small inlet gas lines.

TABLE 10. Problems Encountered with Biomass Gas Burner Systems

- Plugging due to tar condensation
Transfer line (Williams 1984)
Burner head (Williams 1984; Waibel 1979a)
- Plugging due to particulate deposition
Transfer line (Williams 1984)
Burner (Moreno and Goss 1983)
- Flame out due to low heating value (excess water in the wood and product gas)
(Williams 1984)
- Particulate emissions in flue gas (Moreno and Goss 1983; Oliver 1982)
- Erosion of furnace masonry due to alkalinity of gas (Williams 1984)

Tar and particulates have had no noted effect on combustion in the burner. Variations in gas quality due to fluctuations in water vapor content have caused serious problems with flameouts occurring when the heating value of the gas is reduced. This is usually the result of varying moisture content of the feedstock, particularly in fixed bed units.

The Institute of Gas Technology has tested low- and medium-Btu coal gas on a variety of industrial burners. The burners were a forward-flow baffle, kiln, nozzle mix, high-forward momentum, flat-flame, high-excess air, premix tunnel, and boiler burner. Their primary conclusions were that medium-Btu gas can be retrofit on any natural gas burner and produce stable flames and good thermal performance, but low-Btu fuel gas exhibits more flame stability problems and reduced thermal performance when retrofit to natural gas burners (Waibel 1979 a,b).

IGT also doped the gas with char and tar to determine their effect on burner operation. The doping rates ranged from 0.4 to 2.7 gr/scf (1-6 g/m³) char and 1.1 to 2.3 gr/scf (2.5-5 g/m³) tar. This represents less than 1%

carbon conversion to char for a gasifier with a gas production rate of 2 m³/kg feed. For the most part the tar was consumed in the burner although a char-like residue did build up inside the fuel nozzle in some of the tests when tar was fed. It amounted to 3 wt% of total tar added in the baffle burner and 7 wt% in the kiln. Data were not given for any of the other burners. Most of the char was also consumed in the burner with the survival rate ranging from 5 wt% at the lower loadings to 11 wt% at the higher loadings.

Allowable particulate loadings for gasifier/burner systems will be limited by air pollution regulations for emissions in flue gases. Federal New Source Performance Standards (NSPS) require that particulates be removed to levels of 0.03 lb/106 Btu for utility steam generators over 250 x 106 Btu/hr (Moore 1983, Ford 1980). For industrial-commercial-institutional steam generating units over 100 x 106 Btu/hr new NSPS rules have been proposed which set the particulate emission limits for wood fired units at 0.10 lb/106 Btu (Siegel and Petrillo 1984). However, many gasifier/burner applications are not covered by NSPS, but instead will be covered by various state regulations which range from about 0.1 to 0.6 lb/106 Btu (Ford 1980). Determining which regulations will apply is specific to each application and beyond the scope of this study. At the particulate levels specified by the various air pollution regulations, particulates do not appear to cause any significant operational problems with burners, so stricter limits due to burner limitations are not necessary. Since a substantial portion of char particles are consumed in the burner, higher particulate loadings entering the burner may be permissible.

Allowable tar loadings will be limited primarily by the coupling between the gasifier and the burner. If they are close coupled and the transfer line

is well insulated such systems can handle a heavily tar-laden gas such as produced by a fixed bed updraft gasifier. Longer piping runs (>20 m) will cause deposition and possible obstruction. Reduction in the tar content of the gas (compared to a fixed-bed updraft) will be desirable. Little quantitative information is available regarding the behavior of biomass tars and char in off gas piping. Further research in this area would be beneficial.

2.2 DIESEL AND SPARK-IGNITION ENGINES

The heat energy of low-Btu gas can be converted to shaft work or electricity using diesel or spark ignition engines. The four strokes of an Otto cycle (spark ignition) are:

- 1) Intake -- the fuel-air mixture flows with the cylinder,
- 2) Compression -- the mixture is compressed, ignited and combusted,
- 3) Power -- the high-pressure, high-temperature gases expand, and
- 4) Exhaust -- the piston pushes the combustion gases out of the cylinder.

The Diesel engine differs from the Otto engine primarily in that the temperature at the end of the compression stroke is such that combustion is initiated spontaneously. The higher temperature is obtained by continuing the compression step to a higher pressure. In general, the Otto engine has a higher efficiency than the Diesel for a given compression ratio. However, the compression ratio in the Otto engine is limited by fuel quality (pre-ignition difficulties) so that higher ratios can be used in the Diesel engine, and for that reason higher efficiencies can be obtained with Diesel engines. One measure of the fuel quality is the octane number. The higher the octane number of a fuel the less susceptible the fuel is to auto-ignition. High octane fuels are required for Otto engines and low octane fuels are necessary for diesel engines.

Low-Btu gas has a research octane of about 100 and can be used in a spark-ignited gasoline engine by replacing the carburetor with a mixing chamber. Use of low-Btu gas in a diesel engine requires that 5-10 percent diesel fuel be injected for ignition. This type of operation is referred to as dual-fueling. The engine always has the potential to run solely on diesel fuel with minor adjustments to the timing.

A majority of the stationary engines in operation in the U.S. for electrical generation, pumps, and compressors are full diesel engines. Diesel engines are more popular principally because they are less expensive than spark-ignition engines and fuel costs are somewhat less (Kirkwood 1980).

It is usually desirable to cool the gas before admitting it to either engine. The increased density of gas increases the horsepower compared to using hot gas and prevents premature ignition. As a result, a liquid scrubber of some type can be used for particulate and tar removal. A combination of a cyclone separator and a cloth filter has been used extensively, particularly in vehicular applications.

Some of the problems caused by particulates and tars in internal combustion engines are listed in Table 11. The first two items are related and are

TABLE 11. Problems Caused by Particulates and Tars in Internal Combustion Engines (SERI 1979, Kaupp and Goss 1981)

- Engine wear due to mechanical abrasion
- Contamination of crankcase oil
- Corrosion from organic acids
- Deposition in the gas mixer and inlet pipes
- Gum formation on valves (from tar)

the most serious problems. Particulates are the primary cause of engine wear and oil contamination. Particulates come not only from the fuel gas but from the air as well. The last three items in Table 11 involve volatile material from the biomass and are generally not as serious.

The amount of dust that can be tolerated has been the subject of much research in the 1930's to 1950's. Various regulations developed during this time period ranged from 5-50 mg/m³ particulate in the fuel gas entering the engine (SERI 1979). In a recent review Kaupp and Goss (1981) summarized the effect of particulates on engine wear as follows:

- Up to 10 mg/m³ engine wear is of the same order as observed with gasoline.
- At loadings of 50 mg/m³ engine wear was up to five times greater than observed with gasoline.

DuVant, one of the largest manufacturers of gasifier/engine systems currently recommends the following maximum loadings for tars and particulates (DuVant 1981).

- Particulates - 15 mg/m³
- Tars (condensable volatiles) - 15 mg/m³

A recent study for the Electric Power Research Institute indicated that when using low-Btu gas in engines, particulates smaller than 5 micron should be reduced to less than 20 ppm and tars to less than 10 ppm. This corresponds to 20 and 10 mg/m³ respectively. Other possible contaminants that may cause corrosion are H₂S (should be less than 750 ppm) and aluminum, sodium, and vanadium (no limits given) (Compton 1984).

2.3 GAS TURBINES

Low or medium-Btu gas can be used as fuel for combustion gas turbines to produce electricity or shaft work. A basic direct-fired gas turbine system

consists of a compressor, a combustion chamber, and a turbine. Components added to the system to improve efficiency include a regenerator to recover exhaust losses and preheat the air to the combustor, an intercooler between compressor stages, and an additional reheating combustion chamber between turbine stages.

This type of gas turbine cycle (open cycle) uses air as the working medium and burns relatively clean fuels such as natural gas and petroleum distillates. Burning dirty fuels such as coal, biomass, or low-Btu gas directly in a gas turbine is still in the developmental stage. A major problem is erosion and corrosion of turbine blades by particulate matter and hot gases (Pruce 1980).

The indirectly heated gas turbine cycle (closed or semi-closed cycles) has a significant advantage in that it can accommodate a wide variety of fuels including biomass, low-Btu gas, coal, and lignite. In the indirect cycle incoming air is compressed, heated indirectly by combustion gases using a heat exchanger and then expanded through the turbine. The exhaust air is used to combust the fuel outside of the gas turbine. The heart of the indirect fired cycle is the heat exchanger. Several different heat exchangers for gas turbine cycles are under development. Emphasis is currently on metallic and ceramic materials to protect the heat exchanger from corrosion/erosion and to meet the high temperature requirements.

With both types of turbines (direct and indirect fired), it is desirable to use the biomass fuel gas without cooling (and tar removal) to improve the efficiency. Utilization of biomass gas in gas turbines without cooling and cleaning presents many problems similar to those discussed with industrial

process burners such as plugging in the inlet line, flameouts on poor quality gas (high moisture content), and particulate emissions in the exhaust gas stream.

2.3.1 Direct Fired Turbines

The most severe problem with direct fueled gas turbines is likely to be erosion and corrosion of the turbine blades and buckets. Erosion is a function of the type of particle, size of particle, angle of incidence relative to the surface on which it impinges, velocity of impingement, particulate content of the gas stream, and the physical properties of the surface subject to erosion. Both solids and liquid droplets can cause erosion although the mechanism is different. Erosion is inversely proportional to particle size for the same weight loading. Erosion is a power function of velocity and can be reduced by reducing the turbine blade tip speed.

Corrosion of gas turbine blades is related to erosion and deposition. Erosion may abrade protective oxide films and accelerate corrosion. Deposits of alkali metal sulfates and vanadates may react with the metal in the blades to produce sulfidation and corrosion. Corrosion is more severe as temperatures exceed 700°C.

Extensive experience with gas turbines operating not only on coal but also with dusty inlet air and with dirty fuel gases such as blast furnace gas and petroleum catalytic cracker off-gas indicates that the particulate content of the hot gases fed to the turbine must be kept to less than approximately 1 ppm to keep turbine bucket erosion to an acceptable level for turbine inlet temperatures of 1500°F (815°C) or more. DOE has adopted 0.001 gr/scf (~ 0.3 ppm) as a goal for its high temperature gas turbine program (Moore 1983). Dropping the turbine inlet temperature below 1100°F (593°C) makes it possible to obtain turbine bucket lives of around 3 years with about 100 ppm of

particulates if a number of compromises are made in turbine design (Lackey 1979). The maximum allowable particulate loading specified for high temperature gas turbines in the COGAS process is 0.8 lb/106 scf (~ 10 ppm). The limit for condensible hydrocarbons was set at 0.5 lb/106 scf ($\sim 8 \text{ mg/m}^3$) (Robson 1974).

Corrosion of turbine blades by coal derived gas streams is apparently caused by alkali sulfates (Moore 1983; Lackey 1979). Wood contains about the same quantity of alkali as bituminous coal, about 0.1 to 0.2 lb/106 Btu; however, wood has much less sulfur which may result in reduced corrosion. Other studies in an indirect fired gas turbine using gas from biomass found corrosion in the hot zone of the heat exchanger apparently caused by alkali sulfates (see page 41).

The U. S. DOE Fossil Energy Gas Stream Contaminant Control Program has established a goal of 0.02 to 0.04 ppm alkali in coal gas to prevent corrosion in high temperature gas turbines (Moore 1983).

Gas turbine experience most relevant to biomass gases is that of Brown Boveri Turbomachinery Inc. who have installed more than 20 units in the last 25 years to burn blast-furnace gas. These machines are characterized by low pressure ratios (about 4.5) and turbine-inlet temperatures of about 1400°F (760°C). They range in size from 2-14 MW. After extensive testing of hot-gas cleanup equipment, it was found best to cool this gas and clean it with a two-stage wet scrubber and/or an electrostatic precipitator prior to burning it in a combustion chamber ahead of the turbine (Lackey 1979, Pfenniger 1964). The same approach and requirement have been imposed on the low-Btu gas supplied from Lurgi coal gasifiers to the gas turbine of the combined-cycle plant in Lunen, Germany (Lackey 1979).

2.3.2 Indirect Fired Turbines

Use of an indirectly heated gas turbine is one means of avoiding erosion and corrosion of the turbine blades. This cycle depends on a high temperature heat exchanger to transfer heat from the hot dirty combustion gases to a clean gas, typically air which passes through the turbine. There is very little data available for these systems as they are in the early stages of development; however, biomass gas has been used in an indirectly heated gas turbine cycle.

Gas from fluidized bed gasification of high ash agricultural wastes was used to run a small (30 kw) indirectly fired gas turbine in tests by Advanced Energy Applications, Inc. The major difficulties encountered were corrosion in the hot zone (1600°F) of the heat exchanger due to alkali sulfates and fouling in the hot zone from ash deposits (Moreno and Goss 1983). No information was given on the type of gas cleaning that was done, but the conclusion was that development of new high temperature filter would be necessary to prevent ash fouling. Corrosion could be reduced by material changes.

2.4 SYNTHESIS GAS/PIPELINE GAS

A potentially major use for biomass medium-Btu gas in the long term is as a synthesis gas for methanol, hydrocarbons, or methane (SNG). In these applications the gas goes through several processing steps between the gasifier and the catalytic synthesis unit. These include:

- Particulate and Tar Removal
- Shift Conversion
- Acid Gas Removal
- Compression
- Trace Gas Removal

The downstream processing streams and the synthesis catalysts used for these process are all quite similar so they will be considered together. Methane to be transported by pipeline will have to meet additional specifications for the final product gas.

Potential contaminants for methanol synthesis catalysts are shown in Table 12. For some contaminants such as tars and oils the contaminant limits are not well defined. To date most commercial synthesis gas plants are based on natural gas and many of these contaminants have not been a problem. Catalysts for hydrocarbon synthesis (Fischer-Tropsch) and methanation (SNG) are quite similar to methanol catalysts so the contaminant problems should be similar.

Some potential problems other than catalyst poisoning or fouling that can be envisioned are listed in Table 13.

TABLE 12. Potential Methanol Synthesis Catalyst Contaminants
(White 1983, Bennett 1980, Connor 1982)

<u>Synthesis Gas Component</u>	<u>Contamination Level and Potential</u>
C_2H_2	Potential Catalyst Poison @ 3 ppm
C_2H_4 , Higher Olefin	Presently Unknown but Possible Catalyst Poisonous at Higher Concentration
CH_4 , Higher Paraffin	Inert
Sulfur (H_2S , COS , CS_2)	Poison @ 0.03 to 0.2 ppm for Cu based catalyst Poison @ 10 ppm for Zn based catalyst
Chlorides	Poison @ 0.03 to 0.2 ppm for Cu based catalyst
NH_3 , HCN	Possible Catalyst Poison at Modest Concentration
Particulate	Possible Poison and Affect Catalyst Life Due to Bed Plugging
Oil Mist	Possible Catalyst Poison at Modest Amount

TABLE 13. Other Potential Problems in Synthesis Gas Production

Particulates

- Plugging of CO shift converter
- Contamination of acid gas scrubbing liquor
- Erosion of compressor blades
- Deposits in process

Tars or Oil Mist

- Fouling or poisoning of shift catalyst

The most serious of these problems is likely to be erosion of compressor blades by particulates. Operating data from a Koppers-Totzek coal gasifier in Kutahya, Turkey indicate that solid particulates in raw gas are reduced from 12 gr/scf down to 0.002 gr/scf in venturi scrubbers and further cleaned to 0.0001 gr/scf in an electrostatic precipitator if the gas is to be compressed to high pressure for chemical synthesis or for the production of high-Btu gas (Farnsworth 1974). These numbers seem very low in light of other information obtained for gas turbines (see previous section). In an engineering evaluation of a lignite to methanol plant, Davy McKee International specified that the raw product gas be cleaned to 0.01 gr/scf prior to compression (Goyen 1980).

In the case of methane which will be transported by pipeline, some additional limits are placed on the final gas quality. The Office of Coal Research issued the following guideline objectives for development of processes to make high Btu gases from coal. The gas should be deliverable at 1000 psig and have the properties shown in Table 14.

TABLE 14. Properties of Synthetic Natural Gas (SNG) (Strakey 1975)

Heating Value	> 900 Btu/scf (HHV)	CO ₂ < 3%
CO	< 0.1%	Water < 716/106 scf
H ₂ S	< 0.25 gr/100 scf	SpGr 0.59 - 0.62
Total S	< 10 gr/100 scf	Hydrocarbon <40°F at 1000 psig Dew Point
Inerts	< 5%	No poisonous compounds or gum formers

2.5 CONCLUSIONS

The allowable particulate and tar loadings in biomass gas varies significantly depending on the end use. Figure 9 summarizes the requirements for particulates. If the gas is used in a burner to supply energy, particulate removal will be governed primarily by air pollution regulations for particulates. The allowable loadings in the combustion flue gas range from 0.03 to 0.6 lb/106 Btu depending on the size and location of the facility. The specific allowable limit will be site specific.

For internal combustion engines, particulates must be removed down to 10-50 mg/m³ to prevent excessive engine wear. Requirements are even more strict to prevent erosion and corrosion of gas turbine blades; particulates must be reduced to 0.001 to 0.01 gr/scf depending on turbine speed and operating temperature.

Less information is available for gases to be used for synthesis of chemicals, SNG, or gasoline. In general the requirements will be similar to specifications for gas turbine applications and will be dictated by erosion of compressor blades in the compressors used to compress the gas to high pressures required for synthesis.

FIGURE 9. Allowable Particulate Loadings for Various End Uses

Specifications for tars in the product gas are not well defined. Tars will be consumed in any burner (including gas turbine combustors) and increase the heating value of the gas. However, this requires close-coupling the gasifier and the burner. Separation of the gasifier and the burner by any distance can cause significant problems due to tar condensation and precipitation. Tar removal should be considered in this case.

In diesel and spark ignition engines tars must be reduced to 10-50 mg/m³ to prevent deposition in the gas mixer and inlet valves and gum formation on the valves. The limit on condensible hydrocarbons entering the high temperature gas turbine have been set as low as 0.5 lb/106 scf (~ 8 mg/m³) although no reasons have been given for the limitation (Robson 1974).

Tars are known to be poisons for various synthesis catalysts, however, their effect is not well understood, nor have limits been set. For synthetic natural gas entering a pipeline a hydrocarbon dew point of $<40^{\circ}\text{F}$ at 1000 psig ($\sim 100 \text{ mg/m}^3$) has been specified. In addition no gum formers are allowed. Tars would probably be classified as gum formers.

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3.0 GAS CLEANUP TECHNOLOGY

Fuel gases derived from biomass gasification resemble gases derived from coal gasification with respect to major fuel constituents, but differ with respect to major impurities. Because coals are generally much higher in sulfur and ash, these constituents tend to be higher in coal gas than in biomass gas. The spectrum of trace elements derived from coals may also be different from that derived from biomass fuels. Finally, the detailed chemical makeup of tars and oils derived from coals will differ significantly from those derived from biomass gasification. There has been a vast amount of work done on the problem of cleaning coal derived gases and with proper attention to the differences imposed by gasification of biomass fuels much of this technology can be applied.

Various gas cleanup systems exist for removal of both particulates and tars from gases produced by biomass gasification. Often these overlap, particularly when tars are present as liquid droplets. Well-documented data with these systems on biomass gas is limited; however, some actual operating data with biomass is included. Much of the information presented here is based on related experience with coal conversion.

3.1 DEFINITIONS

The primary contaminants of concern in this report are particulates and tars. Particulate is used to refer to solid particles of char and ash. These materials are also commonly referred to as dust. Tar refers to hydrocarbons and oxygenated hydrocarbons which are normally liquid at atmospheric pressure and ambient temperature. In the gasification process the tars may be present as either vapor or as liquid droplets. Liquid dispersions in a gaseous medium are sometimes referred to as sprays or mist. Small solid

particles and liquid droplets suspended in a gas are often collectively referred to as aerosols.

Figure 10 shows a range of particle and droplet sizes and the type of collection mechanism appropriate for various sizes. Although tars are often present as vapor they are removed from the gas as liquid droplets following condensation. In addition to particle size, separation may be effected by other properties of the solid particle or droplets such as electrical resistivity, stickiness, combustibility, corrosiveness, and toxicity (Razgaitis 1977).

FIGURE 10. Particle Size Classification and Useful Collection Equipment

3.2 PARTICULATE REMOVAL

Solid particulate is generally fly ash or char (partially burned feedstock). The class of separators most widely used for removal of particulates are the centrifugal type units (cyclone). For more rigorous removal electrostatic precipitators, baghouses, and other filtering methods are available. These separators are generally operated above the dewpoint of any tars, oils or water that might be present to avoid any sticky combination of tars and char. Depending on the physical properties of the contaminants this may be from 150°C to 500°C.

3.2.1 Cyclones

Cyclones, or inertial separators, operate by imparting centrifugal force to the particles to be removed from the gas stream. This is done by directing the gas flow in a circular path (Fig. 11), which due to their inertia, the particles cannot follow. Design procedures for cyclones with predictable performance are based on theory and supplemented by empirical techniques accumulated from operating experience (Stone & Webster 1974, Zenz 1982). For this prediction, the nature of the particles and especially the particle size distribution must be well defined.

Razgaitis (1977) asserts, on the other hand, that prediction of performance characteristics of cyclones is not straightforward. He provides an elegant and well annotated discussion of the problem and of the limitations (as well as successes) of the various quasi-empirical correlations which have been developed to predict the efficiency of cyclones. Detailed descriptions of conventional and advanced auxiliary injection cyclones are given in this reference with ample literature citation. For example, the most widely applied new-technology cyclone described by Razgaitis is that developed by System Siemens and which is available in the U.S. by license to Aerodyne

FIGURE 11. Cyclone Flow Patterns

Development Corporation. These are direct flow cyclones with vane axial injection and which employ auxiliary air (gas) injection for particulate removal. They can be operated at temperatures up to 900°C and 30 atm. and have exhibited collection efficiencies of 90% for 1 micron particles at pressure drops of only 1.5 cm Hg. For comparison, conventional cyclones designed for a 2 to 3 micron cut off (50% collection efficiency @ this size range) would collect no more than 20% of 1 micron particles (Stone and Webster 1974). Clearly, for collection of sub-micron particles, cyclones will be virtually useless. For collection of particles > 2 micron in diameter they

are an excellent choice. Cyclones are manufactured in almost any size, from miniclones at less than 1/2" diameter to large capacity units up to and over 10 feet. Capacity of larger units can be hundreds of thousands of cubic feet per minute. Series cyclones are quite common and as many as three or four units can be coupled (Fig. 12).

Design and operating data are available for a two-stage cyclone particle removal system for cleaning low BTU gas derived from fluid bed gasification of cotton gin trash (Parnell 1981). Cyclones with tangential inlet and axial dust discharge were designed. The two cyclones were connected in series. The primary cyclone was designed to be less efficient than the secondary cyclone but would still collect most of the particulate. The secondary

FIGURE 12. Two Stage Cyclone System in a Fluid Bed

cyclone was designed for higher efficiency and for collection of smaller particles. With this arrangement the smaller diameter (secondary) cyclone is less subject to plugging.

Operating with an inlet particle burden of 162 g/m^3 the primary cyclone operated to remove 93.1% and to yield an effluent containing 11.1 g/m^3 . The secondary cyclone operating at 98.7% efficiency produced an effluent containing 0.14 g/m^3 .

Omnifuel installed a 12-foot diameter fluidized bed gasifier at Hearst, Ontario. The gas passed through a single cyclone and was burned in four oil heaters. In one test approximately 92 wt% of the particulates in the fuel gas were removed by the cyclone. Particulate loadings were reduced from approximately 3.5 gr/ft^3 (8 g/m^3) in the product gas leaving the gasifier to 0.15 gr/ft^3 (0.4 g/m^3) in the combustion flue gas (Oliver 1982). The 92% efficiency is obtained by dividing the particulates removed in the cyclone by the sum of the cyclone particulates and the particulates in the flue gas. Because a significant portion of the carbon particles carried over to the burners are consumed there and are not accounted for by this measurement, the 92% efficiency probably overestimates the cyclone efficiency.

PNL has tested several gas cleaning systems with their 1 ton/day fluidized bed gasifier. The gasifier generates a raw gas containing a typical particulate burden of 32 gr/ft^3 (70 mg/m^3). A single cyclone generally removed 60-85 wt% of the particulates (Mudge 1983).

In tests with a fixed bed gasifier on a diesel tractor a single cyclone removed 53-67 wt% of the particulates. The low efficiency is due primarily to the low inlet velocity which occurred at low engine loads (Kaupp and Goss 1981).

Most wood fired boilers use multiple small cyclones in parallel (multiclones) for primary particulate control. In the past this was enough to meet air pollution regulations; however, secondary particulate removal systems, usually wet scrubbers, are now necessary for most systems (Brady and Jenkins 1980, Russell 1978, Brown 1978). For example, combustion gas from a wood fired, 60,000 lb/hr, Babcox & Wilcox, stationary grate boiler, was cleaned to 0.38 gr/ft^3 (0.8 g/m^3) in a multiclone and further cleaned to 0.007 gr/ft^3 ($.15 \text{ mg/m}^3$) in a wet scrubber (Scott 1980). Another combination wood (80% of heat input) and oil fired boiler uses two high efficiency cyclones in series for particulate removal. The overall efficiency of the two collectors was 94% and the concentration of the particulates averaged 0.18 g/m^3 (Ananth 1981). Zenz (1982) also makes reference to a "bark boiler" with cyclones only 58% efficient.

In general, high efficiency cyclones (multiclones) have collection efficiencies of 60-85 wt% for hogfuel flyash. High efficiency cyclones in series can increase collection efficiency to over 90 wt%.

Two large cyclones in series have been used to remove particulates for wood combustion gas prior to firing the gas in a 375 kw gas turbine. After the final cyclone the gas contained 8-12 ppm particulates ($.11 \text{ mg/m}^3$). Operation on one turbine for 210 hours and a second for 270 hours with no detectable erosion. A similar 3000 kw turbine unit is under construction (Hamrick 1983).

There are a large number of cases where a cyclone has been employed as a primary particulate remover to be followed by more stringent cleaning devices such as filters, venturi scrubbers or ESP's.

3.2.2 Electrostatic Precipitators

Alone among methods of particle collection, the electrostatic precipitator (ESP) acts solely on the particles to be collected and not on the entire gas stream (Manhattan College 1978). The pressure drop in an ESP is about the same as in an equivalent length of pipe.

The process of electrostatic precipitation consists of corona formation around a high tension wire with particle charging by ionized gas molecules. ESP's operate by using a high voltage, direct current to create gas ions that impart an electrical charge to particles by bombardment. The charged particles are collected by exposing them to an electric field which causes them to migrate and deposit on electrodes of opposite polarity (see Figure 13).

The electrode cleaning system is dependent on the type of precipitator. Dry type precipitators, which can operate up to 540°C, remove particles by mechanical shaking and rapping. Wet type precipitators collect and remove the particles with a thin continuous flowing film of water. They generally operate at less than 65°C (Kennedy 1979).

The usual approach taken to design (size) ESP's for particle collection (Manhattan College 1978, Schweitzer 1979) makes use of the Deutsch-Anderson equation:

$$n = 1 - \exp(-wA/Q).$$

The term w represents the effective migration velocity or precipitation rate parameter which is selected on the basis of experience with various types of particulate. Values range from 0.13-0.67 ft/sec with values of 0.33-0.44 ft/sec often found on precipitators collecting fly ash from firing pulverized coal. Since the desired collection efficiency (n) and gas flow rate (Q) are usually specified, the required collection area (A) can be determined once an appropriate precipitation rate parameter (w) has been selected. In general,

FIGURE 13. Two Stage Electrostatic Precipitator

the precipitation rate parameter decreases with increasing ash resistivity, and increases with increasing sulfur content of the fuel, and corona power density.

It is well known that ESP's perform poorly on ash derived from low sulfur coals (SO₃ injection may be required). Changing from coal with 2% S to a sub-bituminous coal with 0.5% S can result in an efficiency loss from 99.5% to 90% or less (Manhattan College 1978). Many sub-bituminous coals, on the

other hand, are high in sodium (as Na_2O) and this lowers the resistivity resulting in better removal efficiency.

Ash derived from wood consists of oxides of Ca, K, Na, Mg, and Si. Table 15 gives the composition range for wood ash and coal ash.

TABLE 15. Composition of Ash from Wood (Kaupp and Goss, 1981)
Compared with U.S. Coals (Lowry 1963)

<u>Component</u>	<u>Ash Analysis, %</u>	
		<u>U.S. Coals</u>
Al_2O_3	1-10	10 - 35
SiO_2	0-2	20 - 60
CaO	10-60	1 - 20
MgO	1-17	0.3 - 4
Na_2O	1-20	1-4
K_2O	2-4	
Fe_2O_3	0.5-4	5 - 35

These differences can be highly significant to the design of ESP's as well as other separative devices. For example, the wood ash may be much more conductive than coal ash due to its high alkali content, which of course is extremely deleterious to turbine end usage.

Electrostatic precipitators (ESP) are seldom used on boilers firing only hog fuel due to problems with the resistivity of hog fuel flyash (Brady and Jenkins 1980). ESP's have been used on an MSW fired municipal incinerator. Gas from two 39,000 lb/hr Riley Stoker boilers pass through single field ESP with 413 m^2 of collection area. The ESP has a design collection efficiency of 93% and reduced particulates to 0.074 from 0.55 gr/ft^3 (1.2 g/m^3).

Wood wastes are burned in a boiler at the Kettle Falls (Washington) Generating Station to produce 42.5 mw of electricity. Pollution control here is accomplished with roughing cyclones followed by an ESP. This system is designed for an emission level of less than 0.02 gr/ft^3 (40 mg/m^3) (Henriques 1983).

Gas from a PuroxTM fixed bed gasifier feeding MSW were cleaned in a wet scrubber followed by an ESP. Following treatment the gas contained 0.002-0.005 gr/ft^3 (10 mg/m^3) of particulates (Ananth 1981). Hoenig and Cole (1981) indicate 98.7% of the "dust tars" from a Pegasus downdraft unit were removed by an ESP. No indication of the properties of the particulates or tars was given.

PNL's 1 ton/day process development unit fluid bed gasifier originally used a cyclone followed by a venturi scrubber that was later replaced by an electrostatic precipitator (ESP). Both the venturi and the ESP experienced plugging problems caused by a sticky combination of char and tar (Mudge 1983).

ESP's are used for final particulate removal from a commercial Koppers-Totzek coal gasifier. Gases are first cooled and scrubbed in a venturi unit prior to entering the ESP. The ESP reduces particulates from 0.002-0.003 gr/ft^3 ($4\text{-}6 \text{ mg/m}^3$) to 0.0001 gr/ft^3 (0.2 mg/m^3) (Farnsworth 1974).

3.2.3 Baghouses

Baghouse filters are one of the oldest and most widely used techniques for removing particles from gas streams. The primary advantage of bag filters is their very high efficiency in dust removal. Other advantages include low energy use and pressure drop and collection of particles in a dry form which simplifies disposal. Disadvantages include the large size and high cost. In addition temperatures are limited to about 290°C (Kennedy 1979). A typical reverse-flow cleaning baghouse is shown in Figure 14.

FIGURE 14. Typical Reverse-Flow Cleaning Baghouse

The mechanism of the filtering process is complex. Small particles are initially retained on the fibers of the filter material by interception, impingement, diffusion and settling, and electrostatic attraction. After a filter cake accumulates, further collection is by sieving, which is primarily responsible for the high collection efficiency attainable. Bag filters typically allow collection of <2 micron particles with 99.9% efficiency (Stone and Webster 1974). Maximum pressure differential is approximately 6 inches of water prior to cleaning. Typical capacities are $1.9\text{--}2.8 \text{ ft}^3/\text{ft}^2$ (Manhattan College 1978).

Baghouses are becoming more common in large utility industry installations with low sulfur coal where the effectiveness of ESP's is reduced and they would tend to become very large (and expensive). No biomass gasifiers are known to employ bag filters and they have not been used much for wood and bark fired boilers. One problem is their susceptibility to fires caused by glowing char in the flue gas (Ford 1980).

3.2.4 Granular Bed Filters

Granular bed filters employ a stagnant fixed bed of sand or other media to trap particulates by impaction. A typical granular bed filter (Westinghouse) is shown in Figure 15. Fluidizing gas is introduced once a specified

FIGURE 15. Granular Bed Filter

pressure differential (1-3 psi) is reached to clean the captured particulate from the much larger filter media. Typical media sizes are around 15-30 mesh and bed depths are 2-4 inches (Zenz 1982).

Commercial uses of granular bed filtration are limited although Zenz (1982) refers to a down-flowing pebble bed filter 75% to 97% efficient on effluent from a hog mill waste combustor. The bed was an annulus 8.5 ft OD x 6 ft ID, 16 ft tall with gravel 1/8 in to 1/4 in in diameter. Pressure drop across the bed was about 12 inches of water and the unit was operated at 300-350°F. It appears that the particulate was removed from the bulk material by sieving with the pebbles being recycled to the top of the bed.

These devices have been characterized as highly efficient though complex (Razgaitis 1977). In principle a pebble bed filter can be built very large so the filter area is extensive and sufficient to achieve any desired efficiency. The trouble is, sooner or later it has to be regenerated. This can prove quite troublesome, especially with sticky particles.

Granular bed filters currently under development include the Ducon fluidizable bed filter, Panel Bed (fixed backflush) filter, Rex gravel bed (stirred backflush) and various proposed filters under study at Westinghouse, Consolidation Coal Co., Dorfman, Carnegie-Mellon, and Lurgi, M.B. All involve similar principles of collection and regeneration.

A particular advantage of this filtering concept is that the filter media can also serve as a chemisorbant for alkali when operated at high temperatures, 1600°F (870°C) (Mulik 1983). The use of alumina or aluminosilicates allows efficient alkali removal as well as particle collection. Using 2 mm alumina spheres and operating at 1600°F Combustion Power obtained 99.5% particulate collection efficiency on inlet gas loading of 0.5 to 3.5 gr/ft³ (1.1-8.0 g/m³) with median particle size of 5-10 microns (Lee 1983).

3.2.5 Other Barrier Filters

Additional barrier filtration methods are available (Henry, Saxena, and Podolski, 1983) but are not widely used due to their cost or complexity. Porous metal filter elements (Fig. 16) have been used for years in catalyst recovery, nuclear waste calcination, and other situations where particulate removal is desired from hot gas or liquid streams. With proper design and materials (InconelTM) these filters may be operated at 815-925°C (1500-1700°F). Nominal pore sizes range from 0.5 to 100 microns. Typical removal efficiencies are 99.999% for particles greater than 0.05 microns properly designed filter elements. The inherent high cost, slow sintering at high

FIGURE 16. Sintered Metal Filter Assembly

temperatures, plugging, and possible failure due to thermal shock may make these units uneconomical other than for research scale or special applications.

In tests at PNL's fluid bed gasifier high temperature sintered stainless steel filters were placed immediately after the cyclone for additional particulate removal. These filters had a pore size of 10 micron and were operated above the dewpoint of the tar. After the filters the gas was cooled and the tars separated with the condensate in a demister column. This system operated without difficulty (Mudge et al. 1983).

Similar filters are now being constructed based on porous ceramic barriers (Cilberti 1983). These filters are designed to withstand higher temperatures and collect very fine particles with maximum efficiencies. They suffer from many of the problems of sintered metal units and are as yet unproven. Despite the limitations of these filter systems the potential economic advantages of a high temperature/high pressure particulate removal system is enough to justify continuing development for use with commercial coal gasification systems (Rubow 1984).

3.2.6 Wet Scrubbers

Gas scrubbing, using various liquids, can be highly effective for particle removal. Water is the most common scrubbing liquid although other liquids have been investigated. Wet scrubbers have been used extensively on wood and bark fired boilers in the wood products industry (Scott 1980). Wet scrubbers have also been used in coal-fired plants but on a large scale, economics generally favor ESPs or baghouses (Ford 1980).

There are a wide variety of wet scrubbers, but for particulate control in boiler flue gas, venturi scrubbers (Fig. 17) are by far the most common.

FIGURE 17. Venturi Scrubber with Cyclone Separator

Water contacts the dust laden gas in a restricted area (throat). Gas velocities are typically 200-400 ft/sec in this throat area (Manhattan College 1978). Particles are collected by collision with water droplets. The liquid is entrained in the gas stream and must be de-entrained in a mist eliminator. Variable throat scrubbers permit close control of pressure drop and efficiency. Efficiencies are proportional to the design pressure drop. For particles over 10 μm the design pressure drop is typically around 1 inch of water (.25 kPa). For particles less than 10 μm pressure drops of 10-100

inches of water (2.5-25 kPa) have been used (Kennedy 1979). Typical efficiencies are 99.9% for 2 μm particles and 95-99% for 1 μm particles.

An irrigated packed column containing an open type packing such as rings or saddles is also an efficient collector of particles. Collection efficiencies of 50% for 1 μm particles have been demonstrated at laboratory scale (Jackson and Calvert 1966). With counter current irrigation of the packing with a working fluid, the collected particles can be continuously flushed from the packing. Here the primary collection mechanism for particles above 0.5 μm is inertial impaction. Due to their inertia the particles cannot follow the gas flow around the packing. Continuous irrigation of the packing should effectively prevent re-entrainment of agglomerated particles.

Wet scrubbers and cloth filters were used extensively with downdraft vehicular gasifiers in Sweden. Baffle plate wet scrubbers generally removed particulates down to about 50 mg/m^3 , which corresponded to 95-99% collection efficiency (SERI 1979).

A venturi scrubber is used to cool and clean a tar-free gas from a commercial Koppers-Totzek coal gasifier. Particulates were reduced from 12 gr/scf to 0.002 gr/scf which corresponds to 99.98% efficiency (Farnsworth 1974).

A wet scrubber is used with a Halcyon commercial fixed-bed gasifier to remove particulates and condensed tars. Removal efficiencies of 96% for all particles over 5 micron and 98% for all particles over 10 microns were obtained (Finnie 1979).

3.3 TAR REMOVAL

Tar removal lowers the heating value of the gas produced by a biomass gasifier. It may become necessary, however, because of the effect of tars in

plugging lines and instrument ports rendering operation of downstream equipment difficult. Tars may be present in two forms. In a fixed updraft gasifier where the gas exit temperature is low, tars will be present primarily as liquid droplets entrained in the gas. In downdraft and fluid bed gasifiers where the product gas is hot, tars may also be present in the vapor form. Only at high temperatures can particulate and tar removal be separated. At temperatures where tars and oils condense to liquids, removal of tars and oils cannot be divorced from the particulate removal problem (all collect together).

3.3.1 Wet Scrubbers

Wet scrubbers are widely used to cool the gas and remove the precipitating tars and oils and also can be used for solid particulates as previously discussed.

The primary collection mechanism is impingement of the particles on liquid (usually water) droplets. There are a wide variety of wet scrubbers varying primarily in the method of liquid contacting. The most common types are spray towers, packed bed scrubbers, plate scrubbers, impingement scrubbers, baffle scrubbers, and venturi scrubbers. The design and construction of wet scrubbers is covered in detail by Calvert (1972), Lucas (1973), and Kaupp and Goss (1981). Practices in the coke-oven and byproducts industries are widely used for tar removal in all gas manufacturing processes (Parekh 1982).

In a typical spray tower most of the tar is condensed by contact with water sprays. In addition to cooling the gas and condensing tar the liquid washes condensed tar droplets from the gas (together with particles). The tar and liquid flow to a decanter where tar (and particles) settle and separate from the bulk of the liquid (Figure 18).

FIGURE 18. Example of Spray Tower for Tar Removal

Another type of tar scrubber used with many early gas producers was the Lymn washer. It consisted of a series of cones and disks through which the gas and water passed counter-currently. Other scrubbers that have been used with producer gas are rotoclones and Chevron vane separators.^(a)

Table 16 shows the pressure drop and the efficiency of various wet scrubbers. Spray towers are the simplest and least expensive and have the lowest efficiency. Venturi scrubbers (described in 3.2.6 and Fig.16) are the most complex and most efficient.

(a) Information provided by Mr. Kerry L. Rock, Davy McKee, Tulsa, Oklahoma

TABLE 16. Effectiveness of Spray Towers (Lucas 1979; Buonicore 1980)

	<u>Pressure Drop</u> <u>inches water</u>	<u>Particle Size, μm</u> <u>for 80% collection</u>
Spray Tower	0.5-1.5	10
Impingement	2-50	1-5
Packed Bed	2-50	1-10
Venturi	5-100	0.2-0.8

Wet scrubbers all produce a liquid stream which usually requires further treatment for tar recovery prior to disposal. Problems with stable emulsion formation are not uncommon. Oils from biomass gasifiers are quite likely to contain an appreciable fraction of aqueous soluble carboxylic acids, aldehydes, ketones, and substituted phenols such as have been reported in products derived from "flash pyrolysis" and "entrained flow pyrolysis" of wood (Elliott 1985). In addition, the oils which can be decanted are likely to still contain appreciable amounts of water which lower their heating value and reduce their viscosity.

Wastewater samples from the fluid bed, air blown steam gasifier at Texas Tech University have been analyzed at PNL (Petty 1981). Testing confirms the presence of acetic and propionic acids which accounts for pH levels as low as 2.3. Chemical oxygen demand (COD) ranged from 30,800 mg/l to 67,900 mg/l. Nitrogen levels were generally low, but two samples contained in excess of 5400 mg/l (reflected by higher pH (5.5-7.4) which was probably due to the presence of NH_3 . Significantly lower COD has been reported for a downdraft gasifier, about 30 mg/l (Reed 1982).

A recent analysis of condensate from nine experimental biomass gasifiers showed the pH ranged from 2.1 to 5.3 for fixed bed gasifiers and 5.9 to 8.7 for fluid bed gasifiers. Total organic carbon soluble in the water ranged from 10 wt% for the fixed bed gasifiers and was generally less than 0.1% for the fluid bed gasifiers (Elliott 1985).

3.3.2 Centrifugal Extraction

Cyclonic devices are more applicable for solid particulates (as described in Section 3.2.1), but have some application with tar or water droplets greater than 5μ . No data on efficiency was found on using centrifugal devices for biomass tar removal although they have been used (Oliver 1982; Miller 1983). Prater et al. (1984) use a humidifier/cyclone combination to remove 50-90% of the coal tars from a pressurized 1 ton/day fixed bed updraft coal gasifier. More viscous tars may require that the unit's walls be heated to avoid plugging.

3.3.3 Electrostatic Precipitators (ESP's)

Electrostatic precipitators efficiently charge and collect oil mists and tar droplets together with particles. In this operation the collecting plates are washed continuously with water to remove collected material. Operating temperatures for tar removal are generally near 150°F (76°C). Other operational characteristics are essentially the same as with particulates (Section 3.2.2).

3.3.4 Catalytic (Thermal) Cracking

Fluidized bed gasifiers yield a gas containing tars and oils in vapor form which adds significantly to the heating value of the gas. Researchers in the U.S. (Mudge et al. 1981, 1983) and Sweden (Ekstrom, Lindman, and Pettersson 1982) are studying the use of catalysts to convert tars to gases. Catalysts have been used in the gasifier and directly downstream in a secondary vessel. Maintaining catalyst activity appears to be the primary problem with this method (Baker and Mudge 1984).

3.4 OPERATING CONDITIONS, EFFICIENCIES, AND COST

The main features of commercial and developing particulate and tar removal systems are summarized in Table 17. Cyclones can operate at the highest temperatures followed by dry electrostatic precipitators (ESP's) and granular bed filters. Baghouses are limited to about 290°C by the fabrics they utilize. Liquid scrubbers are limited by the vapor pressure of the scrubbing liquid, usually water. Filter systems capable of operating at higher temperatures are under development. The effects of operating at pressure are documented for cyclones, electrostatic precipitators, barrier filters, and other hot gas cleaning methods (Henry, Saxena, and Podolski 1982, Gilbert 1978). Generally, pressurized operation reduces the size of gas cleaning equipment. Wet scrubbers, however, are generally used only on atmospheric pressure gasifiers and consequently have not been researched extensively. Research in this area would be beneficial.

Typical efficiencies for several of the most common gas cleaning systems as a function of particle size are shown in Figure 19. Cyclone separators are the least efficient. Wet scrubbers are somewhat more efficient and baghouses (fabric filters) and ESP's are the most efficient particularly for submicron particles.

Figure 20 shows typical installed capital costs for these same systems. In general the cost of the gas cleanup equipment is directly related to its efficiency.

TABLE 17. Summary of Gas Cleanup Systems (Stone and Webster 1974)

Commercial Systems	Maximum Operating Conditions		Inlet or Face Velocity ₂ CF/min/ft ²	Typical Pressure Drop in H ₂ O	Collects Condensed Tars
	Operated °F/PSIG	Projected °F/PSIG			
Cyclones	1700/high	>2000/high	4000	4 - 6	Yes
Electrostatic Precipitators	900/15	1700/300	200 to 300	15 - 35	Yes
Bag Houses	550/15	---	5 to 10	15 - 30	No
Granular Bed Filters	>900/high	>2000/300+	100	9 - 12	No
Liquid Scrubbers		175/15	--	9 - 12	Yes
Aerodyne Tornado	900/500	1700/500	--	13	Yes
<u>Developing Systems</u>					
Fiber Filters	--	1500/high	100	--	No
Porous Metal	800/15	>1500/high	2 to 5	--	No
Ceramic	3450/15	---	2 to 5	--	No
Liquid Scrubbers	1400/15	1400/100	--	--	Yes
Panel Bed	1000/15	>2000/high	30 - 45	--	No
Ducon	600/15	>1500/high	30 - 90	--	No
Pebble Bed	250/15	1500/high	30 - 100	--	No

FIGURE 19. Typical Gas Cleaning Efficiencies

FIGURE 20. Total Installed Costs for Particulate-Control Devices (Perry 1973)

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4.0 EVALUATION AND RECOMMENDATIONS

4.1 SYSTEMS ANALYSIS

There are a multitude of equipment combinations available for cleaning gases produced by biomass gasification. Each type of gasifier has cleanup options which are best suited to that particular gasifier. This chapter combines gasifier pollutant levels, gas cleanup efficiencies, and end use requirements to evaluate total gas cleanup systems which are applicable. Recommendations on areas for additional research and development are identified.

4.1.1 Methodology

Each type of gasifier was described in Chapter 1. Generic particulate and tar loadings (Fig. 7 and 8) were derived which enables complete systems analyses to be done. Various gas cleanup methods and their efficiencies were compiled in Chapter 3. Figure 19 summarizes relative particulate removal efficiencies for some of the more common devices. No detailed information on tar removal efficiencies was found in the literature.

Calculations of system efficiency and end use applicability consider all of these: pollutant concentrations, gas cleanup devices, and end uses. Because no information on tar removal efficiencies was found several assumptions had to be made to complete the calculations: 1) particulates and tar could not be removed in the same step due to the sludge that would result and 2) when tar removal was necessary, wet scrubbing to condense and remove tars would be required (no other removal techniques were considered applicable to tar removal). This method is used for tar removal in commercial fixed-bed coal gasifiers (Kennedy 1979). While these assumptions may not be strictly true, they represent the current state-of-the-art as we see it.

A typical calculation involves the following steps:

- Obtain gasifier outlet particulate loading and size distribution.
- Get efficiencies from Figure 19 for each cleaning device.
- Calculate outlet concentrations for each combination of gas cleaning devices.
- Compare calculated concentrations with performance standards for each end use.

4.1.2 Results

Table 18 tabulates the various particulate emission levels calculated for each type of gasifier and applicable cleaning technology. For fixed-bed gasifiers several technologies were considered not applicable because they would remove particulates and tar together. For fluid bed and entrained bed gasifiers wet scrubbers as a first cleanup step was considered not applicable as a tar and particulates would both be removed. This also results in a significant energy penalty for these high-temperature gasifiers.

Table 19 shows the gas cleanup combinations that achieve particulate performance standards for the various end uses. Where tar removal was required it was assumed the wet scrubbing would meet the tar removal requirements.

The results, shown in Table 19 show that, in general, particle scrubbing appears to be adequate to deal with biomass gas if simultaneous removal of tar is not required. However additional engineering data is necessary to properly design gas cleaning systems to assure that the system operates reliably and that environmental restrictions are met. These include the size and composition of particulates emitted and the effect of gasifier operating variables on emissions.

Tars and aqueous soluble organics present a problem with aspects not previously encountered. In some applications they can be used as a fuel

TABLE 18. Predicted Particulate Emission Levels

	Fixed Bed		Type of Gasifier			
	Updraft		Fixed Bed		Fluid/Entrained Bed	
	High	Low	High	Low	High	Low
Particulate Burden mg/m ³	1,000	100	6,000	100	100,000	10,000
Particle Size Distribution ^(a) microns	>10 1-10 <1	3% 8% 89%	all >10	>10 3% 1-10 8% <1 89%	>10 1-10 <1	61% 38% 1%
Gas Cleaning Technology						
Single Cyclone	NA	NA	240	8	11,500	1,100
Series Cyclone	NA	NA	10	6	2,300	230
Cyclone & Filter	NA	NA	3.2	.4	39	4
Cyclone & ESP ^(b)	NA	NA	3.2	.8	77	8
Cyclone & Wet Scrubber	NA	NA	3.2	1.6	150	15
ESP ^(b)	NA	NA	60	10	2,060	206
Wet Scrubber	450	45	60	20	NA	NA
Wet Scrubber & Filter	18	2	.6	1	NA	NA
Wet Scrubber & ESP ^(b)	45	5	.06	1	NA	NA

a) Same particle size distribution was used for both the high and low case

b) ESP - electrostatic precipitator

c) For downdraft large particle size distribution was used for high loading and small particle size distribution was used for low loading

TABLE 19. Applicable Gas Cleaning Options

	CLOSE-COUPLED BOILER	DIESEL OR SPARK-IGNITION ENGINE	GAS TURBINE	SYN GAS OR SNG
Performance Standards	200-1500 mg/m ³	10-50 mg/m ³	1-80 mg/m ³	1-80 mg/m ³
Fixed Bed Updraft (2)	None (1)	WS + F	WS(1) WS + F WS + ESP ESP(1)	WS(1) WS + F WS + ESP ESP(1)
Fixed Bed Downdraft	None (1) C	C(1,4) C + F WS(1) WS + F	C(1) 2C(1) C + F C + ESP	C(1) 2C(1) C + F C + ESP
Fluidized Bed	2C(1,3) C + WS C + F ESP(1,6)	C + F C + WS(1) C + ESP(1,6)	C + WS(1) C + F C + ESP(6) (5)	C + WS(1) C + F C + ESP(6) (5)
Entrained Bed	Similar to Fluidized Bed			

LEGEND

C - Cyclone
 2C - Two Cyclones in series
 F - Fabric Filter (baghouse)
 WS - Wet Scrubber
 ESP - Electrostatic
 Precipitator

FOOTNOTES

- (1) Lower level of contaminants is acceptable - higher level would exceed limits.
- (2) Cyclone not effective due to smaller particle size distribution and tar droplets - use wet scrubber to remove tars first if any cleanup is required.
- (3) Assumes 50% of particulate is char and 90% burns in burner.
- (4) Cyclones are effective for this application but wet scrubbers are often used instead because gas cooling is also required. Also cyclone efficiency is affected by large turndown ratio required in some engine applications.
- (5) Pressurized operation may restrict the size or applicability of gas cleanup equipment (particularly baghouses and ESPs).
- (6) ESP is not as effective on particulates with high carbon content and may not be applicable.

directly (no removal required) such as in a boiler, but often they condense and cause plugging problems in transfer lines and other downstream equipment. Tars may be present as vapor or as an aerosol (small liquid droplets). There is no basic understanding of how variables such as temperature, tar loading, tar composition, and velocity affect tar deposition.

In addition there is no information on tar removal in the literature. We assumed the wet scrubbing is effective for biomass tar removal, but this has not been confirmed in large scale equipment. There are several disadvantages to wet scrubbing. The sensible heat of the gas is lost as is the heating value of water soluble compounds. Wet scrubbing generates a wastewater stream which may present environmental problems. Separation and recovery of biomass tars from wastewater has not yet been proven on a commercial scale.

4.2 EVALUATION BY END USE

The results presented in Table 19 indicate there are a variety of gas cleaning technologies that have the potential to meet contaminant removal requirements for biomass gasification. A qualitative discussion of these results by end use follows.

4.2.1 Close-Coupled Boiler or Kiln

In this application tars in the gas will burn if they do not condense out in the transfer line or burner head. Particulates must be reduced to a level that will meet emission standards from the boiler stack. This ranges from 100-1000 mg/m³ depending on the location and size of the plant.

Gas from a fixed bed updraft gasifier can, in most cases, be used directly without cleanup. This is done commercially; however, in some applications a liquid knockout pot to collect condensed tars has been installed between the gasifier and the burner to prevent plugging (Miller 1983).

It is marginal whether gas from a downdraft gasifier can meet particulate emission standards. Use of a single cyclone would assure that the standards could be met.

With fluid-bed gasifiers single cyclones may be adequate for particulate removal in some applications. However, our data on fluid bed gasifier emissions indicate that in most cases two cyclones in series or a cyclone followed by a wet scrubber or a baghouse filter will be required. Two commercial fluidized bed biomass gasifiers use two or more cyclones in series (Murphy 1984; Miller 1983). An electrostatic precipitator (ESP) might also meet particulate removal standards. However, there is no data on effectiveness of ESP with high carbon particulates such as would be in fluid gasifier gas. The effect of tars on fabric filters and ESP is not well understood.

4.2.2 Diesel and Spark-Ignition Engines

These machines are very susceptible to erosion and plugging from particulates and some form of particulate removal will be required for all types of gasifiers. Limits on particulates and tar for this application are in the range of 10-50 mg/m³. In addition, the gas is usually cooled to provide more horsepower (less derating).

With a fixed-bed gasifier tar removal will be required so wet scrubbing is the most logical choice. An additional high efficiency particulate removal step such as a fabric filter might also be required.

Downdraft gasifiers may meet tar limits for diesel and spark-ignition engines but particulate removal will be required. A cyclone or a cyclone/fabric filter combination have been used with some success on many vehicular gasifiers (Kaupp and Goss 1981). For stationary engine applications wet scrubbing has been used (Self Sustained Dual-fuel Power System 1980; KHD Combines Engine and Gasifier 1984).

Because of the high particulate burden and the fact that sensible heat of the gas is lost in engine applications, fluid bed gasifiers are less than ideal for this application. It does appear from the tables that a combination of cyclone and a fabric filter or a wet scrubber or an ESP could meet the particulate standards for engines. Tar removal will also be required so a cyclone/wet scrubber combination is most appropriate.

Despite the fact that much operating experience was obtained with diesel and spark-ignition engines using gas from fixed-bed gasifiers during the 1930's and 1940's there is still doubt about the long-term effects of particulates on engine life, particularly in utility applications (Kohan 1984). Long-term tests on a well-instrumented engine studying various gas cleanup options could lead to wider use of this technology.

4.2.3 Gas Turbines

The most serious problem in direct-fired (open-cycle) gas turbines due to gas stream contaminants is erosion or corrosion of turbine blades by solid particulates. Particulate limits range from 1-80 mg/m³ depending on the type of turbine. Tar will be combusted in the turbine if the unit is close coupled. Tar removal may be required to prevent plugging in long transfer lines.

Particulate removal will be required even with a fixed-bed updraft gasifier. Cyclones cannot be used because of the large loading of tar droplets so wet scrubbing will be required. This reduces the efficiency of a fixed-bed updraft gasifier for this application and generates a wastewater stream. As a result, fixed-bed updraft gasifiers are less than ideal for gas turbine applications.

With a downdraft gasifier, a cyclone or two cyclones in series could meet particulate removal requirements in some cases. However, if the particulate

levels required for gas turbines are indeed on the low end of the range (1-10 mg/m³) a cyclone plus a filter or ESP will be required to meet these levels.

The high particulate burden of fluidized bed gasifiers makes it difficult to reach the very low levels of 1-10 mg/m³. A cyclone plus a filter, or wet scrubber or ESP can reduce particulates to 10-50 mg/m³ which may be acceptable.

With indirect fired (closed-cycle) turbines gas cleanup requirements would be similar to a close-coupled boiler (see Section 2.1).

4.2.4 Synthesis Gas or SNG

For synthesis gas applications (methanol, methane, hydrocarbons, ammonia, etc.) particulate removal and most likely tar removal will be required. Particulate limits are similar to gas turbine applications, 1-80 mg/m³. Tar limits are not known but thought to be small, <100 mg/m³ (see Section 2.4).

With a fixed-bed gasifier a wet scrubber may be sufficient for synthesis gas applications. Depending on the exact particulate burden in the gas and the removal requirements an addition removal step such as a fabric filter or ESP may be necessary.

Gas cleanup requirements for a downdraft gasifier used for synthesis gas production will be the same as discussed for the gas turbine (see Section 2.3) unless tar removal is necessary. If tar removal is necessary a cyclone plus wet scrubber could be used. At low loadings a wet scrubber alone might meet the particulate limits for synthesis gas applications.

A fluid-bed gasifier will require both particulate and tar removal so a cyclone/wet scrubber combination is probably the best choice. It is marginal as to whether this combination can meet the particulate limits. A fabric filter (baghouse) may be required in addition.

4.3 RECOMMENDATIONS

Tars and particulates have been identified as the two most significant contaminants in fuel gas from biomass gasifiers. Recommendations for additional research and development on particulate and tar removal have been identified.

4.3.1 Particulate Removal

As indicated in Section 3 particulate removal technology appears adequate to deal with biomass gas, especially when simultaneous tar removal is not required. However, more detailed data on particulate loadings, particle size and particle composition from different type gasifiers and different types of feedstocks and more definite limits for various end uses is needed for engineering design of large-scale commercial gas cleanup systems.

There is a fair range of particulate limits given for engine and turbine applications and this leads to some uncertainty in design of gas cleanup systems. In addition there is concern about the long-term (10-20 years) effects of particulates on engine and turbine life. Synthesis gas applications are not far enough advanced for this to have become a critical item, but there will undoubtedly be similar concerns over the gas compressors in the synthesis plants. Long-term tests on a well-instrumented engine or turbine could resolve many of these questions and lead to wider use of biomass derived fuel gas in these applications. The test work done by Aerospace Research Corporation on wood-fired gas turbines may provide data that is also applicable to turbines, fueled by gas from biomass gasifiers (Hamrick 1984).

The U.S. Department of Energy, Office of Fossil Energy is developing high-temperature, high-pressure gas stream cleanup systems for coal fired advanced power-generating systems. The results of this effort should be monitored closely for their applicability to biomass systems.

4.3.2 Tar Removal

Removal of tar from gas streams presents a problem with aspects not previously encountered. Tars from fixed-bed biomass gasifiers are most likely present primarily as very small liquid droplets although no information on particle size was found in the literature.

A study of the mechanism of tar formation including the effect of gas velocity, temperature, and wood moisture content on tar loadings and liquid particle diameters would aid significantly not only in gas stream cleanup design but in gasifier design. In conjunction with this, research on the efficiency of various wet scrubbing techniques for tar removal is needed for design of wet scrubbing systems.

Tars from downdraft and fluid bed gasifiers are probably present primarily as vapors although this has not been confirmed. Tar removal will require cooling and water scrubbing. The size of droplets formed by quenching and the effect of the method of quenching on the efficiency of tar removal needs to be studied.

Water scrubbing produces a wastewater stream containing tars and water soluble organics which may present an environmental problem. Research on separation of tars from the aqueous layer and methods of treatment and disposal of the water is needed. Thermal or catalytic cracking of tars to convert them to gas and char should be considered as an alternative to water scrubbing. This has been investigated for both biomass gas (Baker and Mudge 1984) and coal gas streams (Baker and Mudge 1985).

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APPENDIX A

Nomograph for Conversion
of Particulate Loadings

Particulate Loading Nomograph

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